BNL-62015

GLOBAL EMISSIONS INVENTORIES OF ACID-RELATED COMPOUNDS

T.E. GRAEDEL[1], C.M. BENKOVITZ[2], W.C. KEENE[3], D.S. LEE[4], and G. MARLAND[5]

[1]*AT&T Bell Laboratories, Murray Hill, NJ 07974-0636,* [2]*Department of Applied Science, Brookhaven National Laboratory, Upton, NY 11973,* [3]*Department of Environmental Science, University of Virginia, Charlottesville, VA 22903,* [4]*AEA Technology, 364 Harwell, Didcot, Oxfordshire OX11 ORA, United Kingdom,* [5]*Environmental Science Division, Oak Ridge National Laboratory, P.O. Box 2008, Oak Ridge, Tennessee 37831*

Abstract. Computer assessments of the atmospheric chemistry and air quality of the past, present, and future rely in part on inventories of emissions constructed on appropriate spatial and temporal scales and with appropriate chemical species. Accurate inventories are also of substantial utility to field measurement scientists and the regulatory and policy communities. The production of global emissions inventories is the task of the Global Emissions Inventory Activity (GEIA) of the International Global Atmospheric Chemistry Project (IGAC). This paper presents a summary of recent emissions inventories from GEIA and other programs for reference year 1985, with special attention directed to emissions of the acid-related compounds CO_2 ($\approx$6.2 Pg C yr^{-1} anthropogenic), SO_x ($\approx$65 Tg S yr^{-1} anthropogenic and 15 Tg S yr^{-1} natural), NO_x ($\approx$21 Tg N yr^{-1} anthropogenic and 15–20 Tg N yr^{-1} natural), HCl ($\approx$55 Tg Cl yr^{-1} total), and NH_3 ($\approx$45 Tg N yr^{-1} total). The global acid-equivalent flux of about 4.2 Teq H^+ yr^{-1} is about equally attributable to SO_x and NO_x emissions. For some of the acid-related species, historic inventories are available for a century or more; all show dramatic emissions increases over that period. IPCC scenario IS92a is used here as the basis for constructing global acid-related emissions estimates for selected years to 2100; among the results are that acid equivalent emissions are expected to more than double in the coming century.

1. The GEIA Project

Emissions inventories are the basis for studies of the interrelationships of atmosphere and surfaces, and of the interactions of societal development and environmental impact. In certain regions of the world, particularly the more heavily industrialized countries of Western Europe and North America, detailed emissions inventories have been available for a decade or more, though they undergo constant revision and improvement. For other regions, emissions inventories are often unreliable or nonexistent. In all cases, differences in approach make it difficult to compare local or regional inventories with one another and to merge such inventories to create a larger whole. Yet, it is these merged inventories that are needed by modelers working on continental or global scales, or in regions for which locally-generated emissions inventories are not available. Further, continental or global modelers have often been required to prepare their own inventories and have done so in very different ways, since they are not emissions specialists; as a consequence, comparing model results is confounded by the use of widely-varying emissions scenarios.

Are suitable, reliable emissions inventories generally available, even if many are unaware of them? A survey of then-existing emissions inventories performed in 1992 (Graedel et al., 1993) indicated that only the extant CFC inventory was regarded as good; however, that inventory was not available in gridded form. Inventories for CO_2, CH_4, NO_x, SO_x, reduced sulfur, and radon were regarded as fair, other inventories as poor or

The AT&T Corp.'s right to a non-exclusive, royalty free licence in and to any copyright is acknowledged.

Water, Air and Soil Pollution **85**: 25–36, 1995.

nonexistent. The temporal resolution was considered almost uniformly poor. That survey made it clear that the generation of internally consistent, rigorously developed, gridded inventories with good spatial and temporal resolution would be of considerable value.

These needs were the inspiration for the formation of the Global Emissions Inventory Activity (GEIA) under the auspices of the International Global Atmospheric Chemistry Project. The GEIA project has as its goals the establishment of framework for the development and evaluation of global emissions inventories, and the generation and publication of inventories for use by scientists and policy makers worldwide. Table I presents details about the active GEIA projects, together with information on the anticipated release dates of the resulting inventories. GEIA inventories are available electronically through anonymous file transfer protocol (FTP). The resident source is the Data Center at the National Center for Atmospheric Research, whose email address for GEIA data and documents is ncardata.ucar.edu (128.117.8.111). The FTP file contains documentation on line (a README file) for using the data system. Users without FTP access may order data on tape or diskettes from the GEIA Data Management Center at 1-303-442-6866 or by electronic mail (the preferred technique): paulette@rmii.com or hopkins@rmii.com

In this paper, we draw upon GEIA inventories that have been completed or are in progress to present a perspective on the emissions of acid-related gases to the atmosphere. The list includes SO_x and NO_x (precursors to the strong mineral acids sulfuric and nitric), the ubiquitous, weakly-acidic CO_2, HCl (often emitted directly in acidic form), and NH_3 (the only common gaseous atmospheric alkaline species).

