INTERNATIONAL GLOBAL ATMOSPHERIC CHEMISTRY

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## IGAC and Education

Much of the research in atmospheric chemistry conducted in universities and research centers has involved a large number of PhD students and post-doctoral fellows. With the growing attention given to global change issues over the last two decades, educational programs focusing on atmospheric chemistry and biogeochemistry have attracted a large number of outstanding students interested in highly visible "global change" issues and the importance to society of environmental problems such as ozone loss and climate change.

In recent years, however, the transformation of our economies and the importance of information technologies have created new challenges and new opportunities. Because many students have decided to invest their skills in these areas, university departments dealing with environmental issues have had increasing difficulties recruiting highly skilled students. This is apparently true in many countries around the world.

Time has perhaps come for our scientific communities to rethink their priorities, and for the governments and funding agencies to reassess their support of the sciences. An important goal in a highly competitive world will be to maintain a pool of highly qualified scientists and engineers in the research/academic environment. The atmospheric chemistry community needs to participate in this discussion, and to recognize that environmental issues and the methods to address them are evolving.

Environmental problems will remain important, precisely because the globalization of the economy will lead to new global environmental problems. But new concerns will require more integrated and more trans-disciplinary approaches. Key issues for atmospheric chemists lie at the borders between atmospheric chemistry and hydrology, meteorology, biology, oceanography, and socio-economics. Having scrutinized the world primarily from a disciplinary perspective, using global observing systems or global models, scientists in the next decades will probably attempt to apply fully integrated approaches to understand the evolution and the vulnerability of coherent regional areas.

Education must adapt to this evolution and develop the appropriate methodologies. Student training must evolve towards integrative thinking in which the complexity of the Earth system is recognized.

GLOBAL IGBP CHANGE

CACGP

# SCIENCE FEATURES

## Overview of the Global Emissions Inventory Activity (GEIA)

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### GEIA and IGAC

The Global Emissions Inventory Activity (GEIA) has provided scientifically sound, policy-relevant global emissions inventories to science and policy communities since the early 1990s. These inventories are used to assess past, present and future global atmospheric chemistry, air quality and climate change.

GEIA's work cuts across and contributes to all of IGAC's other activities. Its inventories provide a scientific foundation for policy initiatives designed to address urgent environmental issues such as global warming, stratospheric ozone depletion, acid precipitation and biological damage [Graedel *et al.*, 1993]. As these issues become more pressing, GEIA has entered a new phase of growth and responsiveness to an expanding user community.

GEIA includes several hundred scientists from around the world, many of whom are involved in other IGAC Activities. GEIA's work is guided by Co-Conveners Derek Cunnold of the Georgia Institute of Technology (USA) and Jos Olivier of RIVM, the Netherlands National Institute of Public Health and the Environment. Paulette Middleton, RAND (USA) directs the GEIA Center in Boulder, Colorado, the hub of data access and communication (http://weather.engin.umich.edu/geia/). Cunnold, Olivier and Middleton work closely with an international coordinating committee, including Carmen Benkovitz, Brookhaven National Laboratory (USA), Gregg Marland, Oak Ridge National Laboratory (USA), Ann McMillan, Atmospheric Environment Service (Canada), Jozef Pacyna, Norwegian Institute for Air Research and Trevor Scholtz, ORTECH Corporation (Canada).

### Guiding Principles

GEIA was created in 1990 to encourage the development of global emissions inventories of gases and aerosols emitted into the atmosphere from natural and anthropogenic sources. The long-term goal is to provide inventories of all trace species relevant to global atmospheric chemistry. As with all IGAC Activities, GEIA tries to include all interested parties on a volunteer basis. Emissions inventory experts from individual countries, supported by local organizations, conduct their own research while maintaining contact through GEIA's communications network and participation in periodic meetings. The GEIA forum allows participants to discuss their work and draw upon the combined expertise of their fellow members. This coordination ensures that results of their work are compatible and can be combined with a minimum of effort into a uniformly formatted data set.

Inventory development is carried out through individual projects. Currently, more than two dozen projects are addressing emissions for all major greenhouse gases and aerosols. Prior to becoming a GEIA database, the inventory and its documentation must undergo thorough peer review. Each GEIA database is accompanied by documentation and references to scientific publications that describe the effort.

GEIA's operations are organized around four principles:

1. GEIA's goal is to produce emissions inventories for all species of interest on a 1° x 1° global grid and a country-by-country basis.
2. GEIA inventories are accompanied by an assessment of their degree of uncertainty, as well as a comparison with results of top-down global or regional budget studies, if available.
3. GEIA seeks to provide comprehensive information for each database, including:
   - sectoral information
   - natural sources, if appropriate
   - temporal variation, at least by season/month
   - historical emissions, as a separate product
   - periodic updates, e.g., emission estimates for 1985 and 1995
4. GEIA project teams are international and intercontinental.

### Achievements and Plans

GEIA's achievements over the past decade include:

- a growing user community
- more than two dozen projects and many completed and available inventories
- the GEIA website
- ten international planning workshops
- links to related efforts

These achievements contribute to building a strong and solid foundation for the future of GEIA. The dedicated core teams, successful projects, establishment of a website, long-term planning enhanced by frequent international workshops, increasing attention to the user community, and forward-looking approaches to coordination and collaboration are all important elements of GEIA's future.

### Conveners and Participants

The first convener of GEIA was Thomas Graedel, now at Yale University, with Gregg Marland of Oak Ridge National Laboratory succeeding him in 1996, followed in

May 1998 by co-conveners Derek Cunnold of the Georgia Institute of Technology (USA), and Jos Olivier of the Netherlands National Institute of Public Health and the Environment (RIVM). Including the Coordinating Committee, groups responsible for development of the inventories and active users, there are now over 200 members in the GEIA e-mail network. Many project leaders and Coordinating Committee members have been part of GEIA since its inception in 1990.

