CONF-941138--2

BNL 60309

THE GEIA GLOBAL GRIDDED INVENTORY OF ANTHROPOGENIC VOCS

Carmen M. Benkovitz
Environmental Chemistry-Division
Brookhaven National Laboratory
Upton, NY 11973

and

Jan J. M. Berdowski and Chris Veldt
TNO MW
Delft, the Netherlands

April 1995

Presented at
International Conference on the Emission Inventory:
Applications and Improvement
Raleigh, NC
Nov. 1-3, 1994

This research was performed under the auspices of the U.S. Department of Energy under Contract No. DE-AC02-76CH00016.

The GEIA Global Gridded Inventory of Anthropogenic VOCs

Carmen M. Benkovitz
Building 426
Brookhaven National Laboratory
Upton, NY, 11973
USA

Jan J.M. Berdowski
Chris Veldt
IMW-TNO
P.O. Box 6011
2600 JA Delft
The Netherlands

ABSTRACT

Modeling assessments of the atmospheric chemistry, air quality and climatic conditions of the past, present and future require as input inventories of emissions of the appropriate chemical species constructed on appropriate spatial and temporal scales. The task of the Global Emissions Inventories Activity (GEIA) of the International Global Atmospheric Chemistry Project (IGAC) is the production of global inventories suitable for a range of research applications. Current GEIA programmes are generally based on addressing emissions by species; an international working group of interested participants cooperates in the work needed to compile each inventory. The work of the GEIA programme addressing the compilation of a global inventory of anthropogenic emissions of Volatile Organic Compounds (VOCs) gridded with 1° resolution is presented. Past studies were used to identify anthropogenic activities according to their contribution to global VOC emissions; based on results of these initial studies, activity and species groupings for emissions reporting have been selected. Current status of the work of the committee is discussed. Detailed information on available activity rates, emission factors, and speciation profiles for each defined sector is being compiled. Links to investigators working on the compilation of VOC emissions on a regional level have been established.

INTRODUCTION

The International Global Atmospheric Chemistry Project (IGAC) [1] is an international cooperative effort of atmospheric scientists designed to measure, understand, and attempt to predict changes in the chemistry of the global atmosphere over short and long time scales. Of particular interest are changes in the oxidizing capacity of the atmosphere, the impacts of these changes on climate, and the chemical interactions of the atmosphere and biota. These goals are broad and include several environmental issues of urgent concern, including greenhouse warming due to the accumulation of trace gases in the atmosphere, depletion of stratospheric ozone, increased acidity of rainfall, increased oxidant levels in the troposphere, and resulting biological damage.

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 mathematical models of transport and transformations in the atmosphere. These models rely in part on inventories of emissions constructed on appropriate temporal and spatial scales and including the required chemical species. The production of such inventories, initially regarded as adjunct to modeling activities, is now a separate area of research whose importance to the accuracy of results of modeling and assessment activities has been fully recognized. The myriad of problems involved in the compilation of accurate inventories on a local or

regional basis is multiplied manyfold when the geographic area of interest is extended to the multinational, hemispheric, and global domains. Recognizing that the most accurate information on emissions is usually developed by experts from individual countries, the IGAC Steering Committee has defined an activity, the Global Emissions Inventory Activity (GEIA) [2], whose ultimate and very ambitious goal is to establish emissions inventories for a number of trace species, incorporating fluxes from both anthropogenic and natural sources, with recognized accuracy and enough spatial, temporal and species resolution to serve as standard inventories for the international community of atmospheric scientists.

Organic compounds from both anthropogenic and biogenic sources have been identified as important precursor emissions in several current environmental problems, such as the formation of tropospheric ozone and the formation of species resulting in acid deposition. Compiling inventories of the organic compounds involved in these environmental problems has become one of the most challenging questions in atmospheric chemistry and modeling today. A large number of organic compounds are involved, and information on their chemical properties is still being developed. Most large transport and transformation models currently in use cannot calculate the individual concentrations of all of these compounds, and several different mechanisms have been developed to simplify representation of the chemistry in these models. In general, these chemical mechanisms combine the individual organic compounds in a small number of groups; the compounds integrated into each chemical group are selected according to the chemical reactions represented in the models. In addition, regulations addressing emissions of organic compounds have been developed well after the regulations addressing SO_2 and NO_x emissions; therefore, information on source locations, activity rates and emission factors needed to compile inventories of emissions of organic compounds has been developed only in a subset of industrialized nations. Within the GEIA activity, several committees are addressing the compilation of global inventories; currently active committees are listed in Table 1. The compilation of VOC emissions are being addressed by three GEIA committees; this paper describes the current work of the committee on anthropogenic emissions of volatile organic compounds (VOCs).