2. Emissions of Carbon Dioxide

Of the acid-related species, the fluxes of carbon dioxide are far and away the largest; they are dominated by balanced natural flows involving vegetative photosynthesis and respiration and oceanic cycling, total sources and sinks each being of order 210 Pg C yr^{-1} (Moore and Braswell, 1994). Average volcanic CO_2 emissions, at some 18 Tg C yr^{-1} (Williams et al., 1992) are of little importance. The budget is, however, unbalanced by anthropogenic activities, expecially deforestation and the combustion of fossil fuels.

The combustion of fossil fuels and the concomitant production of CO_2 have been the subject of intensive study, as have the minor sources cement production and gas flaring. The procedure is to use United Nations energy statistics for fuel use and to multiply by the carbon content of the fuel burned and a factor reflecting the degree to which, on average, fuel-contained carbon is liberated as CO_2. Country statistics are adjusted for imports and exports, for net changes in storage, and for the production of secondary products that will not be oxidized. Population density is used to distribute emissions within each country. When all such totals are summed, the fossil fuel CO_2 source for 1991 is estimated at 6.2 Pg C yr^{-1} (Marland et al., 1994a). Less well determined is the net CO_2 flux related to land clearing and biomass burning, but in a typical recent year the estimated range is 0.4 – 2.6 Pg C yr^{-1} (Tans et al., 1990), with a preferred value of about 1.7 Pg C yr^{-1} (Enting et al., 1994). Fossil fuel-related CO_2 emissions have recently been gridded to 1° by 1° by Andres et al. (1995). The result for reference year 1990 is shown in Figure 1.

Table I. GEIA Active Emissions Inventory Projects

Project[%]	Director	Spatial Resolution	Temporal Resolution	Ref. Year	Est. Date of Release
VOC(N)[#]	Hewitt, UK Guenther, USA	1° × 1°	Monthly	1990	Available
Aircraft[†]	Wuebbles, USA	1° × 1°	Annual	1990	Available
CO_2(A)[@]	Marland, USA	1° × 1°	Annual	1987	Available
NO_x(A)	Benkovitz, USA Pacyna, NOR	1° × 1°	Annual	1985	Available
SO_x(A)	Benkovitz, USA Pacyna, NOR	1° × 1°	Annual	1985	Available
NO_x(A)[‡]	Scholtz, CAN	1° × 1°	Seasonal	1985	Available
SO_x(A)[‡]	Scholtz, CAN	1° × 1°	Seasonal	1985	Available
CFC-11(A)	Cunnold, USA	1° × 1°	Annual	1986	Available
CFC-12(A)	Cunnold, USA	1° × 1°	Annual	1986	Available
CFC-113(A)	Cunnold, USA	1° × 1°	Annual	1986	Available
N_2O(A+N)	Bouwman, NL	1° × 1°	Annual	1985	Available
NO_x(N)	Levy, US	1° × 1°	Monthly	1985	Available
Bl. C(A+N)	Dignon, USA	1° × 1°	Annual	1985	Available
VOC(A)[‡]	Berdowski, NL Benkovitz, USA	1° × 1°	Annual	1985	Oct 95
NH_3(A)	Bouwman, NL Lee, UK	1° × 1°	Annual	1985	Oct 95
Pb(A)	Pacyna, NOR	1° × 1°	Annual	1985	Dec 95
Biomass burning (A+N)	Goldammer, GER Levine, USA Stocks, CAN	1° × 1°	Annual	1987	Dec 96
Rct. Cl(A+N)	Keene, USA	1° × 1°	Seasonal	1990	Jun 97
Red. S(N)	Tarrason, NOR	1° × 1°	Annual	--	Dec 97
CH_4(A)	Roulet, CAN Matthews, USA	1° × 1°	Annual	1985	Dec 97
Hg(A)	Pacyna, NOR	1° × 1°	Annual	1985	Dec 97
CO(A)	Logan, USA	1° × 1°	Annual	1985	Dec 97
Radon(N)	Kritz, USA	1° × 1°	Annual	--	Jun 98
Data Mgt.	Middleton, USA	--	--	--	--

[%] A = anthropogenic; N = natural.

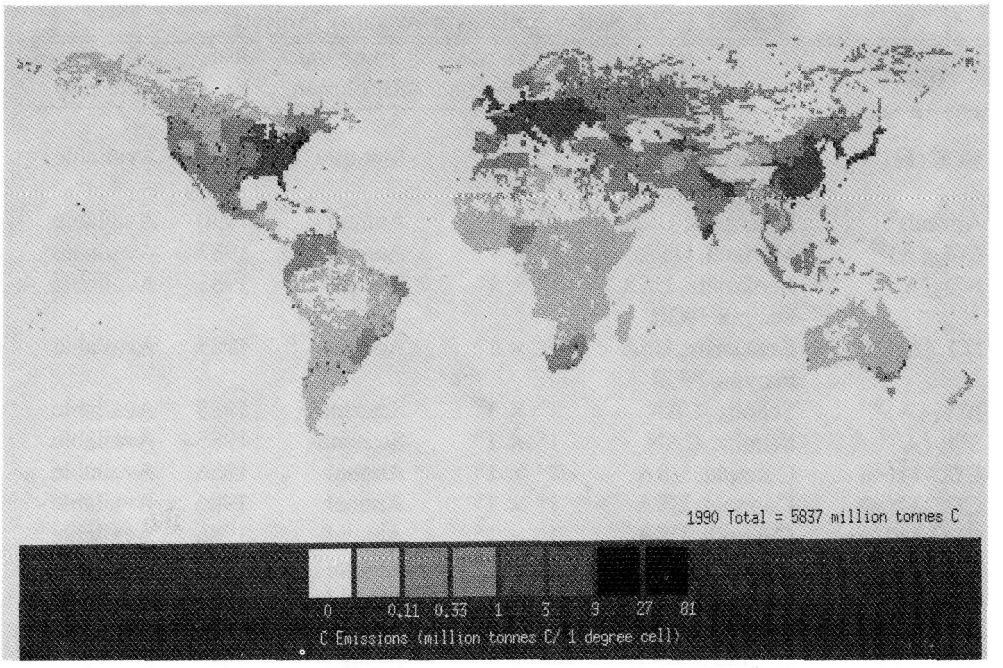
[#] This inventory, described in Guenther et al. (1995), is speciated into isoprene, terpenes, short-lived VOC, and long-lived VOC.