## Inventory Development Activities

The current status of GEIA inventories is listed below. In the following list, (A) indicates emissions from anthropogenic sources and (N) emissions from natural sources.

**Compounds (available):** Ammonia (A&N), Black Carbon (A), Carbon Dioxide (A), Carbon Monoxide (A), Chlorofluorocarbons (A), Lead (A), Mercury (A), Methane (N), Sulfur and Nitrogen Oxides (A), Nitrous Oxide (A&N), Reactive Chlorine Emissions (A&N),Volatile Organic Compounds (A&N).

**Compounds (in progress):** Methane (A) (preliminary 1990 GEIA inventory available at EDGAR website), Organochlorines (A), Radionuclides (N), Dimethylsulfide (Reduced Sulfur) (N), Primary Particles (A,N).

**Source-Specific Emissions (available):** Aircraft Emissions (A), Lightning (N), Nitrogen Oxides in Soils (A), Sulfur from Volcanoes (N).

**Source-Specific Emissions (in progress):** Biomass Burning (A & N), International shipping.

**Other Data (available):** Population, Cropland.

The April 2000 IGAC integration and synthesis project meeting in Aspen, Colorado generated additional support for several key GEIA data development activities, many of which are already in the GEIA Five-Year Plan:

- Provide inventories for roughly every 10 years over the past 30-50 years
- Provide seasonal cycles in the emissions
- Provide formulae for the dependence of natural emissions on soil moisture and temperature
- Further evaluate $NO_X$ emissions from soils
- Provide biomass burning emissions as a function of burning temperature and land use practices
- Combine the GEIA and EDGAR databases
- Provide organohalide emission estimates
- Distinguish between emissions into the canopy and those that reach the global atmosphere

GEIA will be pursuing these and related activities in the coming months. In particular, emphasis is being given to bringing the CO, $CH_4$, biomass burning and aircraft and ship emissions data online. GEIA is also developing enhanced question/answer feedback procedures, expanded formats, and flexible downloading protocols.

## The GEIA Center

The hub of the global network of institutions and agencies developing these emissions inventories is the GEIA Data Management and Communication Center. The Center's activities are supported by the U.S. National Science Foundation (NSF) and National Aeronautics and Space Administration (NASA) and directed by Paulette Middleton of RAND Environment. The Center's role is

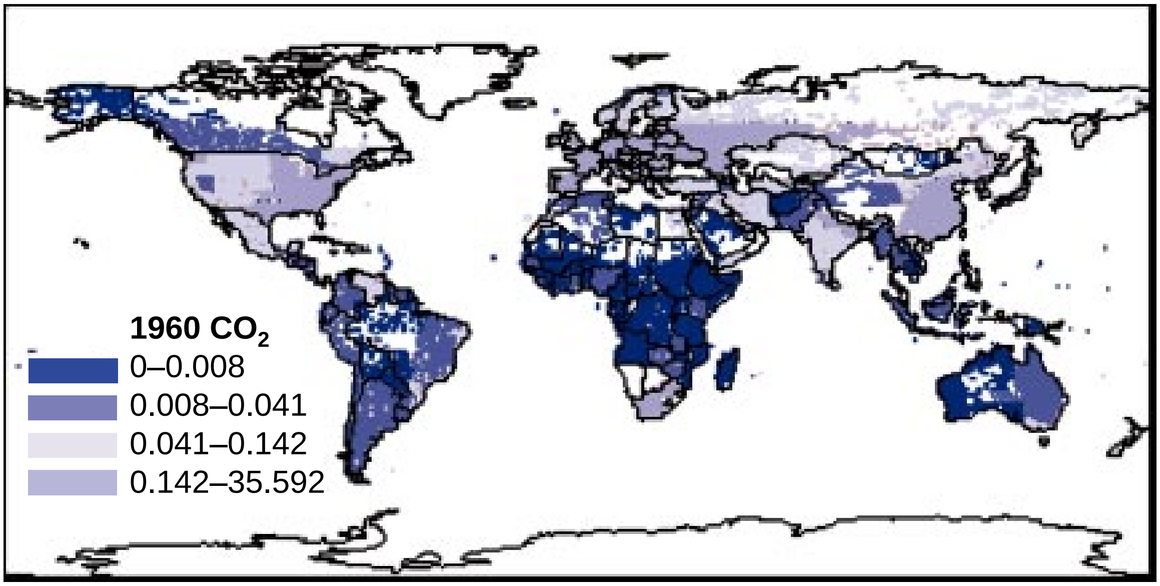

**Figure 1**. A graphic depiction of global distribution of carbon dioxide, one of the highly policy-relevant compounds inventoried by GEIA (http://weather.engin.umich.edu/geia/).

to manage the website at http://weather.engin.umich.edu/geia, to coordinate the distribution of emission inventory data, and to manage GEIA outreach activities.

As part of the expanded outreach strategy to enhance the exchange of information on GEIA and to encourage participation in GEIA, the GEIA Center recently developed the GEIA Newsletter. Each newsletter will have highlights from projects and workshops as well as other timely news related to global emissions inventories. The first newsletter can be found at http://weather.engin.umich.edu/geia/ and can be downloaded in PDF format. This allows the newsletter to be printed in hardcopy form or to be passed on to other interested readers.

In addition to this newsletter, the GEIA Center is expanding its e-mail network, establishing more two-way links to related websites, and developing information materials such as a new brochure. The website survey of users is being upgraded and follow-up discussions with users are being initiated. The survey will be similar to one developed in 1998 to help in the development of GEIA's first 5-year plan. The survey requested information on users and their use of the GEIA data, their evaluation of the databases, and more detailed evaluation of specific data sets and needs. Results so far have been extremely valuable in developing effective ways to improve the quality and usefulness of GEIA data.

Currently the GEIA Center tracks the number of visitors to the GEIA website, the number and type of downloads, and users' comments regarding their use of the data and general suggestions for GEIA. The results of this tracking are located on the GEIA site. For example, there were 540 downloads of data in 1998 and 704 downloads in 1999. The nitrogen oxides inventory was most popular in 1998, while natural VOC inventory was the most popular download in 1999.

