NATIONAL INSTITUTE OF PUBLIC HEALTH AND THE ENVIRONMENT
BILTHOVEN, THE NETHERLANDS

WIMEK report / RIVM report nr. 728001 002

Greenhouse Gas Emission Accounting

Preliminary study as input to a joint International IPCC Expert Meeting/CKO-CCB Workshop on Comparison of Top-down versus Bottom-up Emission Estimates

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September 1997

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This study was commissioned to LUW-WIMEK by the Directorate-General for Environment, Department of Air and Energy of the Dutch Ministry of Housing, Spatial Planning and the Environment, project number 97140031.
This study was commissioned to RIVM by the Directorate-General for Environment, Department of Air and Energy of the Dutch Ministry of Housing, Spatial Planning and the Environment, project number 728001.

This project has also been carried out in the framework of the Dutch National Research Programme on Global Air Pollution and Climate Change (NRP II, project number 954222).
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 Programme (IGBP).

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Summary

This report has been written as input to an IPCC Expert Meeting on "A comparison of Top-down versus Bottom-up Emission Estimates" to be held on 5-6 November 1997 at RIVM, Bilthoven, the Netherlands. This meeting is part of the IPCC/OECD/IEA workplan to improve the IPCC inventory methodology. For carbon dioxide, methane and nitrous oxide *bottom-up* data from the official national inventories (National Communications) were compared with data from EDGAR (Emission Database for Global Atmospheric Research), and *top-down* emission estimates based on results of dispersion- and climate models using measured concentrations of greenhouse gases in the atmosphere.

The purpose of this preliminary study was to investigate the possibilities of comparing different types of emissions inventories; to develop a methodology for this comparison and to use the results in an analysis to identify areas for improvement of the IPCC methodology.
The main conclusion is that an international program to review and evaluate national inventories of greenhouse gases is useful while:

- the use of both *bottom-up* and *top-down* emission data improves the scientific understanding of the global and regional budgets, increases the quality of emission data and improves methodologies to compile national emissions inventories;
- the exchange, review and comparison of data promotes dialogue, the sharing of data and consensus about the data among scientists and policy-makers;
- the use of atmospheric measurements of greenhouse gases together with models may appear to be an objective tool of monitoring progress towards national and global emission reduction goals and may develop into a verification mechanism.

The difference between EDGAR estimates of global CO_2, CH_4 and N_2O emissions due to anthropogenic activities and a central *top-down* estimate are < 13%, 35% and about a factor of 2 respectively. For CO_2 it is not possible at this moment to estimate fossil fuel emissions derived from atmospheric measurements and global carbon dioxide budget calculations with more accuracy then those based on *bottom-up* emission inventories. For CH_4, a global or even a zonal comparison of *bottom-up* emission inventories with *top-down* results of transport models is possible and a reduction of uncertainty for specific sources may be achieved. Uncertainties of N_2O emissions are that large that we may expect that both *top-down* and *bottom-up* emission estimates may benefit from results of a careful comparison of these emission estimates.

The comparison of national inventories with EDGAR data has identified areas for future improvement in the IPCC Guidelines. For CO_2: the landuse sector, agriculture, and biofuels combustion; for CH_4: emission from agriculture, biofuel combustion, landuse, landfills and waste water treatment; and for N_2O: industrial processes, biomass burning, landuse change and waste treatment. Summarised: except for the emission of greenhouse gasses by energy use and the burning of fossil fuel, the emissions from other sectors are still surrounded with rather large uncertainties.

Samenvatting

Dit rapport is geschreven ter voorbereiding van een Internationale IPCC-bijeenkomst van experts met "De vergelijking van Bottom-up en Top-down schattingen van broeikasgasemissies" als onderwerp. Deze bijeenkomst wordt op 5 en 6 November 1997 gehouden bij het RIVM in Bilthoven. Voor de broeikasgassen CO_2, CH_4 en N_2O zijn *bottom-up* gegevens van officiële nationale inventarisaties (de z.g. National Communications) vergeleken met gegevens van EDGAR (Emission Database for Global Atmospheric Research) en *top-down* resultaten van emissieschattingen op basis van verspreidings- en klimaatmodellen die gebruik hebben gemaakt van gemeten concentraties in de atmosfeer.

Het doel van deze voorstudie was om de mogelijkheden te onderzoeken hoe verschillende typen van emissie-inventarisaties met elkaar vergeleken kunnen worden; een methode te ontwikkelen om de vergelijkingen uit te voeren en om op basis van een analyse van de verschillen mogelijke gebieden voor verbeteringen van de IPCC-methode voor emissieschattingen in kaart te brengen. De belangrijkste conclusie is dat een internationaal programma voor review en evaluatie van nationale inventarisaties van broeikasgassen nuttig is omdat:

- het gebruik van zowel *bottom-up* als *top-down* emissiegegevens het wetenschappelijk inzicht in mondiale en regionale budgetten vergroot, evenals de kwaliteit van de gegevens, en de methodiek verbetert om nationale emissieinventarisaties samen te stellen;
- uitwisseling, review en vergelijking van data de dialoog en het bereiken van overeenstemming over de gegevens tussen wetenschappers en beleidsmakers stimuleert;
- het gebruik van metingen van concentraties van broeikasgassen in de atmosfeer zich kan ontwikkelen tot een objectief instrument om voortgang in nationale en mondiale emissiereductie doelstellingen vast te stellen.

De verschillen tussen EDGAR-schattingen van mondiale emissies van CO_2, CH_4 en N_2O ten gevolge van menselijke activiteiten en een centrale *top-down* schatting zijn resp. <13%, 35% en ongeveer een factor 2. Het is op dit moment niet mogelijk om voor CO_2 met een *top-down* analyse gebruik makend van atmosferische metingen de CO_2 emissies van fossiele brandstoffen nauwkeuriger te schatten dan met *bottom-up* methoden. Voor CH_4 is een mondiale en zonale vergelijking van *bottom-up* en *top-down* resultaten van verspreidingsmodellen mogelijk en een reductie van de onzekerheid voor specifieke bronnen is haalbaar. De onzekerheden in de N_2O-emissies zijn zo groot dat verwacht mag worden dat *top-down* en *bottom-up* emissie-schattingen beiden kunnen profiteren van een zorgvuldige analyse van de resultaten.

De vergelijking van nationale inventarisaties met EDGAR gegevens heeft terreinen voor verdere verbetering van *IPCC Guidelines* in kaart gebracht. Voor CO_2 zijn dat: landgebruik, landbouw en verbranding van biobrandstoffen; voor CH_4 zijn dat: landbouw, biobrandstoffen, landgebruik, afvalstorts en afvalwaterbehandeling; en voor N_2O: industriële processen, biomassaverbranding, veranderingen in landgebruik en de behandeling van afval. Samengevat: behalve de broeikasgasemissies door verbranding van fossiele brandstoffen ten behoeve van energiegebruik, zijn de emissies van andere sectoren nog met tamelijk grote onzekerheden omgeven.

Executive summary

This study will serve as input for an IPCC Expert Meeting on national and global greenhouse gas inventories to be held on 5-6 November 1997 at RIVM, Bilthoven, the Netherlands. For carbon dioxide, methane and nitrous oxide, *bottom-up* data from the official national inventories (National Communications) were compared with data from EDGAR (Emission Database for Global Atmospheric Research) and *top-down* results from dispersion models using measured concentrations of greenhouse gases in the atmosphere.

The purpose of this preliminary study was, firstly, to investigate the possibilities of comparing different types of emission inventories e.g. National Communications, science-based *bottom-up* emission inventories and *top-down* budgets derived from atmospheric modelling. Secondly the study aimed at developing a methodology for this comparison and, thirdly, at the use of the results in an analysis to identify areas for improving the IPCC methodology. This comparison will also assist in maintaining the two tracks for establishing emissions i.e. using statistical data to compile emission inventories and using atmospheric data and dispersion models to compile budgets of greenhouse gases together. This may increase the credibility of the inventories, facilitating development of climate policy. Both national inventories and global budgets are characterised by uncertainties. In national inventories these uncertainties occur in field measurements used to determine emission factors, upscaling of emissions to the national level and in the statistical activity data of the national economies. Uncertainties in global budgets come about from uncertainties in processes determining the terms of the budget: the emission and loss terms, the flows between the reservoirs and the magnitude of the reservoirs themselves.

This preliminary study in which only emission data were compared forms a part of an international effort to improve the IPCC inventory methodology (see IPCC/OECD/IEA workplan, July 1997). In this preliminary study only emission data were compared. In a second phase of this study to start in August 1997, an in-depth review of emissions, emission factors and activity data will take place.

The preliminary study, based on readily available information has followed the steps below:

1. The information was inventoried and made available for comparison.
2. An analysis of this information was carried out and a standard for presentation of emission data was developed to enable comparison of *bottom-up* with *top-down* estimates.
3. National inventories submitted to the UNFCCC Secretariat, produced using the IPCC Guidelines, were compared with EDGAR estimates and results of global climate models.
4. The results of the comparison were presented and a first analysis of the differences was carried out.
5. Where differences were encountered, criteria for selection of case studies were formulated.
6. The uncertainties were investigated.
7. The draft report was reviewed by experts as part of the working plan.

Main conclusions

An international programme to review and evaluate national inventories of greenhouse gases is useful for several reasons:

- The *bottom-up* comparison of greenhouse gas emission inventories (in-depth studies of emission inventories) is a powerful method for improvement of plausibility, consistency, accuracy, and appropriateness of IPCC and other national methodologies. It provides a discussion platform and identifies areas for future improvement of the IPCC methodologies.
- Exchange, review and comparison of data promotes dialogue, data sharing and consensus on the data among scientists and policy-makers.
- Using both *bottom-up* and *top-down* types of emission data improves the scientific understanding of the global total budgets, increases the quality of emission data and refines methodologies to compile national emissions inventories, thereby increasing confidence and credibility in the emissions inventory process.
- Evaluation of different *bottom-up* and *top-down* emission data sets increases the credibility of emission inventories, which may facilitate development of climate policy and measures.
- The use of atmospheric measurements of greenhouse gases together with models appears to be an objective tool for monitoring progress in attaining national and global emission reduction goals and may develop into a verification mechanism.