[‡] Emissions are resolved in the vertical into fluxes above 100 m and those below 100 m.

[‡] Will be speciated; divisions yet to be determined.

[†] This inventory, described in Baughcum et al. (1993), is available through the courtesy of the NASA Atmospheric Effects of Stratospheric Aircraft Program. Emissions are resolved in the vertical into eighteen 1 km levels.

[@] Inventories are available for 1950, 1960, 1970, 1980, and 1990.



(Andres et al., 1995).

3. Emissions of Sulfur Oxides

Sulfur dioxide is emitted naturally from volcanoes, and produced in nature by the oxidation of the oceanically-generated gas dimethyl sulfide. The volcano emission rate is variable, of course, but averages 9 to 10 Tg S yr^{-1} (Stoiber et al., 1987, Bates et al., 1992). The dimethyl sulfide source is about 15 Tg S yr^{-1} (Langner and Rodhe, 1991, Bates et al., 1992). Although the mechanisms for the oxidation of DMS in the atmosphere are not well established, current models estimate that the majority of the DMS is converted to SO_2 (Benkovitz et al., 1994). These natural sources in total are smaller than sulfur emissions related to human activities.

Anthropogenic emissions of sulfur oxides (SO_2 plus sulfate) are almost entirely the result of the combustion of sulfur-containing fossil fuels and the smelting of metal sulfide ores. Major emitting sectors include coal combustion, especially in large electric and heat power plants, and combustion of residential fuel oil. In the past three decades more than twenty inventories of emissions have been formulated, with total fluxes ranging from 46–104 Tg S yr^{-1} (Benkovitz et al., 1995). The approach has generally been to use fossil fuel combustion as the principal source of sulfur dioxide emissions, and to compute the emissions from energy statistics. More recent efforts have produced gridded inventories, which have required that population or energy consumption or both be used to allocate emissions to specific geographic areas.

The most complete and encompassing inventory of anthropogenic emissions of SO_x is one just completed by the GEIA SO_x project group using a sequential approach. First, the global inventory is gridded to 1° by 1° on the basis of economic data for sulfur-emitting activities, emission factors, and information on sulfur recovery. Next, in geographical areas where detailed inventories have been performed (South Africa, North America, etc.), the basic inventory is replaced by the detailed, locally-generated data. In some cases (Japan and Western Europe, for example), regional inventories were updated by new GEIA assessments. Finally, major stationary sources such as power plants and smelters were added in for regions such as the former Soviet Union. The overall result is an emissions estimate of about 65 Tg S yr^{-1}. The details of this process and of the inventory itself are described by Benkovitz et al. (1995) and the resulting gridded inventory shown in Figure 2.

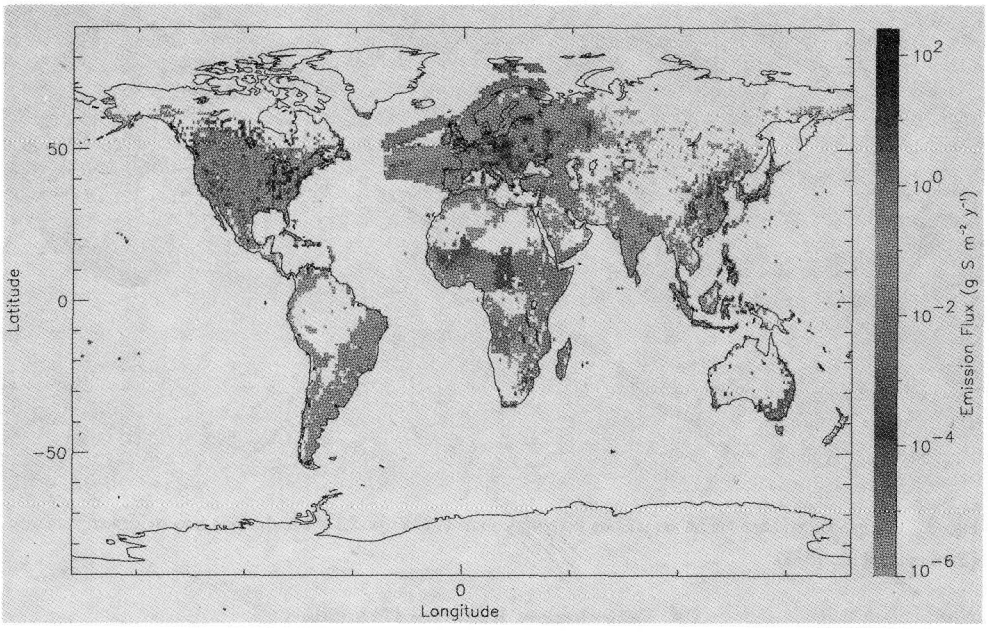


Fig. 2. The global gridded GEIA emissions inventory map (1985 epoch) for anthropogenic oxides of sulfur (Benkovitz et al., 1995).