As previously indicated, the major goal of GEIA is to provide peer-reviewed, documented emission inventories. However a number of emission inventories have been developed for specific needs, e.g., by modelers, independently of GEIA activities. It is the GEIA intent to be a focal point for information on emissions. Therefore, in cases where there is no GEIA inventory for a particular species, the GEIA website has added links to other data sources, e.g., to the EDGAR database.

## Workshops

Workshops are essential to GEIA's productivity and growth. International workshops on global emissions inventories have been held approximately once each year since GEIA was organized. The workshops have usually been organized in conjunction with major, related international meetings. All scientists involved with either emission inventory preparation or using emission inventories are encouraged to attend these meetings.

## Previous Workshops

1. September 1990, in Chamrousse, France, in association with the 7th CACGP Symposium on Global Atmospheric Chemistry.
2. December, 1991 in Baltimore, Maryland, USA, in conjunction with CHEMRAWN (Chemical Research Applied to World Needs).
3. June, 1992 near Oslo, Norway, sponsored by the Norwegian Institute for Air Research.
4. Early 1993 at Amersfoort, The Netherlands, under the auspices of the National Institute of Public Health and Environmental Protection.
5. Late 1993, at NCAR, Boulder, Colorado, USA with modelers in conjunction with a symposium on "Challenges in Atmospheric Chemistry and Global Change: Yesterday, Today and Tomorrow," held by NCAR to honor Paul Crutzen on his 60th birthday.
6. September, 1994, in Fuji-Yoshida, Japan in conjunction with the Eighth CACGP Symposium and the Second IGAC Scientific Conference.
7. July, 1995, in Gothenburg, Sweden in conjunction with the conference "Acid Reign 95".
8. May, 1996 in Toronto, Canada.
9. November, 1997 in Bilthoven, The Netherlands, hosted by the Netherlands Governmental Agency on the Environment and followed by the IPCC Expert Meeting on Methods for the Assessment of Inventory Data Quality on 5-7 November 1997.
10. August, 1998 at the University of Washington, Seattle, Washington, USA, in conjunction with the Joint CACGP/IGAC International Symposium on Global Atmospheric Chemistry.
11. September, 1999 in Bologna, Italy, in conjunction with the Sixth IGAC Scientific Conference.

## Invitation and GEIA Contacts

Everyone is invited to become involved in GEIA and to contribute to emission inventory preparation. Feedback on GEIA databases, their access and suggested improvements are always welcomed. Contributions to the evaluation of existing emission inventories and their uncertainties are especially valuable.

Details on GEIA, the five-year plan, results of a GEIA-wide survey that provided input for the planning process, and the GEIA projects and data themselves, can be found at http://www.geiacenter.org, along with information on future GEIA meetings.

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# Emission inventories of natural sources

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## Introduction

The *Global Emissions Inventory Activity* (GEIA) aims at providing global gridded emissions inventories to science and policy communities for all trace gases, in particular those that are relevant for global atmospheric chemistry. These emissions may stem from anthropogenic or natural sources. Natural sources may relate to processes in the soil (e.g., bacteria), in vegetation (plants), in the oceans (emission from dissolved amounts or produced by algae, or acting as a sink by absorption and dissolution in the water), in the Earth's crust (volcanoes or gas/oil seepage), or in the atmosphere (lightning). These emissions often depend strongly on climate, soil or water characteristics and thereby show a strong temporal variation in seasonality, diurnal cycle or both. Thus, natural sources often have a distinctly different character than have anthropogenic sources, which are comparably constant with respect to seasonality. In addition, the source strength as well as the spatial distribution of natural sources may differ substantially from year to year. An extreme example is volcanic emissions. Besides natural sources, natural sinks may occur locally, e.g., in some ocean areas where compounds are absorbed from the atmosphere.

Even if one is primarily interested in the effect of man-made emissions, the background of natural emissions (or sinks) of the same compounds and of related species must also be considered. For example, large datasets concerning temperature, precipitation, soil characteristics and oceanic parameters must be taken into account. Many of the natural emissions inventories are thus the result of modeling of the underlying processes using these global datasets. The resulting emission inventories either relate to a multi-year average showing the characteristic spatial and seasonal distribution of the natural emission sources, or present episodic inventories for specified years based on geo and climate data for these specific years.

In this paper we will review the GEIA emissions inventories of natural sources and also discuss others published in the literature, comparing their global source strengths with anthropogenic sources.

## GEIA inventories

Species presently covered by GEIA are related to acidification, ozone depletion, tropospheric ozone formation, climate change, aerosol formation and pollutants such as heavy metals and persistent organic pollutants (POPs) that are poisoning people and ecosystems. The extent to which N–, S–, Cl–, and C–containing compounds and other species are emitted by natural sources differs from compound to compound. For example, in Figure 1 total global emissions from natural and anthropogenic sources of $NO_X$, $NH_3$ and $N_2O$ are compared. It clearly shows that the share of man-made emissions in the total source strength can differ considerably but also that uncertainties can be quite large.