Top-down

It is possible to evaluate greenhouse gas emissions on a lesser than global scale e.g. regional or national, using direct measurements and atmospheric models if the required accurate measurements are available. However, at the moment measurement data of greenhouse gas concentrations come from a limited number of remote background stations. Measurements on small spatial and temporal scales, notably in emission source regions, are lacking for most world regions. For CO_2 ,only the global budget can be evaluated. No accurate estimation of anthropogenic sources at zonal or regional level can be made on the basis of the existing atmospheric measurements due to the overriding influence of the large fluxes of CO_2 between the different reservoirs (atmosphere, ocean and biosphere) which also vary in time. For CH_4 global, zonally averaged and several regional emissions can be estimated on the basis of measurements and model calculations. At the moment the regional detail is only feasible for Europe. For N_2O only global and (limited) zonally averaged emissions can be estimated or evaluated at the moment.

Bottom-up

National emission inventories, to be prepared by all countries signing the Framework Convention of Climate Change (FCCC) as part of their National Communications, are now available for many industrialised countries. However, a substantial number of National Communications from developing countries are still lacking. Global *bottom-up* emission data are collected by using the EDGAR database. About 65% of the global CO_2 emission, 55% of the global CH_4 emission and less than 30% of the global N_2O emissions are estimated to have been reported in the first National Communications submitted in 1994 and other county studies. Source categories of IPCC Guidelines and EDGAR have been made compatible. Results of a *bottom-up* versus *bottom-up* comparison of sectoral emissions data from National Communications and EDGAR data have shown that occasional substantial differences for specific greenhouse gases between sectors and countries. Analysis has revealed *bottom-up* comparison of greenhouse gas emission inventories (scoping and in-depth studies of emissions inventories) to be a powerful method for improving plausibility, consistency, accuracy, and suitability

of IPCC Guidelines and other national methods to estimate emissions. As it provides insight into the transparency of the inventory calculations, it supports scientific dialogue and provides a discussion platform for future updates of the IPCC methodology.

Comparison of Top-down versus Bottom-up emission estimates

Various models and datasets are available for a *top-down* versus *bottom-up* comparison of emission inventories. Models used in the Netherlands are the EUROS, LOTOS, and COMET models for Europe; the IMAGE, DIALOOG, TM2 and MOGUNTIA model for the world. In this report we used results from the literature of these models and of various other global climate models. Datasets which contain global measurement data are the CDIAC and WMO-WCDCGG databases. The NOP-CH_4 dataset at RIVM contains CH_4 concentration data of Europe with high spatial and temporal resolution. The most important datasets on national emissions carrying sectoral detail and information on emission factors and activity data are the National Communications database of the UNFCCC Secretariat, the IEA database on energy use, the FAO database on agriculture and forestry, and the EDGAR database of RIVM/TNO on emissions, per country and per gridcell on 1^0x1^0 longitude and latitude. In order to be able to compare *bottom-up* and *top-down* results it is necessary to make these datasets compatible. In the following table the initial differences are given.

Table ES.1 Differences between Bottom-up and Top-down methods

	Bottom-up inventories	Top-down models
Gas	CO_2 CH_4 N_2O	CO_2 CH_4 N_2O
Emission	per sector	per source category
Spatial scale	country	global/zonal/regional/grid
Time scale	base year	any period from hour to year

Evaluation of the greenhouse gas budget using information from measurements and atmospheric transport models, can be employed to test the plausibility of emission inventories. Measurements of radioactive and stable isotopes can be used to provide information on specific source categories, even on a regional scale. However, the capability of the *top-down* method to derive detailed information on emissions on a small spatial and temporal scale is limited at the moment (Table ES.2.)

Table ES.2 Maximum attainable temporal and spatial resolution for inverse modelling

	CO_2	CH_4	N_2O
Spatial resolution	global (zonal)	global, zonal or 1^0x1^0	global and zonal
Temporal resolution	5-10 years average	year (1990)	year (1990)
Period	5 years	year	year
Source categories	fossil fuel land-use change vegetation	anthropogenic natural	anthropogenic natural

National inventories (National Communications) are available for most countries for 1990 and 1995. Since EDGAR 2.0 emission data have 1990 as a base year a comparison with model results is recommended for those years. For CO_2 ,however, deforestation and land cover and land use data are available only for 5-10 year averages and the exchange between atmosphere, biosphere and ocean fluctuates substantially from year to year. Therefore is recommended to use only 5-10 year periods to compare *bottom-up* and *top-down* data.

In developing a standard methodology for *top-down/bottom-up* comparisons national totals per sector of any dataset, e.g. official UNFCCC submissions, need to be distributed over a global grid, thereby providing means to compare results of *bottom-up* estimates with those of a *top-down* analysis using global dispersion models. In this report the unique facility of the

EDGAR software was used to convert country totals per sector into global 2-D distributions and 1-D latitudinal (zonal) distributions, which can then be compared with zonal emission estimates resulting from inverse modelling.

Carbon dioxide

It was concluded for carbon dioxide that global carbon cycle models ignore short-term perturbations due to fluctuations in the climate system. It is therefore not possible to estimate yearly fossil fuel emissions derived from atmospheric measurements and global carbon dioxide budget calculations with more accuracy than the emissions based on *bottom-up* emission inventories. The main sources of uncertainty in establishing the CO_2 budget are the poor world coverage offered by the measuring stations, the yearly variations in sources and sinks, the still unsolved problem of magnitude of oceanic and biospheric sink leading to different assumptions on the budget by modellers. Yearly variations in the CO_2 budget are much larger than the yearly variations in the fossil fuel emissions. Therefore no accurate validation of national emission inventories is possible with budget studies. The uncertainty in the estimated CO_2 emission due to anthropogenic activities derived from atmospheric measurements is 25-35% and 10% or less in the *bottom-up* emission estimates. EDGAR (*bottom-up*) global CO_2 emissions due to use of fossil fuel are well within the range of *top-down* global estimates (difference < 13% between central estimates). Differences of CO_2 emissions due to land-use changes are larger (up to a factor of 3). Besides a global comparison of CO_2 emissions some additional spatial detail may be retained taking the indications of an additional biospheric sink in the Northern Hemisphere into account. The comparison of national inventories with EDGAR data has identified areas for future improvement in the IPCC Guidelines for CO_2, namely land use, agriculture, and biofuel combustion. Differences between national inventories and EDGAR data will be analysed further in the second phase of the project.

Methane

The short-term variations in the atmospheric increase of the methane concentration are in the order of magnitude of the yearly increase. The uncertainty of global methane emissions due to human activities derived from atmospheric measurements is in the order of 30%. The uncertainty in the sources estimated with *bottom-up* methods is about the same, some being higher and others lower (25-35%). Therefore a global or even a zonal comparison of *bottom-up* emission inventories with *top-down* results from transport models is possible and a reduction in uncertainty for specific sources may be achieved. EDGAR *bottom-up* estimates of global CH_4 emissions due to anthropogenic activities are well within the range of 35% from a central *top-down* estimate. A comparison of *top-down* with *bottom-up* emission inventories is possible on a much smaller - regional - scale for north-west Europe thanks to long-term high resolution measuring data being available for this area. Using data of one measuring station, Cabauw in the Netherlands, at a height of 200 m above sea level, it is possible to evaluate the national inventories of surrounding countries up to a distance of 500 km. Sectoral detail is less developed than in the inventories, but it is possible to evaluate the totals. The comparison of national inventories with EDGAR data has identified areas for possible future improvement of the IPCC Guidelines, especially the methodology for estimating methane from agriculture, biofuel combustion, land use, landfills and waste-water treatment.

Nitrous Oxide

Top-down inverse modelling exercises used to derive global and zonal budgets have been carried out. In addition, global 3-dimensional model calculations were used to simulate atmospheric concentration fields where gridded N_2O emissions served as input. Uncertainties are high, both in the inventories and in the emission estimates based on results of global

models. One of the reasons is that the stratospheric removal cannot be accurately quantified as yet. In addition, most of the atmospheric N_2O is of biogenic origin and from biogenic sources and shows especially in soils, large variability, both spatial and temporal. The least uncertain estimate is the trend in the atmospheric increase of nitrous oxide (4-5 Tg N/year). Using this figure and a central estimate for the magnitude of the stratospheric sink (12-13 Tg N/year) it could be shown by a *top-down* analysis that a large part (50%) of the global anthropogenic N_2O emission, derived from *bottom-up* emission estimates, was missing. Based on this information, a careful *bottom-up* analysis of all possible sources led to the identification of a new source of N_2O emissions: cattle and feedlots. This is a clear example of how a *top-down* estimate can improve *bottom-up* emission estimates. Nevertheless uncertainties are still large up to a factor of 2, so large in fact that we can expect both *top-down* and *bottom-up* emission estimates to benefit from results of a careful comparison of emission estimates. On the basis of present models and datasets we expect a comparison of national inventories with global model results to only be useful on a global or zonal scale. The global totals from the EDGAR database for N_2O emissions due to anthropogenic activities are much higher than the National Communications total (factor of 3), partly because a large number of countries did not submit a National Communication. However, totals are still much lower than the IPCC *bottom-up* total (factor of 2) based on the *Revised IPCC Guidelines* and *top-down* global estimates. A first analysis showed differences between EDGAR and National Communications to be large for most sources. The second phase of the study will further analyse these differences. For future improvement of the IPCC Guidelines it is recommended to focus on industrial processes, biomass burning, land use change and waste treatment.