4. Emissions of Oxides of Nitrogen

Unlike the emissions of sulfur dioxide, for which anthropogenic sources clearly dominate, natural sources of oxides of nitrogen may be roughly comparable with anthropogenic sources on a global basis. Soil-biogenic emissions are of order 5 Tg N yr^{-1}, and have been gridded by Yienger and Levy (1995). Biomass burning emissions are of order 9 Tg N yr^{-1} (Andreae, 1991) and lightning emissions, though poorly quantified, are thought to be less than 10 Tg N yr^{-1} (Yienger and Levy, 1995). Thus, the natural total is perhaps 15–20 Tg N yr^{-1}.

Anthropogenic emissions of oxides of nitrogen are almost totally related to the combustion of fossil fuels. As with SO_x, the most comprehensive gridded inventory for

ground-based sources is that recently completed by the GEIA NO_x working group. The inventory was constructed in parallel with that for SO_x, but used a different base inventory and somewhat different supplementary information, in part because the majority of the NO_x emissions are released by the transportation sector. The overall result for all NO_x sources is an emissions estimate of about 21 Tg N yr^{-1}. The details of this process and of the inventory itself are described by Benkovitz et al. (1995) and the resulting gridded inventory shown in Figure 3. Additional anthropogenic NO_x emissions are those from aircraft (Baughcum et al., 1993); these currently constitute about 1.5 Tg N yr^{-1}.

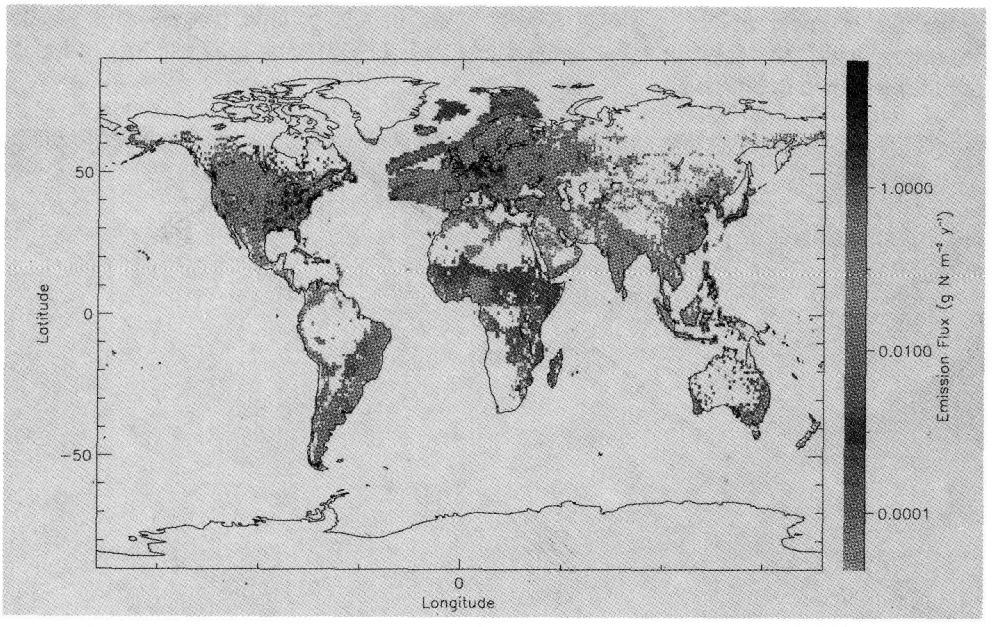


Fig. 3. The global gridded GEIA emissions inventory map (1985 epoch) for anthropogenic oxides of nitrogen (Benkovitz et al., 1995).

5. Emissions of Reactive Chlorine

There are many known sources for atmospheric HCl, but the fluxes from each are quite poorly determined. The dominant flux is generally believed to be HCl production in the marine atmosphere by direct volatilization from deliquescent seasalt aerosol that has been acidified by the incorporation of HNO_3 and/or H_2SO_4. Other sources include biomass burning (Andreae, 1995), volcanic eruptions (Symonds et al., 1988), coal combustion, and waste incineration. An uncertain but potentially large source is the photolysis of Cl-atom precursors such as HOCl and Cl_2, followed by reaction with hydrocarbons.

The current state of understanding of the tropospheric HCl budget is discussed by Graedel and Keene (1995). Their conclusion is that in a year with average volcanic activity we expect the total global injection of HCl into the atmosphere to be of order 55 Tg Cl yr^{-1}, much of it at continental margins where sea salt generation and fossil fuel emissions are both substantial. No gridding of these emissions has yet been accomplished.