Emission inventories are available for the following species:

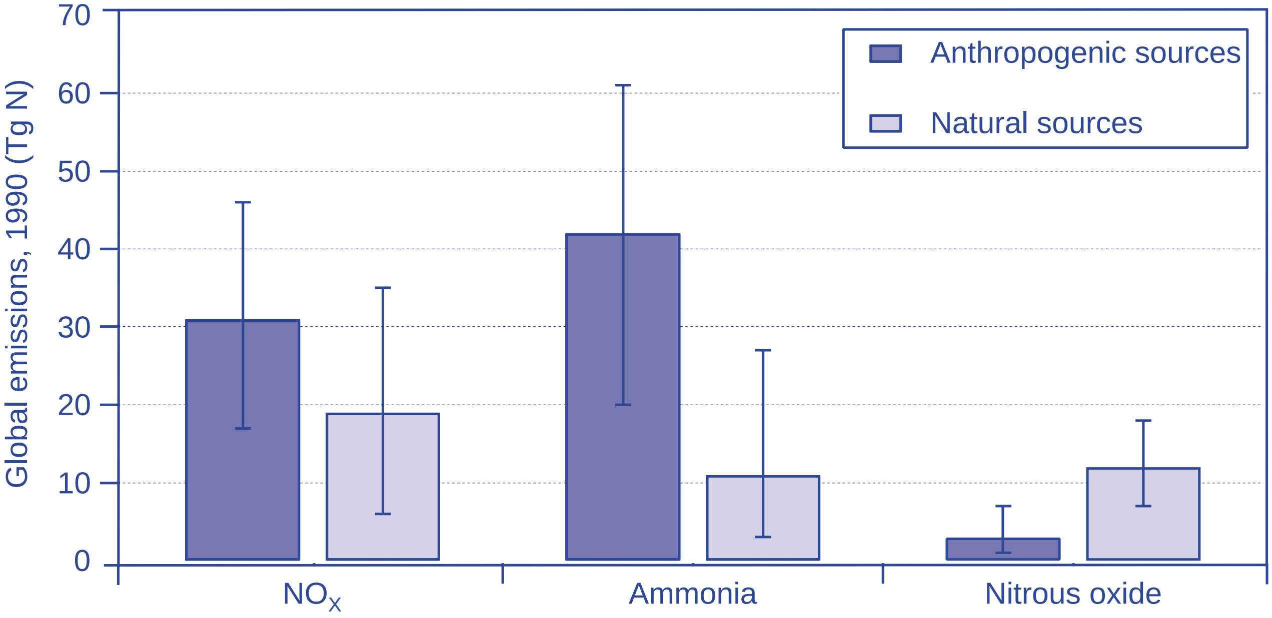

**Figure 1**. Contribution and uncertainty of global anthropogenic and natural N emissions in 1990. [from Olivier *et al.* (1999)].

**Table 1**. Natural emissions of nitrogen and sulfur compounds.

| Source/Compound | Emission (Tg N or S/yr) | Reference |
|---|---|---|
| **$NO_x$ emissions:** | | |
| – soils (including canopy sink) | 5.5 (4-12) | Yienger and Levy [1995] |
| | 13 (10-21) | Davidson and Kingerlee [1997] |
| – lightning | 12.2 (5-25) | Price *et al.* [1997a,b] |
| | 5 (2-10) | Pickering *et al.* [1997] |
| **$NH_3$ emissions:** | | |
| – natural soils | 2.4 (0-10) | Bouwman *et al.* [1997] |
| – oceans | 8.2 (3-16) | Bouwman *et al.* [1997] |
| – wild animals | 0.1 (0-1) | Bouwman *et al.* [1997] |
| **$N_2O$ emissions:** | | |
| – natural soils | 7.5 (3.7-11.3) | Bouwman *et al.* [1995] |
| – oceans | 3.6 (2.8-5.7) | Nevisson *et al.* [1995] |
| – freshwater and coastal ecosystems | 1.9 (1-9) | Seitzinger and Kroeze [1998] |
| **S emissions:** | | |
| – volcanoes: $SO_2$ | 6.7 (25 yr average) | Andres and Kasgnoc [1998] |
| Other S species | 3.7 (25 yr average) | Andres and Kasgnoc [1998] |
| – oceans: DMS | p.m. | [Various models] |

- acidification: $NO_x$ from soils and lightning; $NH_3$ from natural soils, oceans and wild animals
- aerosol formation: $SO_2$ from volcanoes; DMS from oceans
- climate change: $CH_4$ from wetlands, termites, oceans/hydrates; $N_2O$ from natural soils and oceans
- tropospheric ozone: CO from vegetation and oceans; CO soil sink; NMVOC from vegetation
- major reactive chlorine compounds: $CH_3Cl$, $CHCl_3$, $CH_2Cl_2$, $C_2HCl_3$, and $C_2Cl_4$ from oceans; $CH_3Cl$ and $CHCl_3$ from land-based sources; HCl and $ClNO_2$ from sea salt dechlorination.

For the following sources inventories are in progress:

- radionuclides
- emissions from natural biomass burning (wildfires in temperate regions) (for various species).

Natural $CH_4$ inventories do not reside at the GEIA website, but are available at http://www.giss.nasa.gov/data/ch4fung. Inventories for DMS from oceans are published in the literature, but not yet available at the GEIA site. Natural CO emissions from vegetation and oceans are discussed in the literature and used by many atmospheric modellers, but to date no comprehensive emission inventory at 1° x 1° has been compiled. Within the EU-funded project 'POET' which studies the **P**recursors CO, $NO_x$, $CH_4$ and NMVOC of **O**zone and their **E**ffect on the **T**roposphere [Granier, 1999], the gridded inventory for CO from vegetation will be based on the GEIA inventory for NMVOC from vegetation by Yienger and Levy [1995] and the small ocean source of 13 Tg CO will be based on Bates *et al.* [1995]. For the soil sink of CO the project will not provide one dataset, since the models calculate this sink from the calculated surface CO concentration and the assumed deposition velocity. Resulting gridded emission inventories will be made available for the IGAC modeling community at large.