Recommendations

The effectiveness and accuracy of the **Top-down** monitoring and modelling techniques can be increased through the following research activities:

1. Expansion of the network of concentration measuring sites, both background locations and source regions.
2. Measuring meteorological parameters and mixing-layer height simultaneous with concentration measurements.
3. Greater frequency of measurements at sites with seasonally varying source strengths.
4. Greater spatial coverage and accuracy of isotope measurements.
5. Establishment of international primary standards, and improved maintenance and calibration of standards, for individual laboratories and internationally.
6. Standardised collection and cataloguing of measurement data at international and regional centres.
7. Effective availability of measuring data through web sites and FTP sites is very important. The databases at CDIAC (Oak Ridge) and WMO-WCDCGG (Tokyo) are therefore important initiatives which should be supported.
8. Support the development of inverse modelling techniques such as Kalmanfiltering and the application of adjoint models.
9. Increase the temporal resolution of models by making them suitable for actual meteorological information.
10. Improvement of time profiles (seasonality) of emission sources in modelling.

The completeness and consistency of **Bottom-up** national inventories can be increased by the following activities:

1. Comparison of aggregated emission factors between national inventories or between different years in one inventory may identify deviating emission factors; likewise, comparison of sectoral emission strength indicators may indicate apparent typing or calculation errors.
2. Comparison of national inventories with global sectoral emission inventories constructed from international statistics and consistent sets of emission factors may reveal missing sources and identify areas with large deviating emission factors.
3. In-depth review of deviating emission factors should be done to check that they are country specific and not biased compared to similar countries.
4. Global sectoral inventories can be used to estimate the main sources in non-reporting countries as well as their share in the global total.
5. Compilation of global emission databases such as EDGAR and GEIA are important as scientific reference databases to perform quick checks on completeness and strength class of sources and for more detailed investigation of large deviations when identified in the comparisons mentioned above.
6. To be useful for comparison with national inventories, these global inventories should include emissions per country and distinguish key source sectors.
7. Maintenance and update, in particular of emission factors, in scientific databases are important for improvement of the consistency between countries.
8. Carry out measurements near sources to accurately determine specific emission factors

For **comparison of Top-down and Bottom-up** emission estimates the following aspects need to be considered to increase the accuracy of comparison results:

1. For an independent check of national inventories with *top-down* estimates from inverse modelling, often additional information is required on the temporal variation within a year (e.g. seasonality) and on the spatial distribution in a standardised way (e.g. on grid [1D]or latitudinal bands [1D]).
2. Inverse models require these 'fingerprints in space and time' of sectoral emissions as *a priori* input and also often generate their results in this spatial format.
3. For a proper evaluation by *top-down* models it is also important to have a fair description of the natural sources in the target area.
4. The uncertainty in the estimate of annual sectoral emissions of a country is important additional *a priori* information for inverse model calculations.
5. Facilities like the EDGAR system are required to consistently convert national emissions into 1D or 2D distributions as a bridge between national, annual inventories and atmospheric chemistry models.
6. In the future a Top-down and Bottom-up comparison of emission inventories should also include other greenhouse gases.

Glossary

Activity data	Statistical information on the economy of a country, used to calculate emissions
ALE/GAGE	Atmospheric lifetime experiment/Global Atmospheric Greenhouse Gas Emissions.
ANU	Australian National University
BU	Bottom-up analysis
Budget	Stock, fluxes and loss terms of greenhouse gases in the troposphere.
CCB	Research Programme on Climate Change and Biosphere, WIMEK Wageningen, The Netherlands.
CDIAC	Carbon Dioxide Information Analysis Centre at Oak Ridge National Lab.
CH_4	Methane.
CKO	Centre for Climate Research, KNMI,RUU,RIVM Bilthoven, the Netherlands.
CMDL	Carbon Dioxide Monitoring and Diagnostics Laboratory.
CMKW	Centre for Environment and Climate Studies, Wageningen.
CO_2	Carbon dioxide
CoP	Conference of the Parties to the Climate Convention
CORINAIR	Inventory Air of European Union at EEA
CSIRO	Commonwealth Scientific and Industrial Research Organisation, Melbourne.
ECN	Energy Research Foundation, Petten, the Netherlands.
EDGAR	Emission Database for Atmospheric Research (RIVM/TNO) at RIVM, Version 2.0, Bilthoven.
EEA	European Environment Agency, Copenhagen, Denmark.
EEur	Eastern Europe
Emission factor	Emission per unit product per year or per unit area per year.
EUROS	Eulerian European dispersion model for atmospheric research.
EU15	European Union with 15 member countries.
FTP	File transfer protocol.
GHG	Greenhouse gas.
Global inventory	Estimate of total anthropogenic (and natural) emission of a greenhouse gas
Greenhouse gas	Trace gas that traps part of the long wave back radiation of the earth.
IEA	International Energy Agency.
IGAC	International Global Atmospheric Chemistry Project of IGBP.
IGBP	International Geosphere - Biosphere Project.
IMAGE 2	Integrated model to assess the greenhouse effect, Version 2, developed at RIVM
IPCC	Intergovernmental Panel on Climate Change
Isotope study	Study in the origin of an emission using the isotopic composition of a gas in the atmosphere.
GEIA	Global Emission Inventory Activity of IGAC.
GWP	Global warming potential. Over 100 years: $CO_2 = 1$, $CH_4 = 21$, $N_2O = 310$.
Kalman filtering	Statistical technique developed by Kalman.
EKF	Extended Kalman filtering
KEMA	National Research Institute of the Dutch Utilities.
Lifetime	The residence time in the atmosphere of a quantity of a greenhouse gas.
LTO cycle	Landing and Take Off cycle of aircraft up to about 1 km or 3000 feet.
LUW	Agricultural University Wageningen.
MIT	Massachusetts Institute of Technology.
National Communication	National report to the Climate Convention Secretariat to report progress on the climate policy of a country, including national inventories.
NatComm	National Communication
NC	National Communication
National inventory	Estimate of national emissions of greenhouse gases in a particular year, as reported in the National Communication.
NMVOC	Non-Methane Volatile Organic Carbon
NOAA	National Oceanic and Atmospheric Administration.
N_2O	Nitrous oxide.
N_2O-N	Nitrous oxide expressed as Nitrogen.

OECD	Organisation for Economic Co-operation and Development.
OLADE	Organizacion Latino Americana De Energia.
ORNL	Oak Ridge National Laboratory USA.
ppbv	part per billion by volume
ppmv	part per million by volume
Reservoir	Pool of a specific compound, e.g. carbon
RIVM	National Institute of Public Health and the Environment, Bilthoven.
RoW	Rest of the World
SEI Boston	Stockholm Environment Institute, branch located in Boston USA.
TD	Top-down analysis
Trajectory analysis	Calculation of remote emissions of a greenhouse gas using concentration measurements and information on the origin of the air masses (wind speed, wind path, height of the mixing layer).
TM2	Transport Model, version 2, of Max Planck Meteorology Institute, Hamburg.
TNO	Netherlands Organisation for Applied Scientific Research, Apeldoorn.
UN	United Nations
UNFCCC	United Nations Framework Convention on Climate Change.
USCSP	US Country Studies Programme.
VOC	Volatile Organic Carbons, emissions responsible for smog formation.
VROM	Ministry of Housing, Physical Planning and the Environment, The Hague.
V-PDB	Vienna PeeDee Belemnite, a standard marine limestone for isotope studies.
V-SMOW	Vienna Standard Marine Ocean Water, a standard used in isotope studies.
WMO-GAW	World Meteorological Organisation-Global Atmospheric Web.
1-D model	One dimensional model, box model.
2-D model	Two dimensional model: latitude, height; or longitude, latitude
3-D model	Three dimensional model: longitude, latitude, height.

Figure captions

Table captions

1. Introduction

1.1. Background

Under the United Nations Framework Convention on Climate Change (UNFCCC) countries have agreed to stabilise greenhouse gas concentrations in the atmosphere at a level that would prevent dangerous anthropogenic interference with the climate system. The greenhouse gas emissions, projections for 2000, (and beyond) and policy initiatives for reduction of emissions are reported in National Communications to the Climate Convention Secretariat. A common reporting format is used to allow a compilation and synthesis of emission data to be made regularly by the Climate Convention Secretariat. In this study, national inventory data are compared with global inventories and global atmospheric budget studies, in order to identify target areas for future improvements of the IPCC greenhouse gas inventory methodology.

1.1.1 Climate Convention

The United Nations Framework Convention on Climate Change (UNFCCC), was signed in Rio de Janeiro in 1992 by 154 heads of states and later ratified by the same countries. The Convention calls for the stabilisation of greenhouse gas concentrations in the atmosphere at a level that would prevent dangerous anthropogenic interference with the climate system. Such a level is to be achieved within a time frame sufficient to allow ecosystems to adapt naturally to climate change, to ensure that food production is not threatened and to enable economic development to proceed in a sustainable manner. The first Conference of the Parties in Berlin decided that a common methodology was needed for the yearly submission of data on sources and sinks of greenhouse gases. The Guidelines for National Greenhouse Gas Inventories, developed and published by the IPCC (IPCC/OECD/IEA, 1995, three volumes), were adopted as this common methodology for reporting emissions and sinks to the Climate Convention Secretariat. The OECD countries and Eastern Europe (as listed in Annex 1 to the Convention) have already reported emissions for 1988/1990 to 1995. Other countries are expected to do so by April 1997. The European Environment Agency has summarised the 1994 and 1995 data for Europe (EEA, 1997). A third review and synthesis of data by the Climate Convention Secretariat has started and will be based on all Second National Communications.

1.1.2 The objectives of the Climate Convention

As a first step towards achieving the objective of the Climate Convention, many industrialised countries have decided to stabilise their greenhouse gas emissions at 1990 levels by the year 2000. Through the Ad Hoc Group on the Berlin Mandate (AGBM) further emission reductions after 2000 have been negotiated. During the third Conference of the Parties in Kyoto, 1-12 December 1997, a final decision will be made on a legally binding protocol for greenhouse gas emission reductions after 2000.