6. Emissions of Ammonia

Ammonia, the atmosphere's only common alkaline gas, has a number of sources: the bacterial decomposition of animal excreta, soils, vegetation, biomass burning and the oceans. Several estimates of global fluxes have been made including those of Soderlund and Svensson (1977), Bottger et al. (1978) and Schlesinger and Hartley (1992). The most recent inventory to be published is that of Dentener and Crutzen (1994), which is the only one disaggregated on a global grid (10° by 10°). The total emissions of NH_3 N of Dentener and Crutzen's inventory amount to some 45 Tg N yr^{-1}, which is slightly lower than the lowest end of the range given by Schlesinger and Hartley, i.e., 50–128 Tg N yr^{-1}. Schlesinger and Hartley's 'best' estimate of 75 Tg N yr^{-1} is likely to be an overestimate as the dominant source, domestic animals, was calculated using emission factors developed in Europe. These factors are likely to be inapplicable to large areas of the world, specifically in less developed nations where the N content of feedstuff is much lower. The emissions calculated by Dentener and Crutzen were broken down to 25 Tg N yr^{-1} (animals, domestic and wild), 7 Tg N yr^{-1} (oceans), 6 Tg N yr^{-1} (fertilizer), 5 Tg N yr^{-1} (vegetation), and 2 Tg N yr^{-1} (biomass burning). All these estimates are likely to be highly uncertain as emission factors for these sources are not well developed. The spatial distribution of these emissions show that the largest source areas are those with the largest concentrations of domestic animals, e.g., India and northern Europe. Various source data and other information are being used in a GEIA subproject group to produce a 1° ×1° inventory of ammonia emissions which is expected to be released in 1996 (Lee et al., 1995).

7. Total Acid Gas Emissions

The effects of acid-related gases on soils, vegetation, statuary, and so forth depend not only on the individual species but also on the total flux of acid delivered to the receptors. In an effects context, CO_2 is of quite minor interest despite its high rate of emissions, since its solubility and its acid dissociation constants are low and its contributed acidity is thus seldom important. The same is, of course, not true of the strong mineral acids H_2SO_4, HNO_3, and HCl, which can be expected to eventually reach the surface, fully ionize, and thus contribute to received acidity. For perspective, therefore, we can convert the emission fluxes of the mineral acids or their precursors into acid equivalent fluxes, using conversion factors of 0.4 for SO_x to sulfate (Charlson et al., 1992) and 0.8 for NO_x to nitrate (L. Kleinman, Private communication, 1995). The results are 2.0 Teq H^+ yr^{-1} for SO_x (83% anthropogenic), 2.2 Teq H^+ yr^{-1} for NO_x (57% anthropogenic), and 1.6 Teq H^+ yr^{-1} for HCl (apparently mostly from acid-displacement reactions involving anthropogenically derived precursors). HCl generated by acid-displacement does not correspond to a net production of atmospheric acidity and, thus, its net influence is already included implicitly in the sum of SO_x and NO_x (4.2 Teq H^+ yr^{-1}). This total should be reduced by 3.2 Teq H^+ yr^{-1} to account for neutralization by the alkalinity-related flux of NH_3, and by 0.2–0.8 Teq H^+ yr^{-1} (Chameides and Stelson, 1992) to take sea salt alkalinity into account. Thus, the global total is of order 0.5±0.3 Teq H^+ yr^{-1}. Further neutralization doubtless takes place as a consequence of continental dust, but the global dust flux is too uncertain to include here, and, of course, geographical variations in emitted and deposited fluxes of all these species are very large. Thus, on a global basis, the atmosphere is roughly neutral, but neutrality is unusual at smaller scales: different

geographical regions tend to be predominantly acidic or predominantly alkaline.

8. Historical Acid-Related Emissions and Emissions Trends

It is often useful for science and policy purposes to have emissions inventories for periods in the past, and some historical inventories have been completed over the past decade or so. The most detailed and reliable are those for anthropogenic CO_2, which have been compiled on an global annual basis for the period 1751-1991 (Marland et al., 1994b). Over that period, the anthropogenic flux has increased from 2.6 Tg C yr^{-1} to 6.2 Pg C yr^{-1}, a factor of 2400! More recently, the inventories for 1950, 1960, 1970, and 1980 have been gridded to 1° by 1° by the GEIA CO_2 project (Andres et al., 1995).

In the case of sulfur oxides, two regional historical emissions inventories have been compiled: for Western Europe back to 1880 on a 150 km by 150 km grid (Mylona, 1993), and for the United States on a state-resolved basis back to 1900 (Gschwandtner et al., 1986). The inventories assume coal burning as the major source, and make estimates as to the sulfur content of the coals burned and the degree of emission control technology in place. For the European inventory, a peak rate of about 10 Tg S yr^{-1} occurred about 1940, followed by a post-war decrease and then a rapid increase to about 28 Tg S yr^{-1} in 1975. Since that time, emissions controls have produced about a 15% decrease in emissions. The United States sulfur dioxide emissions inventory has three peaks: 12 Tg S yr^{-1} around 1925, 14 Tg S yr^{-1} around 1944, and 17 Tg S yr^{-1} around 1970. As in Europe, SO_x emissions have since decreased by about 15%. A global inventory for 1970 to 1986 on a grid of 4.5° in latitude and 7.5° in longitude has been constructed by Hameed and Dignon (1992) using a similar approach. Those data show an 18% increase over the sixteen-year period, due largely to substantially increased emissions from Asia. Under development is an historical SO_x inventory for every ten years from 1860 to the present on a 5° × 5° global grid (Rodhe et al., 1995).