INVENTORY DEVELOPMENT

Inventory Specifications

Volatile Organic Compounds (VOC) in this inventory will include anthropogenic emissions of all organic compounds that exist in the gaseous state at ambient conditions as well as semi-volatile organic compounds from some large contributing source types, except for methane and fluorocarbons. The methodology to be followed in estimating emissions from each activity sector is as follows:

$$\text{VOC emissions} = (\text{activity rate}) \times (\text{emission factor}) \quad (1)$$

$$\text{speciated emissions} = (\text{VOC emissions}) \times (\text{speciation factors}) \quad (2)$$

For some sources the methodology is more complex, e.g., when the emission rate is influenced by environmental parameters, for example, evaporation losses from traffic; these effects will also be taken into account when estimating their emissions.

A first cut estimate of global emissions of VOCs from anthropogenic activities was compiled by Veldt [3], and was recently adapted by Berdowski [4]. Anthropogenic sources contributing to VOC emissions were prioritized according to their estimated contribution to these emissions; 87% of total emissions were contributed by sources in four sectors: small scale combustion, mobile sources, fuel production and distribution, and solvent use. Sources in other sectors that have smaller but significant contributions include the chemical industry, fossil fuel combustion processes, and uncontrolled waste burning. Due to the different quality of the information available for each of these sectors, emissions will be reported on separate files for the sectors listed in Table 2. Possible subdivisions of each sector will be decided as work progresses.

VOC speciation for global modeling up to the present demands less detail than present knowledge can supply. Selection of chemical species and groups was based therefore on the latter, allowing for any desired level of further aggregation. Speciation profiles for emissions for the first three

source groups in Table 2, comprising two thirds of anthropogenic VOC emissions, is known to a reasonable detail. These profiles were used to compile Table 3, which present the groups selected for the GEIA inventory. A compromise was made between reactivity, data availability and source strength.

Version 1 of the inventory will estimate emissions in 1990. The target temporal level is seasonal, which also represents a compromise between available information and modelers' needs.

Data Sources

United States and Canada. For the United States and Canada the U.S. Environmental Protection Agency (EPA) and the Atmospheric Environment Services (AES), Canada compile detailed inventories of VOC emissions [5]. Therefore, emissions from U.S. and Canadian sources for all sectors will be taken from the EPA 1990 Interim Inventory. The spatial allocation to the GEIA global 1° resolution grid will be accomplished using the 20-km resolution gridded population file developed for the National Acid Deposition Assessment Program (NAPAP) [6], with subsequent aggregation to the GEIA grid. The speciation profiles in the Air Emissions Species Manual [7] will be used to speciate these emissions to the individual chemical species level; results can the be aggregated to the selected groups.

Emissions from countries other than the United States and Canada will be addressed by sectors.

Small scale combustion. In this sector estimates of emissions from biomass burning in small appliances (i.e., for household heating, cooking, etc) and of emissions from small scale combustion of coal are covered; estimates of emissions from large biomass burning are being addressed by another GEIA committee. A proposal defining emission and speciation factors for this sector was prepared by C. Veldt (TNO, Delft, The Netherlands, 1992) and has been sent to a limited number of experts for comments. Responses and comments from the reviewers will be combined and a final proposal developed. Work on developing the activity rate information for this sector is being addressed as a cooperative effort between TNO/RIVM in The Netherlands and Harvard University in the U.S. using statistics from international organizations such as United Nations (UN), Food and Agriculture Organization (FAO), and the Biomass Users Network (BUN) (King's College, London, U.K.). Harvard has developed the information for African countries and China; TNO will help in compiling the information for the ten most important countries with respect to residential biomass burning, except for China and India.