### N– and S–inventories

For the natural sources of the nitrogen compounds $NO_x$, $N_2O$, $NH_3$, mainly soils under natural vegetation and oceans, the reader is referred to Bouwman *et al.* [1995; 1997] for detailed descriptions of inventories of $N_2O$ and $NH_3$, to Yienger and Levy [1995] for $NO_x$ from soils and vegetation and to Price *et al.* [1997a,b] for $NO_x$ from lightning. Davidson and Kingerlee [1997] present an alternative estimate of global $NO_x$ emissions from soils, but they do not provide gridded emissions. In Nevisson *et al.* [1995] a more detailed description is provided of $N_2O$ emissions (and regional sinks) from oceans. Furthermore, Seitzinger and Kroeze [1998] have developed a global inventory of $N_2O$ emissions from freshwater and coastal marine ecosystems, originating from N inputs from mainly anthropogenic sources (so-called indirect $N_2O$ emissions, mainly from agriculture). The vertical profile of lightning emissions may be taken from Pickering *et al.* [1998], who also provide an uncertainty range of 2-10 Tg N, whereas Price *et al*. [1997b] conclude from a constraint analysis that the range would be 5-25 Tg N. As is the case for soils, alternative datasets also exist for lightning in the literature, but these are not always available on a 1° x 1° grid. As illustrated in Figure 1, total natural emissions of nitrogen gases have a share of 40%, 20% and 80% in the global total emission of $NO_x$, $NH_3$ and $N_2O$, respectively.

On the sulfur emissions from volcanoes we refer to the separate paper by Robert Andres [this issue].

### CO, $CH_4$ and NMVOC inventories

Interestingly, for CO at present no inventories on a 1° x 1° basis are reported for the natural sources from vegetation and oceans. Müller [1992] presented a 10° x 10° inventory for oceanic emissions of 162 Tg based on Erickson [1989], but has revised his estimate to 20 Tg, and Bates *et al.* [1995] estimate the ocean source at 13 Tg CO. However, various estimates are reported in the literature and in the 1997 GIM/IGAC model inter-comparison of 3D tropospheric CO distributions a range of source strengths was used [Kanakidou *et al.*, 1999]. For vegetation/soil the average was 160 Tg within a range of 100-280 Tg; for oceans these values were 50 (13-162) Tg. Table 2 provides an overview of the various estimates found in the literature, including the 'best estimate' provided by Khalil [2000] in the introduction of the special issue on CO of *Chemosphere: Global Change Science*, No. 1 of 1999. For comparison, we note that the second IPCC Assessment Report of 1996 estimated vegetation and ocean emissions to be in the range of 60-160 Tg and 20-200 Tg, respectively. Within the framework of EDGAR 3.0, an estimate is made of CO emissions from vegetation fires in temperate regions, which is 35 Tg in 1995 [Olivier *et al.*, 2000]. In addition, the global budget includes a soil sink, of which the total strength and spatial distribution are generally calculated from the surface CO concentration and the assumed deposition velocity.

Matthews and Fung [1987] presented a gridded inventory of $CH_4$ from natural wetlands based on an extensive analysis and arrived at a source strength of 110 Tg. Matthews [2000] reviews the literature up to 1997 and concludes that estimates of wetland emissions are converging to a level around 100 Tg, however with an un-

**Table 2**. Natural emissions of CO, $CH_4$ and NMVOC.

| Source/Compound | Emission (Tg N/yr) | Reference |
|---|---|---|
| **CO emissions:** | | |
| - vegetation | 75 (50-100) | Seiler and Crutzen [1990] |
| | 90 (20-200) | Khalil and Rasmussen [1990] |
| | 160 (100-280) | Kanakidou *et al.* [1999] (model intercomparison) |
| | 230 (100-400) | Khalil [1999] |
| - oceans | 100( 10-190) | Seiler and Crutzen [1990] |
| | 13 (6-30) | Bates *et al*. [1995] |
| | 50 (20-80) | Khalil and Rasmussen [1990] |
| | 50 (13-162) | Kanakidou *et al.* [1999] (model intercomparison) |
| | 10 (0-30) | Khalil [1999] |
| - temperate vegetation fires | 35 (25-75) | Olivier *et al.* [2000] |
| **$CH_4$ emissions:** | | |
| - wetlands | 110 (50-170) | Matthews and Fung [1987], Matthews [2000] |
| - termites | 20 (?-?) | Fung *et al.* [1991] |
| | 20 (18-22) | Sanderson [1995] |
| - hydrate/clathrates | 10 (?-?) | Fung *et al.* [1991] |
| - gas seepage | ? (8-65) | Hovland *et al.* [1993] |
| | 55 (50-60) | Lambert and Schmidt [1993] |
| - open ocean | 3.5 (3-4) | Lambert and Schmidt [1993] |
| - oil seepage | p.m. | |
| **NMVOC emissions**: | | |
| - vegetation: isoprene | 503 (200-800) | Guenter *et al*. [1995] |
| monoterpene | 127 (50-500) | Guenter *et al*. [1995] |
| - oceans: isoprene | 5 (?-?) | Guenter *et al*. [1995] |
| - gas seepage | p.m. | Hovland *et al.* [1993] |
| - marine oil seepage | 0.6 (0.2-6) | Wilson *et al.* [1974] (maximum if all oil discharged is emitted into the air) |

certainty still of about 50%. Recently, Darras et al. [1999] have reported on an IGBP-DIS Wetland Data Initiative to determine the global extent of wetlands. In this paper the Matthews and Fung dataset was compared to three other wetland datasets (ISLSCP, DISCover and Ramsar). Fung *et al.* [1991] and Gornitz and Fung [1994] provide gridded inventories for $CH_4$ from termites and hydrate/clathrate in the Soviet Arctic and between 76°-85° N of 20 and 10 Tg $CH_4$, respectively. These inventories are, however, highly uncertain. An alternative inventory of termites' emissions, with the same global source strength of 20 Tg is presented in Sanderson [1996]. All of these inventories, except for the termite $CH_4$ of Sanderson, are available at http://www.giss. nasa.gov/data ch4fung.