Historical emissions inventories for oxides of nitrogen are much less easy to develop, because the emission fluxes from fossil fuel combustion (the main source) are strong functions of temperature and thus of historic boiler and internal combustion engine technology. Notwithstanding these difficulties, a total United States inventory has been developed by Husar (1994) for the period 1900-1980, and a state-by-state inventory for the United States for 1900-1980 has been presented by Gschwandtner et al. (1986). Both studies arrive at 1980 totals of about 6 Tg N yr^{-1}, with an interim peak at 1930 resulting from extensive railroad and smelter activity and reasonably steady emissions increases since that time due largely to increased vehicle populations and larger numbers of gas-fired electrical power plants. On a percentage basis, the 1900 to 1980 increase was about 900%. A global NO_x emissions inventory for 1970 to 1986 on a grid of 4.5° in latitude and 7.5° in longitude has been constructed by Hameed and Dignon (1992); it indicates that emissions during that period increased by about 30%.

The tropospheric reactive chlorine budget is significantly influenced by anthropogenic sources and is definitely not in equilibrium. It is thus useful to examine what is known about the temporal concentration trends, especially in view of several observational studies showing that column HCl concentrations during the period 1977-1990 showed an upward trend of 1-11% per year (e.g., Rinsland et al., 1991). The most obvious candidate process as a source of increasing HCl is enhanced volatilization from seasalt, since, as

discussed above, emissions of SO_x and NO_x in more developed countries rose gradually from 1900-1940, rose rapidly from 1940-1975, and then decreased from 1975 onwards as a result of the installation of emissions control technology.

9. Projected Trends in Acid-Related Emissions

The development of emissions scenarios for the future is a contentious and rather arbitrary exercise, but is of great use to the scientific and policy communities so long as the inherent limitations are kept in mind. A well-known effort of this kind is that of the Intergovernmental Panel on Climate Change (Houghton et al., 1992), which has constructed emissions scenarios for greenhouse gases for several years between 1990 and 2100. The greenhouse gases include the acid-related species CO_2, SO_x, and NO_x, so it is of interest to use those projections as a basis for thinking about possible future acid-related emissions.

Houghton et al. (1992) give ungridded global emissions estimates for the years 1990, 2000, 2025, 2050, and 2100 for a "no climate policy" scenario (Scenario IS92a) that assumes certain levels of economic growth, certain types and quantities of energy supplies, and modest but increasing constraints on emissions of SO_x and NO_x. For CO_2, their 1990 anthropogenic emissions total is equal to that of the new GEIA inventory (Andres et al., 1995), so we adopt IPCC scenario IS92a as an initial model for the projection of emissions to future years. In the case of both SO_x and NO_x anthropogenic emissions, the new GEIA inventories give somewhat lower global totals , but are for reference year 1985. Accordingly, the IPCC values again seem reasonable. IPCC makes no estimates for HCl emissions, but if we assume a linear relationship between NO_x emissions and those of HCl (on the basis of mineral acid liberation of seasalt HCl), we can scale current emissions appropriately.

Given this scenario for future acid-related emissions, the values for SO_x, NO_x, and HCl can be converted to acid equivalents to provide some rough idea of the future prospects for acid-related influences on the planetary surface. The result, shown in Table II, is a doubling in the rates of emissions of acid equivalents between the present time and the year 2100. While today's acid equivalent totals due to SO_x and NO_x are roughly equivalent, the portion attributable to NO_x is predicted to increase in the future, as shown in Figure 4.

A more complete future scenario can be provided by considering the differing economic growth rates, energy use strategies, and emissions controls of regions and nations, and thus producing geographically-resolved scenarios for the emissions of acid-related species. An early effort along this line is that of the World Energy Council (1993), which has scenarios to year 2020 that estimate emissions of CO_2, S, and N for each of nine world regions. Many of the tools needed to further develop gridded emissions scenarios for all species of interest have been completed or soon will be, and IPCC is refining projections of relevant development parameters for the coming century. Hence, one can anticipate that within several years a full suite of gridded future emissions scenarios will be completed and ready for use by the scientific and policy communities.