Mobile sources. Emissions for this sector will be taken from an inventory compiled by Z. Samaras [8] that uses methodology developed and implemented for the Computer Programme to Calculate Emissions from Road Traffic (COPERT) [9-11]. The mobile sector will be divided into further subcategories, which are being developed by comparing the subcategories used in the U.S. EPA and the Samaras inventories. A comparison of these two mobile source inventories has been carried out by Samaras [12]. Emissions from the Samaras inventory will be allocated to the GEIA grid based on the 1° resolution population file developed by Logan (Jennifer Logan, Harvard University, manuscript in preparation, 1994). Speciation factors for European emissions have been developed by Veldt and van der Most [13], and are used in atmospheric chemistry modeling work covering Europe. Methodology is still needed to improve comparability of the inventories from this sector developed in the U.S. and Europe.

Solvent use. This is a difficult sector to address due to insufficient available data. An initial literature search has been conducted, and is still in the process of updating results, to compile and process activity and profile data for these sources. Emissions in western Europe are fairly well known, but emissions outside North America and western Europe comprise half of the global total. For the largest of these other countries, paint consumption is more or less known; however, it is estimated that 20 to 30% of the global emissions from this sector have to be inventoried by mere extrapolation. Results have summarized and sent for comments to experts through the U.S. Chemical Manufacturer's Association (CMA) and ICI in the United Kingdom.

Fossil fuel production, storage and distribution. The activity rate information will be compiled from several sources such as the information on refineries developed for the project to estimate global emissions of methane from petroleum sources conducted for the American Petroleum Institute [14], and

for the Long Term Ozone Simulation (LOTUS) project [15]. The emission factors are being developed based on emission factors from the U.S. EPA [16], the UNECE-EMEP (United Nations Economic Commission for Europe-European Monitoring and Evaluation Program) Emission Inventory Guidebook [17] and the EDGAR (Emissions Database for Global Atmospheric Research) work [18-20].

Chemical industry. Information on plant capacities in the Worldwide Petrochemical Survey [21] is being used as the starting point for information on activity rates for this sector. Information available in this database is the result of voluntary answers to survey questionnaires, and thus does not provide a complete listing of all existing chemical plants. Additional information on production is being assembled from literature surveys and from other sources such as the International Trade Commission, the United Nations Statistics, etc. The emission and speciation factors are being generated in conjunction with the development of the UNECE-EMEP Emission Inventory Guidebook [17].

Large combustion sources. This sector is not a significant contributor to global VOC emissions. Sharing information on locations and activity rates of large combustion sources will be discussed with the GEIA Committee on Anthropogenic Emissions of SO_2/NO_x. Activity rates and emission factors for small boilers outside North America have been prepared within the framework of the LOTUS [15] and EDGAR [19, 20] projects. For activity data, fuel consumption statistic of the International Energy Agency (IEA) and the UN have been used.

Waste. This sector suffers from a very diffuse definition and lack of information. Possible activities mentioned are managed landfills, smoldering dumps, etc. Work on the definition of this sector and on gathering information on activity rates will be carried out in conjunction with the GEIA Committee on Methane Emissions. Estimating emissions from this sector will be postponed to the second version of the VOC inventory.

Other sources. VOC emissions from aircraft in flight are being addressed by a group under the National Aeronautics and Space Administration (NASA); initial inventories of NO_x and VOC emissions for several scenarios are currently available from NASA [22]. Work on the development of speciation factors for VOC emissions is currently under way.

SAMPLE PRELIMINARY RESULTS

Table 4 summarizes the progress to date of the GEIA global inventory of anthropogenic emissions of VOCs and presents the current estimates of the total emissions by source type. Emission estimates for the four source types not being addressed by this working group were obtained as follows.

- Estimates for large biomass burning were derived from Andreae [23].
- Estimates for waste incineration and for miscellaneous sources not included in any of the other categories were derived from Berdowski [4].
- Estimates of emissions from aircraft are totals for the 1990 scenario described in Stolarski [22], converted to equivalent mass of carbon.