Table 3: Natural emission of reactive chlorine species.

| **Source/Compound** | **Emission (Gg Cl/yr)** | **Reference** |
|---|---|---|
| Oceanic emissions: | | Khalil *et al.* [1999] |
| $CH_3Cl$ | 460 | |
| $CHCl_3$ | 320 | |
| $CH_2Cl_2$ | 160 | |
| $C_2HCl_3$, | 20 | |
| $C_2Cl_4$ | 20 | |
| Land-based emissions: | | Khalil *et al.* [1999] |
| $CH_3Cl$ | 100 | |
| $CHCl_3$ | 200 | |
| Sea salt dechlorination: | | Erickson III *et al.* [1999] |
| HCl | 7600 | |
| $ClNO_2$ | 60 | |

Finally, within the framework of the POET project [Granier, 1999], a $CH_4$ and NMVOC inventory for emissions from oil and gas seepage from continental shelves and from land will be developed. This will be based on Hovland *et al.* [1993] for $CH_4$ from gas seepage through the seabed, presenting a global total estimate of 8-65 Tg of $CH_4$, of which a fraction will pass the water column and into the atmosphere. This paper builds on a study by Wilson *et al.* [1994], which provides an estimate of natural marine oil seepage of 0.2-6 Tg oil discharged into the water with a best estimate of 0.6 Tg. A part of the oil will dissolve and emit into the air. Lambert and Schmidt [1993] argue that the oceanic source of methane is likely to be 50-60 Tg, of which 3.5 (3-4) Tg is emitted from open ocean. Clark *et al.* [2000] measured that gas bubbles at the ocean surface above seeps contain about 60% $CH_4$ and about 10% NMVOC. The seepage inventories are on the continental shelf in addition to the hydrate/clathrate source estimated by Fung *et al.* [1991].

The major natural source of NMVOCs is vegetation, although the global source strength is rather uncertain [Guenter *et al.*, 1995]. The main compound groups emitted by plants and trees are isoprene and monoterpenes. Also, there are a few small oceanic sources (see Table 2). In a separate paper in this issue the natural NMVOC inventory is discussed in more detail by Alex Guenther.

### Reactive chlorine emission inventories (RCEI)

Recently a comprehensive set of gridded inventories of reactive chlorine compounds has been published in a series of eight consecutive papers in *Journal of Geophysical Research - Atmospheres*, 104 (D7), 8331–8440, April 1999. These include estimates of natural sources, of which the global source strength is summarized in Table 3. Oceans appear to account for about 12% of the global annual emissions of methyl chloride, much lower than often has been assumed. For chloroform, both oceans and land appear to be major sources.

## Acknowledgement

I thank Jean-Francois Müller for the informative discussions on the natural source inventories to be used in the POET project.

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# A time-averaged inventory of subaerial volcanic sulfur emissions prepared for the GEIA

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Volcanoes emit gases, aerosols, and fine particulate matter in modes ranging from continuous to ephemeral. From 1975 to 1985, an average of 56 volcanoes erupted yearly. While some showed continuous activity, others erupted less frequently or only once, so that 158 volcanoes actually erupted over this time period. This number increases to 380 volcanoes with known eruptions this century, 534 volcanoes with eruptions in historical times, and more than 1500 volcanoes with documented eruptions in the last 10,000 years [McClelland *et al*., 1989].

Sulfur (S) is a common element in many volcanically-emitted compounds. Sulfur dioxide ($SO_2$) and hydrogen sulfide ($H_2S$) are the primary S-containing gases emitted. Other S-containing species usually occur in smaller quantities. The distribution between these various species differs not only between volcanoes, but also during a single volcanic episode at one volcano. Likewise, the flux, or S mass emitted per unit time, varies for one volcano and between volcanoes.

Like many anthropogenic S emissions, volcanoes can be represented as point sources for S venting to the atmosphere. However, many volcanoes occur in areas where anthropogenic point sources are not so significant. Volcanic S sources often occur in areas not strongly affected by other natural and anthropogenic S. Graf *et al*. [1997] estimated that 64% of quiescently degassed S ultimately transforms into sulfate. Since much of this sulfate forms in areas where anthropogenic sulfate has not reached saturation in radiation balance terms, volcanic S emissions create a sulfate burden with disproportionate radiative effects. General circulation model calculations indicate that direct and indirect radiative forcings due to volcanically-derived sulfate are on par with that due to anthropogenically-derived sulfate. Chin and Jacob [1996] found similar results in that volcanic S composes 20–40% of the sulfate in the mid-troposphere globally and 60–80% over the North Pacific. It also accounts for a significant percentage of the sulfate burden in the upper troposphere at high latitudes. These disproportionate effects are realized even though volcanic S accounts for only 7% of the total S emitted to the atmosphere in their chemical transport model, yet it accounts for 18% of the column sulfate burden globally. Thornton *et al*. [1996] show that volcanic $SO_2$ becomes an important cloud condensation nucleus source in the North Pacific. These three studies suggest that volcanic S plays a role similar to that of anthropogenic S in direct and indirect radiative effects and cloud lifetime effects [Charlson *et al*., 1992]. Some of these effects are due to the ability of volcanoes to loft S high into the troposphere and stratosphere where the S residence times increase significantly.

Due to the importance of volcanic S in the global S cycle, especially the atmospheric portions thereof, a time-averaged inventory of subaerial (directly venting to the atmosphere) volcanic S emissions was compiled. Primarily for the use of global S and sulfate modelers, it incorporates the temporal, chemical and spatial inhomogeneities inherent to the subject that are necessary for more accurate regional and global atmospheric chemistry transport models [e.g., Chuang *et al*., 1997; Weisenstein *et al*., 1997; Chin *et al*., 1996; Feichter *et al*., 1996; Pham *et al*., 1995]. The secondary purposes of this inventory are to compile the existing $SO_2$ data and facilitate the creation of other emission inventories for volcanic species amenable to species/S ratios [e.g., Andres *et al*., 1993; Nriagu, 1989; Symonds *et al*., 1988; Lantzy and Mackenzie, 1979]. This work was originally reported in Andres and Kasgnoc (1998). More information about the Global Emissions Inventory Activity (GEIA), its products, and the inventory presented here is available at the GEIA website (http://weather. engin.umich.edu/geia).