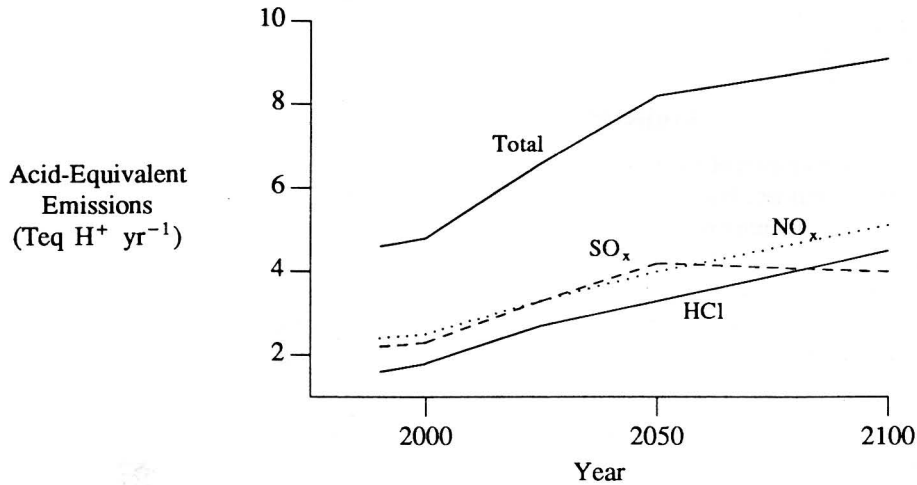


Fig. 4. Projected global emissions of acid equivalents, 1990-2100. The total is that for anthropogenic and natural SO_x and NO_x only, since emissions of HCl are thought to be primarily by acid displacement rather than additive. NH_3 future emissions are not projected in this work, but can be expected to neutralize a portion of the total acidity.

Table II. Future Global Scenarios for Anthropogenic Emissions of Acid-Related Gases and Acid Equivalents*

Emittant	1990	2000	2025	2050	2100
Emissions Fluxes:					
CO_2 (Pg C/yr)	6.2	7.2	11.1	13.7	20.4
SO_x (Tg S/yr)	73	77	117	151	144
NO_x (Tg N/yr)	25	28	43	53	72
HCl (Tg Cl/yr)	55	62	95	117	158
Acid Equivalents:					
SO_x (Teq/yr)	1.8	1.9	2.9	3.8	3.6
SO_x natural (Teq/yr)	0.4	0.4	0.4	0.4	0.4
NO_x (Teq/yr)	1.4	1.5	2.3	3.0	4.1
NO_x natural (Teq/yr)	1.0	1.0	1.0	1.0	1.0
Total (Teq/yr)	4.6	4.8	6.6	8.2	9.1

* Biomass burning emissions are not included in these figures.

10. Summary

Reliable, gridded emissions inventories are now available, or soon will be, for all of the principal acid-related gases. Those inventories will, of course, be improved as more detailed local information becomes available, especially for emissions from the less developed countries. Nonetheless, there is no scientific reason to avoid standardizing on the inventories now available for use in model studies.

Historical estimates of emissions are much less reliable than those for the present epoch, but historical inventories based on unified global statistics and methodologies are beginning to appear. They will be particularly useful in model studies linked to historical data. Much effort remains to be done in this regard; in particular, a gridded, global historical inventory of NO_x emissions would be a valuable project, and could be accomplished. Scenarios for future emissions are also beginning to be attempted, and will evolve rapidly during the next few years.

Emissions inventories are the base on which many scientific studies rest. Their quality and their availability have improved markedly during the past few years, especially so far as global, gridded inventories are concerned. Nonetheless, emissions inventories are, strictly speaking, never correct and never completed. In a world in which population, economic activity, and technology are constantly evolving, the construction and maintenance of reliable emissions inventories remains an effort in need of continuous financial support and the dedicated participation of outstanding scientists and engineers.

Acknowledgements

This paper is largely based on the work of the GEIA inventory project teams. We are grateful for the contributions of all the participants in these projects; there are too many such to name here. The overall effort is guided by the GEIA Steering Committee, which consists of A.F. Bouwman, D.M. Cunnold, T.E. Graedel, P.B. Middleton, and J.M. Pacyna. The activities of GEIA have been greatly assisted by in-kind released time contributions by the institutions of the GEIA participants, and by the financial support of Chemical Manufacturers Association (USA), Dutch Ministry of Housing, Physical Planning, and Environment (NL), National Aeronautics and Space Administration (USA), National Center for Atmospheric Research (USA), National Institute of Public Health and Environmental Protection (NL), National Oceanic and Atmospheric Administration (USA), National Science Foundation (USA), Norwegian Institute of Air Research (NOR), TNO Institute of Environmental and Energy Technology (NL), and World Meteorological Organization (SUI), to help facilitate data management activities, workshops, and travel expenses. The work of D.S.L. is supported by the Hadley Centre, UK. This research is part of the International Global Atmospheric Chemistry (IGAC) Core Project of the International Geosphere-Biosphere Program (IGBP).

References

Andreae, M.O.: 1991, in *Global Biomass Burning: Atmospheric, Climatic, and Biospheric Implications*, Levine, J.S., Ed., pp. 3-21, MIT Press, Cambridge, Mass.

Andreae, M.O.: 1995, Paper presented at AGU Chapman Conference on Biomass Burning and Global Change, Williamsburg, VA.

Andres, R.J., Marland, G., Fung, I., and Matthews, E.: 1995, *Global Biogeochem. Cycles*, submitted.

Baughcum, S.L., Metwally, M., Seals, R.K., and Wuebbles, D.J.: 1993, in *The Atmospheric Effects of Stratospheric Aircraft: A Third Program Report*, edited by R.S. Stolarski and H.L. Wesoky, pp. 185-208, NASA Ref. Pub. 1313, Nat. Aeronautics and Space Admin., Washington, D.C.

Bates, T.S., Lamb, B.K., Guenther, A., Dignon, J., and Stoiber, R.E.: 1992: *J. Atmos. Chem.*, **14**, 315-337.