Figure 1 presents the relative contributions of each sector to the global totals.

To distribute the emissions to the GEIA 1° resolution grid, the point sources will be located directly. Sectors where emissions have been calculated by country will use the global population figures as surrogates to distribute their emissions. Population figures were allocated to a 1° resolution grid by Logan (Jennifer Logan, Harvard University, manuscript in preparation, 1994).

SUMMARY

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 mathematical models of transport and transformations in the atmosphere. These models rely in part on inventories of emissions constructed on appropriate temporal and spatial scales and including the required chemical species. Organic compounds from both anthropogenic and biogenic sources have been identified as important precursor emissions in several current environmental problems, such as the formation of tropospheric ozone and the formation of species resulting in acid deposition. Compiling inventories of the organic compounds involved in these

environmental problems has become one of the most challenging questions in atmospheric chemistry and modeling today. A large number of organic compounds are involved, and information on their chemical properties is still being developed; in addition, information on source locations, activity rates and emission factors has been developed only in a subset of industrialized nations. The GEIA the committee on anthropogenic emissions of VOCs is working on the development of a global inventory of these emissions for 1990 with 1° spatial resolution. The specifications for this inventory developed so far are:

- Volatile Organic Compounds (VOC) to be inventoried include anthropogenic emissions of all organic compounds that exist in the gaseous state at ambient conditions as well as semi-volatile organic compounds from some large contributing source types, except for methane and fluorocarbons.
- The general methodology to be followed in estimating emissions is based on activity rates, emission factors, and speciation factors; when the emission rates are influenced by environmental parameters this effect will also be taken into account.
- Emissions will be reported on separate files for the seven sectors listed in Table 2.
- Chemical speciation of emissions will be reported in the groups listed in Table 3.
- Version 1 of the inventory will estimate seasonal emissions in 1990.
- VOC emissions from U.S. and Canadian sources will be taken from the EPA 1990 Interim Inventory. The spatial allocation will be accomplished using the 20-km resolution gridded population file developed for NAPAP. The speciation profiles in the Air Emissions Species Manual will be used to speciate emissions.
- Emissions from countries other than the United States and Canada will be addressed by sectors.
- Activity rate information for the small combustion sector is being developed using statistics from international organizations such as the UN, FAO, and BUN. A proposal defining emission and speciation factors for this sector has been developed and sent to a limited number of experts for comments.
- Emissions for mobile sources will be taken from an inventory by Samaras that uses methodology developed and implemented for COPERT.
- Emissions from the solvent use sector are fairly well known in North America and western Europe; however, these countries emit only half of the global total. It is estimated that 20 to 30% of the global emissions from this sector have to be inventoried by mere extrapolation.
- The activity rate information for the fossil fuel production, storage and distribution sector is being compiled from several sources such as the American Petroleum Institute and the LOTUS project. The emission factors are being developed based on emission factors in the U.S. EPA AP-42 report, the UNECE-EMEP Emission Inventory Guidebook, and the EDGAR project.
- The activity rate information for the chemical industry sector is being compiled from several sources such as Worldwide Petrochemical Survey, literature surveys, the International Trade Commission, the United Nations Statistics, etc. The emission and speciation factors are being generated in conjunction with the development of the UNECE-EMEP Emission Inventory Guidebook.
- The large combustion sector is not a significant contributor to global VOC emissions. Information on locations and activity rates of large combustion sources will be shared with the GEIA Committee on Anthropogenic Emissions of SO_2 and NO_x. Activity rates and emission factors for small boilers outside North America have been prepared within the framework of the LOTUS and EDGAR projects from statistic of IEA and the UN.
- The waste sector is currently ill-defined; emissions from this sector will not be included in Version 1 of the VOC inventory.
- VOC emissions from aircraft are being compiled by a project under NASA support.
- Distribution of emissions estimated on a country basis will be accomplished via the use of global population data gridded to 1° resolution.

Version 1 of the GEIA global inventory of anthropogenic emissions of VOCs is scheduled to be released to the scientific community in 1995.