1.1.3 Obligation of countries to report on sources and sinks of greenhouse gases

Countries listed in Annex 1 to the Convention have the obligation to periodically report progress on their stabilisation target. Results between 1990 and 1994 show increasing emissions for carbon dioxide, decreasing emissions for methane and stabilisation for nitrous oxide (UNFCCC/CP/1996/12/Add. 2). But *"How can the emission of greenhouse gases be accurately contained within some limit yet to be determined if nobody knows for sure what the emissions are?"* (Nature, 6 April 1995). The emission estimates are characterised by uncertainties which could affect the assessment of policies and measures.

1.1.4 Uncertainty

Emission inventories are characterised by uncertainties related to the difficulty of extrapolation of measurements from experimental research (emission factors) and uncertainties in the statistical data produced by a country (activity data). The uncertainty is considered lowest in the largest sector of greenhouse gas emissions, namely about 10% in carbon dioxide emission estimates from fossil fuel combustion. The uncertainties in estimates of carbon dioxide emissions from deforestation however are much higher. This is because the methodology to assess emissions from land use change, including carbon dioxide, methane and nitrous oxide is in its early stages. Further research will be needed to increase the accuracy of this methodology. The uncertainty in methane emissions is often cited at 20-30% for most anthropogenic sources. The uncertainty in nitrous oxide emissions is often estimated higher. The most important reason for the large uncertainties in methane and nitrous oxide emissions is the temporal and spatial variability in biogenic sources.

1.2. Purpose of this study

This study is part of a larger project. The purpose of this preliminary study was to investigate the feasibility for comparing different types of emission inventories (e.g. National Communications, science based (Bottom-Up) emission inventories and (Top-Down) budgets derived from atmospheric modelling) in order to target areas for improvement of IPCC inventory methodology. This will be carried out through the analysis of available information and taking into account the reported uncertainties of the inventories. The analysis will be based on: National Communications of countries to the Climate Convention Secretariat; emission inventories of other country studies; global emissions databases; results of (inverse) modelling calculations; data assimilation studies where measurement data are assimilated in models to enhance their performance.

This study will aims at proposing a methodology for comparison of Bottom-up and Top-down emission data. Bottom-up inventories are defined here as all inventories based on a simple accounting method with an estimation of emissions from activity levels and emission factors. Top-down estimates are defined here as results from atmospheric transport and chemistry models with measurement data of atmospheric concentrations of GHGs.

In the next phase of the project further analysis of target areas will be done in order to make recommendations for improvement of IPCC methodology.

Questions to be answered in this study:

1. What bottom-up inventories (national, regional or global) are available for CO_2, CH_4 and N_2O? How are they derived?
2. How are emission data presented?
3. How could these inventories be compared and what can we learn from this comparison? Is it possible to develop a preliminary methodology for comparison?
4. To what extent are uncertainties in emission estimates quantified?
5. What can we learn from a preliminary comparison with respect to emissions inventories? Where can IPCC methodologies for emissions inventories be improved in the future?
6. What is the best format for comparison of bottom-up inventories with top-down inverse modelling results?
7. What are the experiences of experts in the field of emission estimation? Can they comment on preliminary results?

1.3. Method

The following steps will be taken to address the research questions:

1. Inventory and documentation of the available material that can be used in Top-down/Bottom-up comparisons.
2. Analysis of existing studies, specifically concerning the way in which emission data are presented.
3. Explore the possibilities of developing a standard presentation for emissions data (e.g. sectoral subdivision) to enable comparison of Top-down and Bottom-up estimates.
4. Description of the uncertainties in emissions estimates and model outcomes, based on existing material.
5. Presentation of the results to improve insight into the uncertainties of the various emissions estimates.
6. A preliminary comparison based on an analysis of case studies from different world regions.
7. Review of the draft results by international experts.
8. Incorporation of comments and completion of the report.

Step 1: Inventory and documentation of available material for comparison

A catalogue of available material at the different institutes co-operating in this phase will be maintained. A comparison of national inventories with global inventories will make use of official national communications and related background documents, the results of the United States Country Studies Programme (USCSP, summarised by Braatz *et al.*, 1996), and EDGAR data (Emission Database for Global Atmospheric Research) (Olivier *et al.*, 1996). Comparison with additional other data is also possible, e.g. CORINAIR 94 inventories for the European Union (Jol, 1996). A comparison with GEIA data (Global Emissions Inventory Activity of the International Geosphere Biosphere Programme) will be more difficult as often in GEIA no sectoral overviews and no emission factors are available. For the comparison with top-down budgets, aggregated national inventories will be used, supplemented with EDGAR emission inventory data.

Step 2: Analysis of the existing studies

Bottom-up comparison of emissions inventories
The national emissions inventories submitted to the Climate Convention Secretariat which are produced using the *IPCC/OECD/IEA Guidelines for National Greenhouse Gas Inventories (1995)* are characterised by uncertainty ranges. Some evaluation mechanism is needed for the emissions estimates of the direct greenhouse gases (carbon dioxide, methane and nitrous oxide). An independent check through comparison with global atmospheric concentration measurements could be a starting point. Bottom-up comparison of national inventories with other international emissions databases, e.g. EDGAR, CORINAIR, GEIA, is not, strictly speaking, a completely independent check but can be used to identify target areas of large uncertainty or for improvement of the methodologies.

Comparisons are done to check the consistency and accuracy of the national inventories and to reduce the uncertainties in different sources of emissions data. The comparison will point at the major areas of uncertainty and future possibilities to improve the IPCC Methodology. In this preliminary study, global emission inventories derived from emission databases like

EDGAR and GEIA will be compared with national inventories. This is a method to compare global budgets with national estimates as a tool to reduce uncertainties and to check for inconsistencies and gaps in inventories.

Top-down comparison with atmospheric budgets

Working Group 1 of the IPCC published global budgets of greenhouse gases based on atmospheric concentration measurements and atmospheric modelling (IPCC, 1990, 1992, 1994, 1996). We note that the term budget used in the context of this report is different from the use in the UNFCCC documents, where budget means the total emissions to be reduced by a country. The IPCC Second Assessment Report (1996) shows a range in sources and sinks of greenhouse gases, so the measurements and models are also characterised by uncertainties. Inverse modelling gives information on the emissions that explain a certain global concentration field. Atmospheric models are running with "a priori" emission estimates. Forward modelling can be done with national inventories as an "update" of the "a priori" emission estimates. In this report existing model exercises for carbon dioxide, methane and nitrous oxide will be evaluated.

Step 3: A standard presentation of emissions data to enable comparison of Top-down and Bottom-up estimates

One of the aims of the preliminary study was to develop a methodology which allows national inventories to be compared with international inventories and with results from atmospheric budget studies, notably inverse modelling results (i.e. global budgets).

National inventories present results as national totals per year for economic sectors. The results of global models (in the reverse mode) offer different temporal and spatial resolution (e.g. total emission per month, per grid or per latitudinal band, per source group). By measuring concentrations of isotopes, a split may be possible between different sources.

Before a comparison of top-down modelled emissions output with data from bottom-up national inventories can be made, a compatible format must be obtained. Different spatial and temporal comparisons are possible:

- National totals per year could be compared with model results.
- National totals per month could be constructed from yearly totals if a comparison is made with monthly calculations from models.
- National totals per sector could be compared with model outcomes for groups of sources.
- National totals per sector per month could be compared with detailed model calculations.
- National totals per sector could be distributed at gridcell level, thereby providing a means to compare global 2D distributions or 1D latitudinal distributions with global results of inverse modelling.

The EDGAR software provides the unique facility of converting national totals per sector of any dataset, e.g. official UNFCCC submissions, to a 1^0x1^0 grid, thereby providing the required format for comparison of bottom-up results with top-down results of inverse modelling, either 1D or 2D.

In the present study we will explore the possibilities of developing standard methodology for top-down/bottom-up comparisons, and comparisons of emission inventories. Here we will start with comparisons by IPCC sector. EDGAR sectors are sometimes combined and sometimes split to make them comparable with IPCC sectors (See Appendix 1). The level of detail for the comparison is chosen to be the Summary Tables 7A and 7B from the Reporting Guidelines volume 1: Reporting Instructions *(IPCC/OECD/IEA, 1995).* In addition, the EDGAR software provides a unique facility for converting national totals per sector of any dataset, e.g. official UNFCCC submissions, to a distribution in a 1x1 degree grid.

Step 4: Uncertainties in emission estimates and model results

In national inventories different sources of uncertainty can be recognised including:

1. Uncertainties in the national and international statistics on activity levels in the economy;
2. Uncertainties arising from upscaling of experimental research. This is a translation of measured emissions in national emissions per unit of activity or per unit of area (emission factors);
3. Uncertainties in the accuracy of the extrapolation methods developed to estimate emissions.

The accuracy will be checked for both national and global inventories. The precision of methods is related to the extrapolation from measurements at the field level to the national level. This has not been analysed in this preliminary study.

Step 5: Presentation of the results to assess uncertainties

Different authors have published validation studies for carbon dioxide, methane and nitrous oxide (Fung *et al.* 1991; Ciais *et al.* 1997a and 1997b; Hein *et al.* 1997; Kaminski *et al.* 1997; Hartley and Prinn, 1993; Bouwman and Taylor, 1996). However these validations were performed at the global scale and did not consider the level of detail contained in the national inventories as submitted to the Climate Convention Secretariat. The three gases under consideration have different peculiarities.