Benkovitz, C.M., Berkowitz, C.M., Easter, R.C., Nemesure, S., Wagener, R., and Schwartz, S.E.: 1994: *J. Geophys. Res.*, **99**, 20725-20756.

Benkovitz, C.M., Scholtz, M.T., Pacyna, J., Tarrason, L., Dignon, J., Voldner, E.C., Spiro, P.A., Logan, J.A., and Graedel, T.E.: 1995, *J. Geophys. Res.*, submitted.

Bottger, A., Ehhalt, D.H. and Gravenhorst, G.:1978, *Atmospheric Cycles of Nitrogen Oxides and Ammonia* (in German), Report 1558, KFA Julich, Germany.

Chameides, W.L., and Stelson, A.W.: 1992, *J. Geophys. Res.*, **97**, 20565-20580.

Charlson, R.J., Schwartz, S.E., Hales, J.M., Cess, R.D., Coakley, J.A., Hansen, J.E., and Hofmann, D.J.: 1992, *Science*, **255**, 423-430.

Dentener, F., and Crutzen, P.J.: 1994, *J. Atmos. Chem.*, **19**, 331-369.

Enting, I.G., Wigley, T.M.L., and Heimann, M.: 1994, *Future Emissions and Concentrations of Carbon Dioxide: Key Ocean/Atmosphere/Land Analyses*, Tech. Pap. 31, CSIRO Div. of Atmos. Res., Mordialloc, Vic., Australia.

Graedel, T.E., and Keene, W.C.: 1995, *Global Biogeochem. Cycles*, **9**, 47-77.

Graedel, T.E., Bates, T.S., Bouwman, A.F., et al.: 1993, *Global Biogeochem. Cycles*, **7**, 1-26.

Gschwandtner, G., Gschwandtner, K., Eldridge, K., Mann, C., and Mobley, D.: 1986, *J. Air Pollut. Contr. Assoc.*, **36**, 139-149.

Guenther, A., Hewitt, C.N., Erickson, D., et al., 1995: *J. Geophys. Res.*, **100**, 8873-8892.

Hameed, S., and Dignon, J.: 1992, *J. Air Waste Manage. Assoc.*, **42**, 159-163.

Houghton, J., Callander, B., and Varney, S.: 1992, *Climate Change 1992: The IPCC Supplementary Report*, Cambridge, U.K.: Cambridge University Press.

Husar, R.B.: 1994, in *Industrial Metabolism: Restructuring for Sustainable Development*, Ayres, R.U., and Simonis, U.E., Eds., Tokyo: United Nations University Press.

Langner, J., and Rodhe, H.: 1991, *J. Atmos. Chem.*, **13**, 225-265.

Lee, D., Bouwman, A.F., Dentener, F., et al.: 1995, in preparation.

Marland, G., Andres, R.J., and Boden, T.A.: 1994a, in *Trends '93: A Compendium of Data on Global Change*, Boden, T.A., Kaiser, D.P., Sepanski, R.J., and Stoss, F.W., Eds., pp. 505-584, Rpt. ORNL/CDIAC-65, Oak Ridge National Lab., Oak Ridge, TN.

Marland, G., Andres, R.J., and Boden, T.: 1994b, in *Global Climate Change: Science, Policy and Mitigation Strategies*, C.V. Mathai and G. Stensland, Eds., Proc. Intl. Spec. Conf., Air and Waste Mgt. Assoc., Pittsburgh, PA.

Moore, B. III, and Braswell, B.H., Jr.: 1994, *Ambio*, **23**, 4-12.

Mylona, S.: 1993, *Trends of Sulphur Dioxide Emissions, Air Concentrations and Depositions of Sulphur in Europe Since 1880*, EMEP/MSC-W Rpt. 2/93, Norwegian Meteorological Institute, Oslo.

Rinsland, C.P., Levine, J.S., Goldman, A., Sze, N.D., Ko, M.K.W., and Johnson, D.W.: 1991, *J. Geophys. Res.*, **96**, 15523-15540.

Rodhe, H., et al.: 1995, in preparation.

Schlesinger, W.H. and Hartley, A.E.: 1992, *Biogeochemistry*, **15**, 191-211.

Soderlund, R. and Svennson, B.H.: 1976, in *Nitrogen, Phosphorus and Sulfur Global Cycles*, SCOPE Report 7, Ecological Bulletin, Stockholm, Sweden.

Stoiber, R.E., Williams, S.N., and Huebert, B.: 1987, *J. Volcanol. Geotherm. Res.*, **33**, 1-8.

Symonds, R.B., Rose, W.I., and Reed, M.H.: 1988, *Nature*, **334**, 415-418.

Tans, P.P., Fung, I.Y., and Takahashi, T.: 1990, *Science*, **247**, 1431-1438.

Williams, S.N., Schaefer, S.J., Calvache V., M.L., and Lopez, D.: 1992, *Geochim. Cosmochim. Acta*, **56**, 1765-1770.

World Energy Council: 1993, *Energy for Tomorrow's World*, St. Martin's Press, New York.

Yienger, J.J., and Levy, H. II: 1995, *J. Geophys. Res.*, **100**, 11447-11464.