ACKNOWLEDGMENTS

This research is part of the Global Emissions Inventory Activity (GEIA), a component of the International Global Atmospheric Chemistry (IGAC) Core Project of the International Geosphere-Biosphere Program (IGBP). C.M. Benkovitz performed this research under the auspices of the Atmospheric Chemistry Project of the NOAA Climate and Global Change Program and of a grant from the Chemical Manufacturers Association. The work of J.J.M. Berdowski and C. Veldt is sponsored by the Dutch Nation Research Program on Global Air Pollution and Climate Change and by the Netherlands Organization for Applied Scientific Research TNO.

REFERENCES

1. Galbally, I.; *The International Global Atmospheric Chemistry Programme*, pp Pages.
2. Graedel, T.E., *et al.* Glob. Biogeoc. Cycles **1993** *7* 1-26.
3. Veldt, C., "VOC: A Little Learning is a Dangerous Thing", in *TNO/EURASAP Workshop on the Reliability of VOC Emissions Data Bases*, TNO Institute of Environmental Sciences: Delft, The Netherlands, Year; pp 115-136.
4. Berdowski, J.J.M., "Anthropogenic NMVOC Emissions - GEIA Status Report", in *5th GEIA Workshop*, Fuji Yoshida, Japan, Year;
5. U.S. Environmental Protection Agency; *Regional Interim Emissions Inventories (1987-1991)*, EPA-454/R-93-021, 1993; pp Pages.
6. Modica, L.G.Dulleba, D.R.; *The 1985 NAPAP Emissions Inventory: Development of Spatial Allocation Factors*, Rep. EPA-600/7-89-010b, 1990; pp Pages.
7. Shareef, G.S.Bravo, L.A.; *Air Emissions Species Manual - Volume 1, Volatile Organic Compounds Species Profiles*, EPA-450/2-90-0001a, 1993; pp Pages.
8. Samaras, Z.Veldt, C., "Global Emissions from Road Transport", in *Third GEIA Workshop on Global Emissions Inventories*, National Institute of Public Health and Environmental Protection, Bilthoven, The Netherlands, Report 481507002: Amersfoort, The Netherlands, Year; pp 1-6.
9. Samaras, Z.Zierock, K.-H.; *CORINAIR Working Group on Emission Factors for Calculating 1985 Emissions from Road Traffic. Volume 2: COPERT - Computer Programme to Calculate Emissions from Road Traffic. Final Report to EC, Study Contracts B 6611-54-88 and B 6611-55-88*, EUR No. 12260 EN, 1989; pp Pages.
10. Samaras, Z.Zierock, K.-H.; *CORINAIR Working Group on Emission Factors for Calculating 1985 Emissions from Road Traffic. Volume 3: Results Obtained with COPERT. Final Report to EC, Study Contracts B 6611-54-88 and B 6611-55-88*, EUR No. 12260 EN, 1989; pp Pages.
11. Eggleston, H.S., Gorissen, N., Joumard, R., Rijkeboeer, R.C., Samaras, Z. Zierock, K.-H.; *CORINAIR Working Group on Emission Factors for Calculating 1985 Emissions from Road Traffic. Volume 1: Methodology and Emission Factors. Final Report to EC, Study Contract No. 88/8611/0067*, EUR No. 12260 EN, 1989; pp Pages.
12. Samaras, Z.C.Zachariadis, T.I., "A Comparative Assessment of Methodologies to Estimate Motor Vehicle Emissions (MOBILE - COPERT)", in *International Conference "The Emission Inventory: Perception and Reality"*, Pasadena, CA, Year;
13. Veldt, C.Most, P.F.J.v.d.; *Emission Factors - VOC from Internal Combustion Engines (in Dutch)*, Publikatiereeks Emissieregistratie Nr. 10, 1993; pp Pages.
14. Radian Corporation; *Global Emissions of Methane from Petroleum Sources*, 1992; pp Pages.
15. Berdowski, J.J.M.Zandveld, P.; *LOTOS-90. The Emission Database for the Long Term Ozone Simulation Model for 1990*, R94/... (in preparation), 1994; pp Pages.
16. U.S. Environmental Protection Agency; *Compilation of Air Pollutant Emission Factors*, AP-42, 1985; pp Pages.
17. McInnes, G.; *UNECE-EMEP Guidebook on Emissions Inventories*, (in preparation), 1995; pp Pages.