Carbon dioxide
Partitioning of the anthropogenic flux of carbon dioxide from fossil fuel combustion, burning of natural vegetation and deforestation over the most important sinks, namely the oceans and the biosphere, is still unclear. The extra input of carbon dioxide from human activities is masked by the seasonal exchange between atmosphere and biosphere. Also, it is unclear how global warming will affect this partitioning over the reservoirs in the future. A warming from a CO_2 increase may increase the fertilisation effect. It is unclear how the biosphere and the ocean uptake will be affected in such a scenario.
Methane.
For methane, most biogenic sources are natural but these are also enhanced by anthropogenic influence, e.g by agricultural practices. So it is difficult to distinguish purely natural from purely anthropogenic sources. The fossil fuel methane input is poorly constrained because the emission of C isotopes from the nuclear energy sector seems to be higher than earlier thought (Vermeulen *et al.* 1996). For Europe the validation of methane emission inventories has been performed by different research groups (Vermeulen *et al.*, 1996; Zhang, 1996; Janssen *et al.*, 1997; Van der Wal 1997; Stijnen, 1997). Their experience will be used to develop an overall method for validation of the IPCC methodology.

Nitrous oxide
Nitrous oxide is relatively inert in the troposphere, but again, exchange with the biosphere makes it difficult to complete the budget. Annual variations of concentrations are caused by variation in emissions.

In this step we will compare the uncertainties of model results with the uncertainties of the national inventories. It is assumed that if the model results are more precise than the inventory results, a verification may take place that leads to a reduction of uncertainties.

Step 6: Preliminary case studies for different world regions

A comparison will be made between countries and for the following world regions: European Union, rest of OECD, Eastern Europe and former Soviet Union, Rest of the World 1 (country studies available), Rest of the World 2 (only EDGAR results available).
The result of this preliminary comparison is a recommended methodology for comparison of Top-down and Bottom-up inventories. Recommendations will be formulated for a comparison of Bottom-up inventories (BU-BU); comparison of Bottom-up against Top-down inventories (TB-BU); and comparison of Top-down inventories (TD-TD).

Step 7: Review of the draft report

Experts have been asked to review the report and to provide suggestions for the organisation of the Expert Meeting in November 1997. The review has taken place in July when the first draft of the report was completed.

Structure of this report

The remainder of the report is divided in chapters, one chapter for each greenhouse gas, along with a concluding chapter. In each chapter the available information will be described: how emissions are reported, what methods of comparison are available, how the uncertainties are described by others, some examples of comparisons, what can be learned on methodology of other authors for comparisons, and a description of a recommended method for comparison.

2. Analysis for carbon dioxide

2.1. Introduction

The Top-down with Bottom-up emission inventory comparison presented here is based on readily available information only. National Communications as summarised in the UNFCCC Second review and synthesis report for the Second Conference of the Parties in Geneva (UNFCCC/CP/1996/12/Add. 2) and US country study results as summarised by Braatz *et al.* (1996) will be compared with sectoral data from EDGAR (Olivier *et al.*, 1996). The GEIA CO_2 inventory does not show sectoral detail (Andres *et al.* 1996), therefore no comparison could be made with GEIA data. Top-down models for CO_2 will be described and a method for comparison will be developed. Available models of atmospheric budgets for carbon dioxide have been published by Tans (1989, 1990), Conway *et al.* (1994), Ciais *et al.* (1995, 1997a, 1997b), Keeling *et al.* (1989) and Kaminski *et al.* (1997).

2.1.1 Description of the available databases (EDGAR)

A global emissions source database called EDGAR (*Emission Database for Global Atmospheric Research*) has been jointly developed by TNO (Institute for Applied Technical Research) and RIVM (National Institute for Public Health and the Environment) to meet the needs of both policy-makers and atmospheric modellers. The EDGAR database provides estimates of annual emissions for 1990 on a sectoral basis of the direct greenhouse gases CO_2, CH_4 and N_2O, of the indirect greenhouse gases CO, NO_x and non-methane VOC (precursors of tropospheric ozone), of SO_2 and also of various ozone-depleting compounds (halocarbons, e.g. CFCs). Version 2.0 consists of a complete set of activity data, emission factors and grid maps for both anthropogenic and most biogenic sources to allow estimation of the total and sectoral source strength of the various gases with a $1^{\circ}x1^{\circ}$ resolution. A partial validation, by comparing our estimates per major source with global total estimates of *the Intergovernmental Panel on Climate Change* (IPCC) generally showed good agreement (Olivier *et al.*, 1996). The construction of the database was part of the Dutch *National Research Programme on Global Air Pollution and Climate Change* (NRP-1) and was embedded in the *Global Emissions Inventory Activity* (GEIA), which is a component of *the International Global Atmospheric Chemistry Programme* (IGAC) Core Project of the *International Geosphere-Biosphere Programme* (IGBP). The EDGAR database, located at RIVM, serves as an analysis tool and as an emission generator for other atmospheric modelling groups, both within RIVM and TNO, and externally. In addition, it functions as the database to provide RIVM's climate model IMAGE with the base data to drive the model calculations on future emissions scenarios. In Olivier *et al.* (1996) a description is given of the construction and contents of the database, including set-up of the emission source categories; description of sources and related data (activity levels, emission factors, maps used to allocate emissions on grid); resulting emission inventories (by region and on grid) including a first validation; and uncertainties and limitations. The database has the possibility to convert country totals per year to a grid of 1 x 1 degree, using various distribution functions based on maps of point sources like power plants, of human population density, cattle population density and other variables. The database can calculate zonal totals of 10 degree bands for comparison with other inventories. An update is currently underway which will include 1995 data.

The database calculates emissions of greenhouse gases from activity data and emission factors stored in the system using a "process approach". The underlying information is organised by source category, by country or region or as gridded maps, and for a number of sources, by season. The following source groups are available in the system:

- energy production and use (by sector and fuel type; including road traffic, stationary combustion)
- industrial production (for several products)
- biofuel use (e.g. fuelwood)
- agriculture (rice production, animal breeding by type, fertiliser use)
- biomass burning (deforestation and savannah burning)
- waste treatment (landfills, waste burning)
- natural sources (soils, vegetation, oceans, wetlands, lightning).

In general, emissions are calculated from activity data and emission factors. Subsequently, the per country emissions can be allocated to a $1^{\circ}x1^{\circ}$ grid using information on point source locations of power plants, on the distribution of hard coal consumption for power generation, or on the number of people distributed over the grid cells. Some emission sources, e.g. N_2O emissions from organic soils, require a more sophisticated approach (e.g. by applying a temperature dependent model). The activity data in EDGAR are obtained from international statistics, e.g. IEA (energy data), UN (industrial production and consumption), and FAO (agricultural data). This data is mostly collected or estimated at the country level, except for three biogenic sources where gridded data is used as basic activity data (e.g. of soil types). Emission factors are either defined uniformly for all countries, such as for CO_2, or evaluated for individual countries or for groups of countries (regions). In the latter case OECD countries are distinguished from Eastern Europe and former USSR, and other non-OECD countries. In some cases, such as for road traffic, emission estimates for individual countries and independently defined activity levels are used to derive country specific emission factors. When available, major point sources are included as distribution parameters; thematic maps on $1^{\circ}x1^{\circ}$ grid are used as allocation functions to convert country emissions to gridded emissions. For fuel combustion in industry and electric power generation, point source information and area source data are used from the TNO-MW database 'LOTOS' and from US-EPA to distribute country totals for Europe and the USA, respectively. For other regions population density is used as a correlate. The same approach is used for some industrial sources.

2.1.2 GEIA

GEIA is the Global Emissions Inventory Activity of the IGAC, the International Global Atmospheric Chemistry Project of IGBP, the International Geosphere-Biosphere Project. GEIA produced gridded inventories for most of the atmospheric trace gases and pollutants. Currently, the major difference between the inventories of EDGAR and GEIA is that GEIA inventories provide the best global gridded inventories available to date for specific compounds, but they often lack sectoral details. Furthermore, as GEIA inventories are partially compiled by concatenation of existing regional emission inventories on grid, these inventories may not always cover all source categories, nor may they have options to distinguish major source categories or aggregated sectoral emission factors. The EDGAR inventories, however, have been developed using sometimes less detailed national data, but they are as comprehensive as possible and are complete and consistent in geographic coverage, sources and compounds. The sectoral details provided by the EDGAR database are a major advantage for policy applications, which are often directed at specific sectors, and for modellers, who often require additional assumptions that are mostly sector-specific (e.g. seasonal variation, or stack height). In addition, EDGAR inventories include activity levels and emission factors separately, which increases the applicability for policy purposes. Of course, both types of inventories exchange information to the extent possible, in view of their structural differences. For CO_2 the GEIA inventory does not include a sectoral split. Thus it can not be used for comparisons with official country results. Both inventories use similar emission factors. Minor differences can be found in the assumptions on completeness of combustion, and on the fraction of feedstocks

which is oxidised within a year. The assumptions in GEIA are similar to the IPCC defaults as used in the Guidelines. A detailed comparison of GEIA and EDGAR is described in Andres *et al.* (1996).

2.2. Monitoring carbon dioxide concentrations in the atmosphere

Systematic measurements of atmospheric concentrations of carbon dioxide on a global scale have been carried out by Tans *et al.* (1989), Tanaka (1987), Ciais *et al.* (1995, 1997) and Keeling *et al.* (1993). Those used in this analysis are provided in table 2.1 and figure 2.1.