This inventory relies upon the 25-year history of S (primarily $SO_2$) measurements at volcanoes. Subaerial volcanic $SO_2$ emissions indicate a 13 Tg/a $SO_2$ time-averaged flux, based upon an early 1970s to 1997 time frame. When considering other S species present in volcanic emissions, a time-averaged inventory of subaerial volcanic S fluxes is 10.4 Tg/a S (Table 1). These time-averaged fluxes are conservative minimum fluxes since they rely upon actual measurements. The temporal, spatial and chemical inhomogeneities inherent to this system gave higher S fluxes in specific years.

This inventory is based upon an extensive compilation of the available, measured volcanic S fluxes. These data were collected from 214 published references, personal communications, and three volcanological conference presentations. Two to three times more references were searched. The conference presentations, as well as two electronic mail messages to the VOLCANO listserv, allowed many opportunities for inventory data discussion with volcanologists and atmospheric scientists.

Due to the nature of the available data and the volcanic source inhomogeneities, the GEIA inventory presented here is time-averaged. If the inventory was constructed for the benchmark GEIA year of 1990, there would be too few data for a reliable extrapolation. Additionally, if the focus is long-term volcanic source modeling, then a multi-year source average will produce more accurate results than a single index year.

The above volcanic $SO_2$ flux, derived from the COSPEC and TOMS archives, does not include examples of larger eruptions that occurred prior to this modern instrumental record. For example, the 1963 Agung eruption released 7 Tg $SO_2$ over two days [Self and King, 1996], the 1783-1784 Laki eruption released 122 Tg $SO_2$ over eight months [Thordarson et al., 1996], and the 14.7 Ma Roza eruption released 12,420 Tg $SO_2$ over 10 years [Thordarson and Self, 1996].

The impact of these larger events has not been as well studied as that of the 1991 Mount Pinatubo eruption which scales between Agung and Laki. Resulting effects of Pinatubo include a 4 $W/m^2$ decrease from 40ºS to 40ºN and 8 $W/m^2$ from 5ºS to 5ºN. Globally averaged, the Pinatubo eruption caused a negative forcing greater than the positive, anthropogenically enhanced, greenhouse forcing for the latter part of 1991 and most of 1992. This translated to an ENSO-adjusted, global, tropospheric cooling around 0.5ºC in 1992 [McCormick *et al*., 1995].

Natural processes, including volcanoes, emit approximately 24 Tg/a S to the atmosphere [Bates *et al*., 1992; Spiro et al., 1992]. The volcanic S flux calculated here is 43% of the total natural S flux. Anthropogenic activities emit approximately 79 Tg/a S to the atmosphere [Bates *et al*., 1992; Spiro *et al*., 1992; Andreae, 1990]. The volcanic S flux calculated here is 13% of the anthropogenic flux. The bulk of the anthropogenic flux is located in the Northern Hemisphere while volcanic fluxes occur in more focused belts around the world.

In summary, Table 1 details a conservative, time-averaged inventory of subaerial volcanic S flux that incorporates the temporal, spatial and chemical inhomogeneities of this source. While this average flux is based upon the actual, modern instrumental record, TOMS measurements indicate that the 1991 Pinatubo eruption released an equivalent amount of S in one event. Even larger contributions are indicated for eruptions that predate the modern instrumental record.

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**Table 1**. GEIA Volcanic S inventory. Global distribution and elevation of these sources are given in the online version. Mg/d = megagrams ($10^6$)/day, Tg/a = teragrams ($10^{12}$)/annum.

| | Mg/d $SO_2$ | Tg/a $SO_2$ | Tg/a S |
|---|---|---|---|
| Continuous $SO_2$ emissions | 26,200 | | |
| Sporadic $SO_2$ emissions | 200 | | |
| Total measured $SO_2$ emissions* | 26,400 | 9.66 | |
| TOMS measured $SO_2$ emissions# | | 3.7 | |
| Total $SO_2$ emissions | | 13.4 | 6.7 |
| Other S species$ | | | 3.7 |
| Total volcanic S inventory | | | 10.4 |

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* Total measured $SO_2$ emissions are primarily based upon COSPEC measurements of smaller emission sources (includes 49 continuous and 25 sporadic volcanoes).

\# TOMS measurements are satellite-obtained measurements of larger emission sources.

$ Other S species based upon direct samples of volcanic gases (to obtain other S/$SO_2$ ratios) and separately measured $SO_2$ fluxes.

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# IGAC-GEIA natural VOC emissions

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Volatile organic compounds (VOC) are among the trace gases that have a role in controlling the distribution of chemical species in the atmosphere. Global emissions of VOC are dominated by natural (e.g., biogenic) sources. Scientific investigations of biogenic VOC emissions were initiated in the Soviet Union in 1928 and a considerable VOC flux measurement database was produced by the 1950s [Isidorov, 1993]. Biogenic VOC emissions research in North America began in the 1950's [Went, 1960] and reached a period of peak activity in the mid 1970s to early 1980s due to speculation that these compounds had a role in urban ozone formation. These efforts included systematic enclosure surveys of North American vegetation species as well as above-canopy flux measurements of emissions from entire ecosystems [e.g., Rassmussen, 1972; Zimmerman, 1979; Winer *et al*., 1982; Arnts *et al*. 1982].

This period of activity in North America was curtailed when some modeling studies failed to demonstrate that biogenic emissions were an important component of urban ozone formation [Altshuller, 1983]. At the same time, researchers in Europe [e.g., Knoeppel *et al*., 1981; Hov *et al*., 1983; Isidorov *et al*., 1985], Asia [e.g., Yokouchi *et al*., 1983] and Australia [Ayers and Gillett, 1988] were increasing efforts in this research area and making significant contributions.