18. Baars, H.P., Berdowski, J.J.M. Veldt, C.; *Preliminary Study on a Global Emissions Database (EDGAR)*, R91/136, 1991; pp Pages.
19. Berdowski, J.J.M. Change 1992 10 5-6.
20. Olivier, J.G.J., Bouwman, A.F., Maas, C.W.M.v.d. Berdowski, J.J.M. Environmental Monitoring and Assessment (accepted) 1994
21. Oil and Gas Journal Energy Database, *Worldwide Petrochemical Survey*. 1990, Penwell Publishing Co.: Tulsa, OK.
22. Stolarski, R.S.Wesoky, H.L.; *The Atmospheric Effects of Stratospheric Aircraft: A Third Program Report*, NASA Reference Publication 1313, 1993; pp Pages.
23. Andreae, M.O.; *Biomass Burning: Its History, Use and Distribution and its Impact on Environmental Quality and Global Climate;* in *Global Biomass Burning: Atmospheric, Climatic and Biospheric Implications;* MIT Press, Cambridge, MA, 1991; pp. 3.

Table 1. GEIA committees, January 1994.

Species	Source Type	Temporal Resolution	Director
SO_2	Anthropogenic & Natural	A	J.M. Pacyna, NOR C.M. Benkovitz, USA
		S	T. Scholtz, Can
Reduced S	Natural	M	L. Tarrason, NOR
NO_x	Anthropogenic	A	J.M. Pacyna, NOR C.M. Benkovitz, USA
		S	T. Scholtz, Can
CO_2	Anthropogenic	A	G. Marland, USA
CO	Anthropogenic	A	J. Logan, USA
NH_3, N_2O	Natural	S	A.F. Bouwman, NL
CH_4	Anthropogenic	A	N. Roulet, Can E. Matthews, USA
VOC	Anthropogenic	S	J. Berdowski, NL
VOC	Natural[2]	2 seasons	N. Hewitt, UK A. Guenther, USA
CFCs	Anthropogenic	A	D. Cunnold, USA
Pb	Anthropogenic	A	J. Pacyna, NOR
Hg	Anthropogenic	A	J. Pacyna, NOR
Radioisotopes	Natural	A	M. Kritz, USA
Biomass Burning	N/A	S	B.J. Stocks, CAN J.S. Levine, USA J. Goldammer, GER
Aircraft[1]	Anthropogenic	A	D. Wuebbles, USA
Data Management	N/A	N/A	P. Middleton, USA

A=Annual S=Seasonal M=Monthly N/A=Not applicable

1 Currently includes NO_x and VOCs. VOCs to be speciated (groupings to be determined).

2 Speciated into isoprene, terpenes, short-lived VOCs and long-lived VOCs.

Table 2. Source classification selected for reporting anthropogenic emissions of VOCs. Listed in order of decreasing contribution to global emissions.

Small scale combustion
Mobile sources
Solvent use
Fossil fuel production and distribution
Chemical industry
Fossil fuel combustion
Other.

Table 3. Chemical groups selected for reporting of anthropogenic emissions of VOCs. Each name represents an individual group; groups are listed with others of similar structure.

Ethane	Benzene	Formaldehyde	Methylene chloride
Propane	Toluene	Acetaldehyde	1,1,1-Trichloroethane
N-butane	o-Xylene	Saturated aldehydes C≥3	Trichloro-ethylene
Iso-butane	mp-Xylene	Unsaturated aldehydes	Tetrachloroethylene
N-pentane	Ethylbenzene		Non-speciated Cl-hydrocarbons
Iso-pentane	Styrene		
Hexanes	Trimethylbenzenes		
Heptanes	Other alkyl aromates C=9	Acetone	
Alkanes C≥8	Alkylaromates C≥10	Methylethylketone	Other HC compounds
Non-speciated alkanes	Non-speciated aromates	Non-speciated ketones	Other HCO compounds
		Methylisobutylketone	
Ethylene			
Acetylene		Formic Acid	
Propylene	Methylalcohol	Acetid Acid	
1-Alkenes C=4	Ethylalcohol	Non-speciated carbonic acids	
2-Alkenes C=4	Isopropylalcohol		
1-Alkenes C=5	Butylalcohols		
2-Alkenes C=5	Non-speciated alcohols		
Alkenes C≥6			
Non-speciated alkenes			

Table 4. Summary of the progress of the GEIA global inventory of anthropogenic emissions of VOCs. Present estimates for global annual emissions for 1990 (Tg/year) are included.