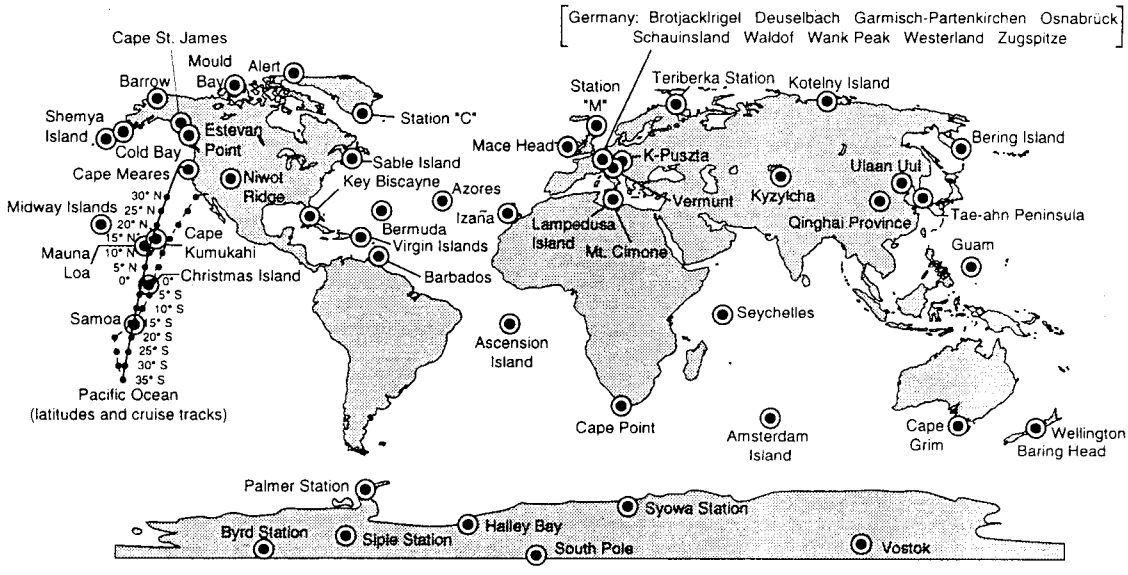


Figure 2.1 Map of CO_2 background measuring stations.

Table 2.1 Overview of CO_2 databases of concentrations and their specifications.

Network	measuring period	location	sampling period	ref.
NOAA/CMDL	1981-1988	26 global background stations	weekly, continuous	Tans *et al* (1989)
Tohoku Univ (Japan)	1882-1988	shipboard	2.5 week	Tans *et al* (1989)
INSTAAR	1992	40 sites δ^{13}	weekly	Ciais *et al* (1995)
CSIRO	1992	4 sites δ^{13}		Ciais *et al* (1995)
WMO	1972-1989	$2x2^0$ ΔpCO2 in the oceans	seasonal	Tans (1990)
SIO	since 1958	4 sites	hourly	Keeling (CDIAC 1993)

A database containing most of the systematic global atmospheric measurements of greenhouse gases concentrations in the atmosphere has been established by the initiative of the WMO-GAW program. This is the World Data Centre of Greenhouse Gases (WDCGG) database located in Tokyo (WMO-GAW, 1996). Another global database is the CDIAC (Carbon Dioxide Information Analysis Center) data centre in Oak Ridge Labs USA (CDIAC, 1993). Both are easily accessible through the internet. The sites of the measuring stations are chosen in such a way as to yield data that are expected to be representative of large-scale air masses and therefore of large scale CO_2 sources and sinks and the major features of atmospheric transport. They should therefore be considered as background concentration measuring stations. Measurement frequency is not high (only once a week in remote areas). In north-west Europe measuring stations can be found in the more polluted areas. In the US and Canada measuring stations are operational. For more detailed insight into the regional pollution in Russia, Asia, Africa and Latin America, more measurement stations are needed in those polluted areas

2.3. Top down modelling

2.3.1 Methodologies for CO_2

The accuracy of the comparison of Top-down with Bottom-up emission inventories largely depends on model specifications and the kind of model. It also depends on the spatial and temporal resolution of the available data. We can distinguish between different types of models. An integrated model like e.g. IMAGE 2 has a grid level of detail for emissions but uses a sub-model for the atmosphere that is not as detailed as a general circulation model (GCM). The advantage is that scenarios can be calculated rather quickly on future concentrations given developments in population, energy use, land use and deforestation along with related emissions of greenhouse gases. In atmospheric chemistry and transport models a distinction can be made between one-dimensional models and two or three-dimensional models. In one-dimensional models output is calculated for one longitude and for different layers in the atmosphere. In two-dimensional models output is calculated for zonal averages and in different layers for the atmosphere. In three-dimensional models output is given for longitude and latitude and height. Different atmospheric model types can be distinguished. Atmospheric models which use the actual physical and chemical processes to calculate output are called deterministic models. Other models use differential equations to come up with the same parameters. Adjoint models calculate emissions in the forward mode and in the inverse mode and are highly sophisticated for inverse modelling. Stochastic models are used to introduce noise in existing models to calculate uncertainties or are used as a tool for interpolation between base emissions inventories and measured concentrations. In theory the best results in the comparison of Top-down with Bottom-up inventories can be expected when using the three-dimensional models with stochastic models for interpolation between existing measuring stations. National inventories are not generally gridded but can be gridded using distribution functions for human population density, cattle population density, energy use, transport and so on. Output from inverse modelling has the form of totals per grid per gas per month. This output has to be split into sectoral emissions in order to make a comparison. Trajectory analysis and ^{14}C data can be used to enhance this sectoral detail. National inventories per year can be split into totals per month using information on the seasonal distribution of the sources.

2.3.2 Global CO_2 model IMAGE

IMAGE 2 is the second version of the Integrated Model to Assess the Greenhouse Effect (Alcamo (ed.), 1994; Alcamo and Kreileman, 1996). The model can be used to validate the existing CO_2 budgets. IMAGE 2 was developed for scenario analysis for the period 1990 to 2100. The model is calibrated with data for the period 1970 to 1990. Carbon dioxide emis-

sions are calculated from energy statistics, deforestation data, changes in the biosphere, ocean uptake and climate feedback's like the carbon dioxide fertilisation effect and the extra CO_2 emitted from warming of tundra and other soils. In IMAGE 2 total emissions are calculated and this is translated into a concentration in the atmosphere. The model including scenario analysis is described in a special issue of Soil, Water and Air Pollution (Alcamo ed., 1994) and in a special issue of Global Environmental Change (Alcamo and Kreileman, 1996; Leemans *et al.*, 1996). IMAGE 2 can not be used in the inverse mode. But it can calculate atmospheric concentrations of greenhouse gases from very detailed source inventories and future emissions under different scenarios of population growth and economic developments, including land use change and changes in the future eating habits of people. Global budgets can be compared with the IMAGE results for different periods in time and between different authors, e.g. Heimann 1991-1994 and IPCC 1980-1989.

2.3.3 Budgets comparison and model results for CO_2

Working group I of the IPCC published global budgets of greenhouse gases based on emission inventories and based on measurements and modelling of the compartments of the climate system: the air, ocean and biosphere (IPCC Climate Change 1990, 1992, 1994, 1995). In this paragraph the possibilities to derive emissions of greenhouse gases from atmospheric measurements and models are discussed (this is the Top-down analysis).

The partitioning of the anthropogenic emission flux of carbon dioxide from fossil fuel combustion, burning of natural vegetation and deforestation to the most important reservoirs and sinks namely the atmosphere, the oceans and the biosphere is not well determined. The increase in atmospheric CO_2 due to the extra input due to human activities should be filtered from the large yearly and seasonal fluxes of CO_2 between the atmosphere, the biosphere and the oceans. The accuracy of the estimation of the anthropogenic CO_2 input into the atmosphere depends on the spatial and temporal resolution of the available data and the model specifications.

Long term budgets (10 year time scale).

Most of the global carbon cycle models used in IPCC Assessments address only the longer term (10 year time-scale) direct perturbation of the global carbon cycle due to anthropogenic emissions (IPCC, Climate Change 1995). The effect of additional fossil fuel and biomass carbon injected into the atmosphere is a long-lasting disturbance of the carbon cycle see figure 2.2 . The measuring record itself provides support that anthropogenic emissions are a source of the observed increase, see figure 2.3.

When seasonal and short-term interannual variations in concentrations are neglected, the rise in atmospheric CO_2 is about 50% of anthropogenic emissions (Ciais *et al.*, 1995). These aspects are in accordance with our understanding of the carbon cycle and agree with model simulations, (see figure 2.4). Based on long term time series of atmospheric CO_2, measurements and model calculations, long term CO_2 budgets can be established (see table 2.2 IPCC 1995; Heimann, 1997) and figure 2.4 (Kreileman and Leemans, 1997) using the IMAGE 2 model.

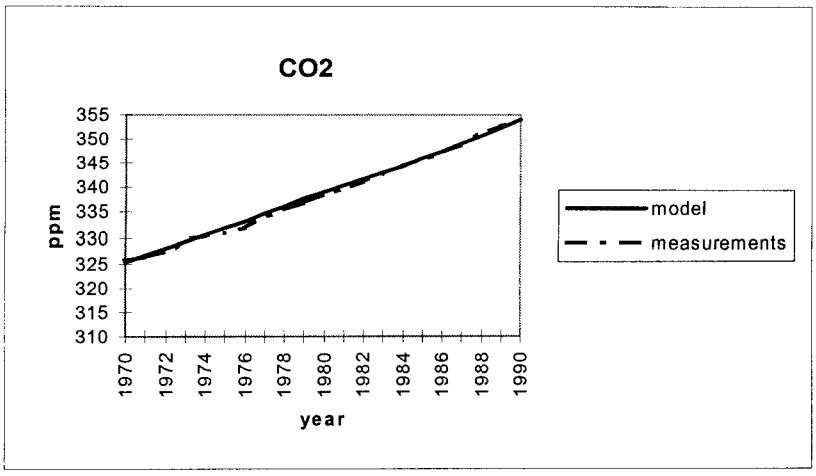


Figure 2.2 Time series of measured and modelled CO_2 concentration in the atmosphere at Mauna Loa (Keeling *et al.* 1993.)