The implementation of IGAC-GEIA was a significant step towards creating an international community of scientists interested in biogenic VOC emissions. Over thirty scientists from five continents attended the first IGAC-GEIA natural VOC working group meeting, held in Boulder Colorado in 1992. One outcome of this meeting was the initiation of an effort to produce a global biogenic VOC emission inventory [Guenther *et al*., 1995]. An equally important result was the strengthening of interactions among these scientists. This resulted in collaborations both within and outside of the IGAC-GEIA framework. In addition, the increasing interest in this field led to special sessions on biogenic VOC at international meetings as well as meetings solely devoted to this topic including a biennial Gordon Research Conference that focused on biogenic hydrocarbons and the atmosphere.

Among the challenges faced in developing a global natural VOC emission inventory are the large number of compounds and sources. The first IGAC-GEIA model included only one compound (isoprene) and three VOC categories (monoterpenes, other reactive VOC, and other VOC). Emissions from different sources were discussed but were not identified separately in the inventory. The next generation of the IGAC-GEIA natural VOC emission model will predict emissions of about 40 individual

VOC compounds from seven different sources. Each source is responsible for the emission of more than one chemical species and some compounds are emitted by more than one source. The results of the past decade of biogenic VOC emissions research is being incorporated into the next generation model. This includes improved landscape characterization, emission factors, emission activity algorithms, and other model components.

The initial product of the IGAC-GEIA natural VOC working group was an inventory providing monthly total emission on a 1° latitude by 1° longitude grid. It was immediately clear that this was not sufficient for many chemistry and transport modelers. Since many biogenic VOC have very short lifetimes (minutes or hours), emission estimates were needed at relatively high spatial and temporal resolution. In addition, biogenic VOC emissions are strongly influenced by environmental conditions (e.g., temperature) and predictions for a specific scenario could vary greatly from that provided in the static IGAC-GEIA inventory. Thus many modelers by-passed the static inventory and directly used the model procedures to generate emission estimates for their own specific scenarios. As future efforts focus on Earth system models that simulate chemistry, climate and emissions, as well as the feedback coupling between them, it will be even more important to provide dynamic emission models rather than static emission inventories. There is also a need for a consistent approach to modeling emissions of different compounds from the same source, such as NO and VOC emissions from soil microbes. Even more important is coordination of procedures for modeling emission and deposition of the same compound since some emission routines may already account for deposition of a trace gas as it is transported through a vegetation canopy. These are all issues that IGAC-GEIA is considering as it moves to its next phase.

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## Correction to IGACtivities #21

1. In the "Measurement methods for atmospheric OH" article, the reference for Figure 1 was incorrect. The figure has been adapted from Dorn *et al.*, 1996.
2. In the same article, the wrong graphic and caption was used for Figure 6. The correct figure is provided below.

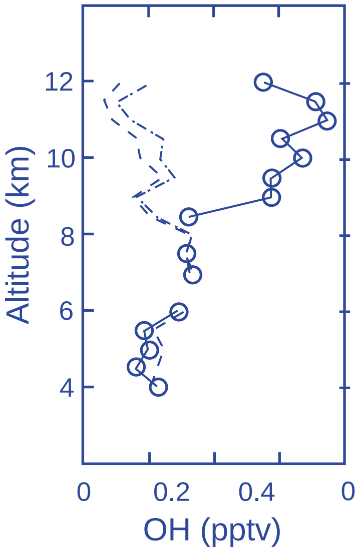

**Figure 6**: Altitude profiles of measured (open circles) and modeled OH for 10 May 1996 during SUCCESS. Measurements and models are averaged into 0.5 km altitude bins. Models with (dash-dot line) and without (dashed line) acetone are compared. (Adapted from Brune, W.H., *et al.*, *Geophys. Res. Lett.*, 25, 1701-1704, 1998, copyright by the American Geophysical Union)

# Announcements

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**From Air Pollution to Climate Change**

Institute of Environmental Research
Chulalongkorn University
Bangkok, Thailand
22-23 January 2001

Tel. (66 2) 218-8126 to 9; Fax. (66 2) 255-4967
http://www.start.or.th/IGAC7/

*For information, contact:*
Professor Jariya Boonjawat (jariya@start.or.th)

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## GCTE / IGAC Workshop

"Nitrogen Isotopes in Global Change Research"

**Contacts**:
Dave Evans (devans@comp.uark.edu)
Peter Hogberg (peter.hogberg@sek.slu.se)

**Location and dates**:
TBA

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## A Changing Atmosphere

8th European Symposium on the Physico-Chemical
Behavior of Atmospheric Pollutants
European Commission; DG Research;
Joint Research Centre;
17-20 September 2001
Torino, Italy

Tel +39 0332 789958; Fax +39 0332 785704
Further information: frank.raes@jrc.it
http://www.ei.jrc.it/ap/events/torino2001/

## Challenges of a Changing Earth

A Global Change Open Science Conference
Amsterdam, The Netherlands
10-13 July 2001

International Geosphere-Biosphere Programme (IGBP)
International Human Dimensions Programme on
Global Environmental Change (IHDP)
World Climate Research Programme (WCRP)

Phone: +46-8-16-64-48; Fax: +46-8-16-64-05
http://www.sciconf.igbp.kva.se/

For further information, contact:
Rowena Foster (Rowena.Foster@dwe.csiro.au)

### Rationale and Philosophy

The Global Change Open Science Conference will present the latest scientific understanding of global environmental change at three levels: (i) the integrated level of the IGBP core projects and the IGBP as a whole; (ii) cross-cutting research involving the WCRP and IHDP, as well as regional research coordinated by START and other groups; and (iii) the individual level of the research projects which contribute to IGBP/ WCRP/IHDP networks and provide the broad, substantive base on which the integrating activities of the three programmes are built.

The Conference will also look forward. The last day will present the visionary and creative new approaches in Earth System Science, for studying the thresholds, nonlinearities and teleconnections of a complex planetary system in which human activities are intimately interwoven with natural processes. It will outline a research programme for our current era of increasing human domination of many global-scale processes - the "Anthropocene".

IGAC*tivities* Newsletter

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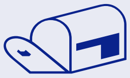

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