Sources	Global Emissions	Activity	EF	Profile	Emission	Geographic Unit	
Small scale combustion	48.6	+/-	+	+	+/-	-	Country
Mobile sources	33.2	+/-	+	+	+	+/-	Country
Solvent use	20.0	+/-	+/-	+/-	+/-	+/-	Country
Oil & gas production	18.0	+/-	+	+	_	+/-	PS
Oil refining	4.4	+	+	+	+	+	PS
Gasoline distribution	2.5	+	+	+	+	+	Country
Chemical bulk products	0.8	+	+/-	+/-	+/-	+	PS
Large combustion plants	0.4	+/-	+	+	+/-	-	PS
Waste incineration	(8.0)[1]	-	-	-	-	-	
Miscellaneous	(10.0)[2]	-	-	-	-	-	
Large biomass burning	(24.0)[3]	n	n	n	n	n	
Aircraft	(0.2)[4]	n	n	n	n	n	

+ = first order approach finalized; +/- = first order approach finalized > 75%; - = first order approach not started or finalized < 75%.
PS = Point Sources. n = not being addressed in this work.
[1,2] Derived from Berdowski [4]
[3] Estimate from Andreae [23] as Tg carbon yr^{-1}.
[4] Totals from the 1990 scenario in Stolarski [22] as Tg carbon yr^{-1}.

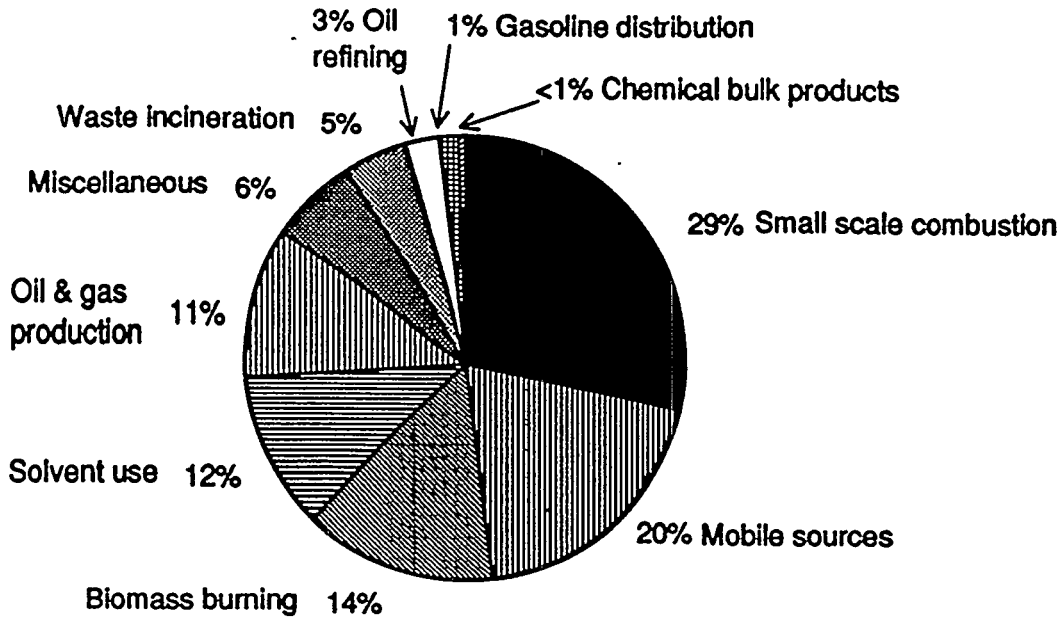

Figure 1. Preliminary estimates of the relative contributions of source types to the global total VOC emissions in 1990.