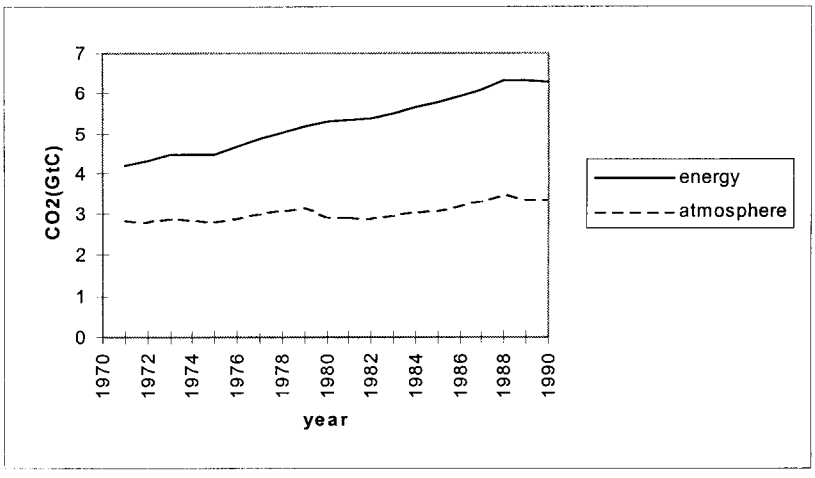


Figure 2.3 Time series of CO_2 fossil fuel emission and atmospheric increase (Kreileman and Leemans, 1997)

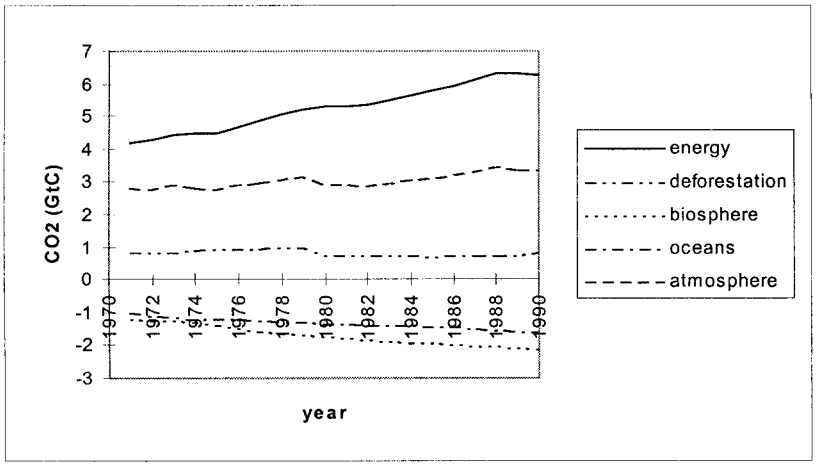


Figure 2.4 Modelled CO_2 emissions using the IMAGE 2 model (Kreileman and Leemans, 1997).

Table 2.2 CO_2 budget comparison IPCC and Heimann.

CO2 budget (GtC/yr)	1980-1989 (IPCC 1994)	1991-1994 (Heimann, 1997)
(1) Fossil fuel and cement production	5.5 ±0.5	6.1 [4]
(2) Atmospheric increase (observed)	3.2 ±0.2	2.3 [5]
(3) Ocean uptake (model calculated)	2.0 ±0.8 [1]	2.0
based on observed O_2/N_2 ratio trend (1989-1994)	1.9 ±0.8 [1]	
based on observations of $^{13}C/^{12}C$ ratio	2.1 ±1.5 [2]	
(4) Net balance of terrestrial biosphere (= net sink) = (1)-(2)-(3)	0.3 ±1.0	1.8 ±1.1 [1] (or 2.0)
(5) Land use change emissions (primarily tropics)	1.6 ±1.0 [7]	
Net emissions from tropics	1.0 [7]	≈ 0.0 [7]
(6) Regrowth of temperate latitude forests (based on forest statistics)	0.5 ±0.5	0.0 (see (7))
(7) CO_2 uptake by other terrestrial processes (=net sink) (a.o. CO_2 fertilisation effect, N-fertilisation, climate effects) = (4)+(5)-(6)	1.4 ±1.5 (0.8 NH and 0.6 Tropics/SH) [8]	1.9 ±1.5 [3] (= 0.5 + 0.8 + 0.6 [6])

[1] 1.9 ±0.8 (Keeling)
[2] 2.0 +1.5 (1970-1990; Heimann)
[3] extra tropical northern hemisphere
[4] (1994; IPCC 1995)
[5] (own estimate) : = 6.1-2.0-1.8
[6] Extra 6.1-5.5 = 0.6 CO_2 emission is compensated with extra CO_2 uptake in the northern hemisphere,
To keep the interhemispheric gradient the same and at the same time reduce the atmospheric increase
So the total northern hemispheric sink has been increased from 1.3 (= 0.5 + 0.8) to 1.9
Total was 0.5 + 0.8 = 1.3
New total = 1.9
[7] Tropical net emissions: 1.6 - 0.6 = 1.0
[8] Own estimate of NH and Tropics ratio

Table 2.2 indicates that the storage in the atmospheric reservoir is the most accurately known (Δ < 10%). Ocean uptake is less accurately known ($\Delta \approx 50\%$) and uptake of anthropogenic CO_2 by the biosphere is the least accurately known ($\Delta \approx 100\%$). Only the increase of CO_2 in the atmosphere is derived directly from observations. However, the CO_2 emissions due to fossil fuel burning and land use change are devided between the ocean, biosphere and atmosphere as the main reservoirs and thus cannot be directly derived from atmospheric measurements. The increase of CO_2 in the other reservoirs are derived from model calculations sometimes supported by isotope measurements of Carbon and/or Oxygen (O_2) or flux measurements of ΔpCO_2 in ocean waters (Tans *et al.*, 1990). The amount of uncertainty of the storage of CO_2 in these 3 reservoirs is much larger than the uncertainty of the fossil fuel emissions (Δ = 10%) based on emission inventories (Marland *et al.*, 1985). Thus estimates of the fossil fuel emissions derived from atmospheric measurements and CO_2 (Top-down) budget calculations cannot be calculated over this period with more accuracy than those based on emission inventories. A crude estimate about uncertainty ranges is:

Δ fossil fuel = 10% derived from emission inventories (Marland *et al.*, 1985).
Δ fossil fuel = $((3\%)^2 + (11\%)^2 + (7\%)^2 + (21\%)^2)^{1/2}$ = 25% derived from a budget analysis using the data of Table 2.2.

The range in the uptake of CO_2 in the oceans is about 1 GtC and the range in the net uptake of the biosphere is about 2 GtC. These ranges should not be added because both sinks are interdependent (anti correlated) so the uncertainty range of the sinks is about 25-30%. Fossil fuel emissions are a part of the carbon budget. We estimate that fossil fuel emissions derived from an analysis of a long term budget (5-10 years) have an uncertainty range of 3-7 GtC.

Short term variations

An evaluation of long term time series shows that the rise of the atmospheric CO_2 concentrations is about 50% of the anthropogenic emissions (IPCC 1995). The oceans and the biosphere are important sinks for fossil CO_2 and take up about half of the emissions. Figure 2.5. shows the variation in the growth rate of the CO_2 concentrations since 1958 in ppm/yr at the Mauna Loa station in Hawaii (IPCC 1995).

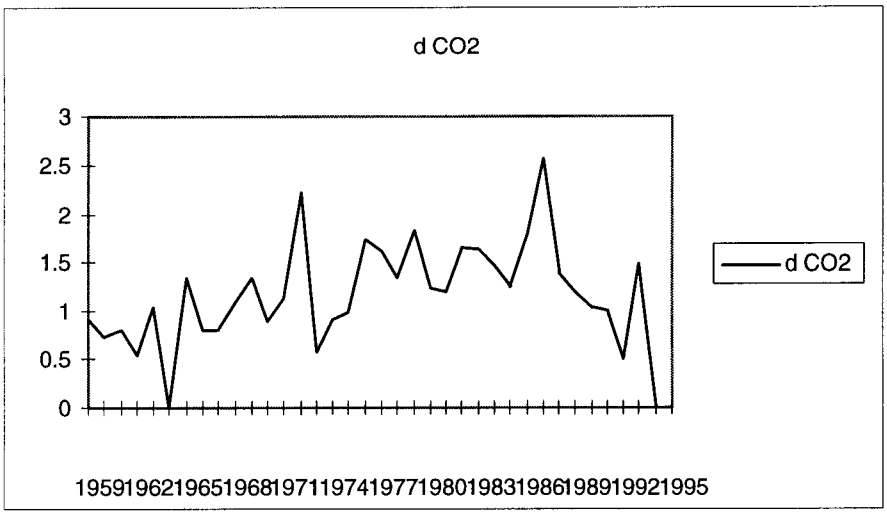


Figure 2.5 Yearly variations of the growth rate in the atmosphere 1959 - 1995

This figure shows that yearly variations of the growth rate can be between 0.5 and 2.5 ppm/yr. This means variations between 1 and 5 GtC which is in the order of 25-100% of the fossil fuel emission. Figure 2.6 shows that these yearly variations in atmospheric increase are much larger than the yearly variations or the trend of fossil fuel emissions.

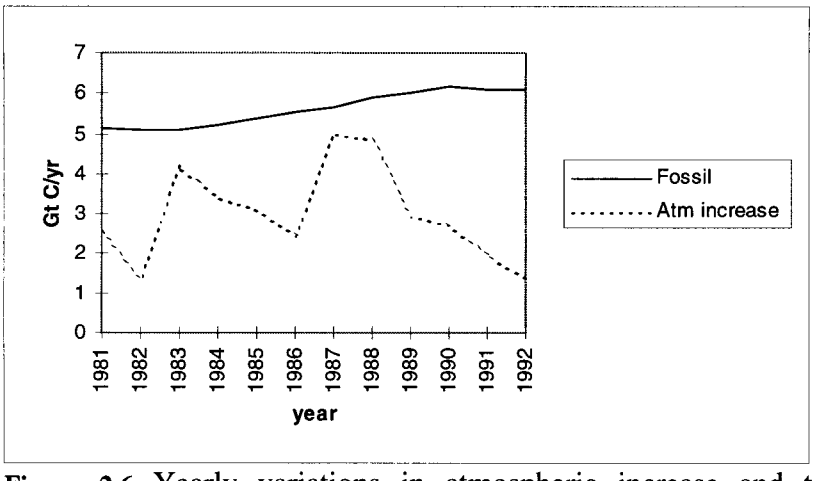


Figure 2.6 Yearly variations in atmospheric increase and trend of fossil fuel emissions (Kreileman and Leemans, 1997).

Both the magnitudes of ocean and biospheric fluxes vary substantially from year-to-year. The interannual CO_2 variations reflect imbalances in the exchange fluxes between the atmosphere and the biosphere and the ocean. Because global carbon cycle models ignore short term perturbations due to fluctuations in the climate system they are not able to reproduce these shorter-term atmospheric CO_2 variations. It is therefore not possible to estimate fossil fuel emissions derived from atmospheric measurements and global CO_2 (Top-down) budget calculations for a year with more accuracy then based on emission inventories.

Table 2.3 Overview of referenced literature on inverse modelling

model type	spatial resolution	temporal resolution	period	Ref.
2-D model	20 latitudinal grids and 10 in vertical	monthly windfields	1981-1985	Tans (1989)
3-D model	oceans 2^0x2^0	diurnal seasonal cycles, 4-hourly windfields and monthly averaged convective transport	1981-1987	Tans (1990)
2-D model	20 latitudinal grids and 10 in vertical	monthly windfields	1981-1992	Conway *et al.* (1994)
2-D model	20 intervals of equal area	14 days	1990-1992	Ciais *et al.* (1995)
3-D model	8^0x10^0 and 9 layers in the vertical	monthly windfields	1984	Keeling *et al.* (1989b)
3-D model	8^0x10^0 and 9 layers in the vertical	monthly windfields	1987	Kaminski *et al.* (1997)

2.3.4 The zonal budget

To evaluate short term variations in the CO_2 budget we need to quantify the fluxes of the oceans, biosphere and the human society on a smaller temporal and spatial scale than used in the models. In the next analysis we use the data sets described in Table 2.1 and the global models described in Table 2.2.

The models have been run in the inverse mode and emissions have been estimated to fit:
- the overall concentration field
- the averaged yearly increase (1.5 ppm)
- the vertical concentration gradient (4 ppm at 70^0)
- the zonal concentration gradient (up to 10 ppm between Northern (70^0) and Southern Hemispheres)

  This mean latitude gradient of CO_2 is primarily due to the emission of CO_2 from fossil fuel combustion, about 90% of which occurs in the northern hemisphere (Conway *et al.* 1994)
- the seasonal cycle (up to 20 ppm at 70^0 NH)
- $\delta^{13}C$ data to estimate the fluxes from and to the biosphere (Ciais *et al.*, 1995)

In the following an interpretation is given of results from model calculations. Results of the estimated zonal fluxes from the different compartments (fossil fuel, biosphere and ocean) to the atmosphere are brought into one format to make the results of the different authors and model calculations comparable. The results are shown in the figures 2.7 - 2.17. In the second phase of

this project this format will be used for a comparison with (bottom-up) national inventories aggregated to zonal totals, e.g. EDGAR results as shown in Figure 2.22.

Table 2.4 Overview of content of figures on model calculations.

Figure	Scenario type/ dominant sink	Period	Yearly averaged atmospheric increase (GtC)	Reference
7	GMCC dataset	1981-1985	4.6	Tans (1989)
8	Tohoku dataset	1882-1885	4.4	Tans (1989)
9	ocean scenario	1981-1987	2.7	Tans (1990)
10	biosphere scenario	1981-1987	2.7	Tans (1990)
11	ocean measurements	1981-1987	3.7	Tans (1990)
12	1990	1990	3	Ciais *et al.* (1995)
13	1991	1991	1.9	Ciais *et al.* (1995)
14	1992	1992	1.6	Ciais *et al.* (1995)
15	Ocean	1984	2.5	Keeling (Ciais 1995)
16	Biosphere	1981-1988	3	Tans (1990)
17	Ocean and biosphere	1992 (El Nino)	1.4	Ciais *et al.* (1995)

Discussion of the results.

Note that although the partitioning between ocean and biosphere may differ, the resultant atmospheric increase does not vary for a given period if the budget is matched. This means that without additional information (isotopes or analysis of yearly variations or independent measurements of fluxes as in figure 2.9) an univocal partitioning of CO_2 storage and emissions from ocean and biosphere is not possible.
In the following the four main sources of uncertainty are dealt with:

1) The measuring data set and representativeness of the measuring data;
2) Ocean or biosphere as the main sink;
3) Yearly variations in sources and sinks;
4) Different budget assumptions between various authors.

1. Representativeness of the measuring data

The differences in estimated emissions between the figures 2.7 and 2.8 are caused by differences in data sets. The GMCC data are mainly sampled at remote ocean sites. The Tohoku dataset also contains information of continental emissions. The Artic source that was found in the GMCC/CMDL data is considered as an artifact of the 2-D model due to the fact that continental data are lacking.

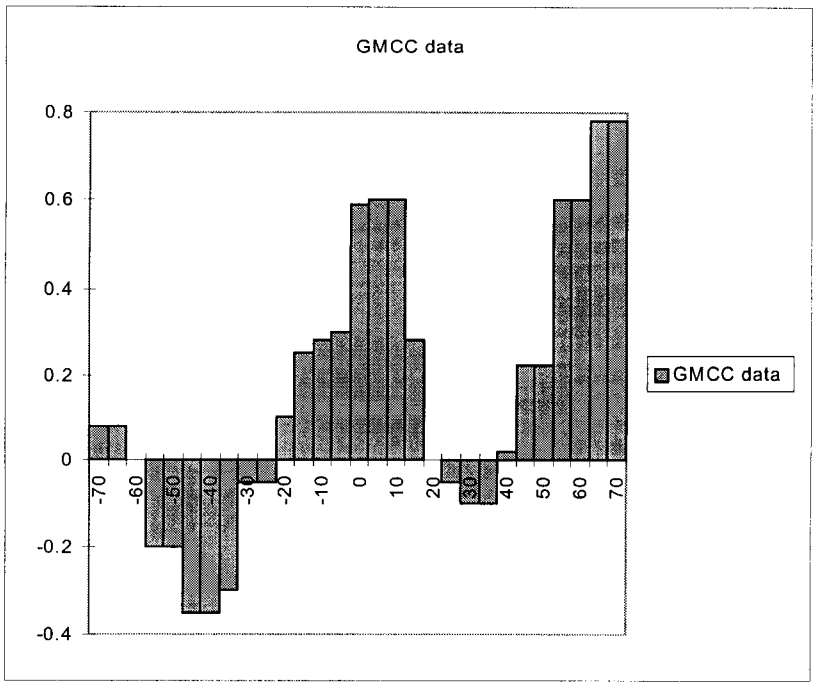


Figure 2.7 The net zonal CO_2 fluxes derived from inverse modelling with the GMCC dataset (Janssen, 1997, Based on Tans *et al.*, 1989)

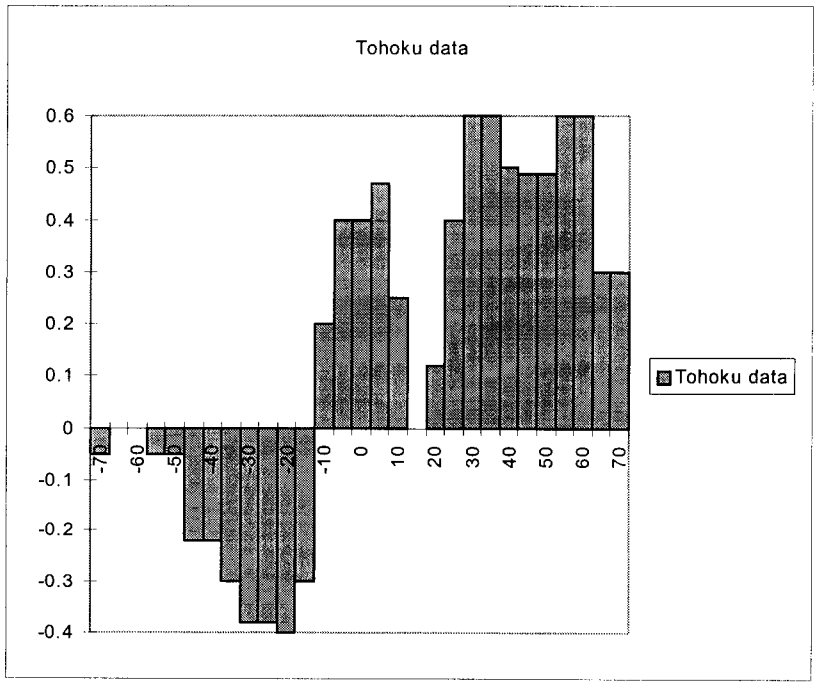


Figure 2.8 The net zonal CO_2 fluxes derived from inverse modelling using the Tohoku data-set (Janssen, 1997).

This can be dealt with using a 3-D model in the inverse mode as has been done by Kaminski et al (1997). Fluxes were derived from the biosphere by analysing the monthly variations of CO_2 concentrations at background stations with the adjoint of a 3-D atmospheric model for 1987. They show that local CO_2 fluxes from the biosphere can be substantially larger than the zonal average up to a factor of 10.

2. Partitioning of ocean and biosphere

The differences between figures 2.9 and 2.10 are caused by the assumption that the surplus of fossil CO_2 will mainly be taken up by the oceans (figure 2.9) or the biosphere (figure 2.10).

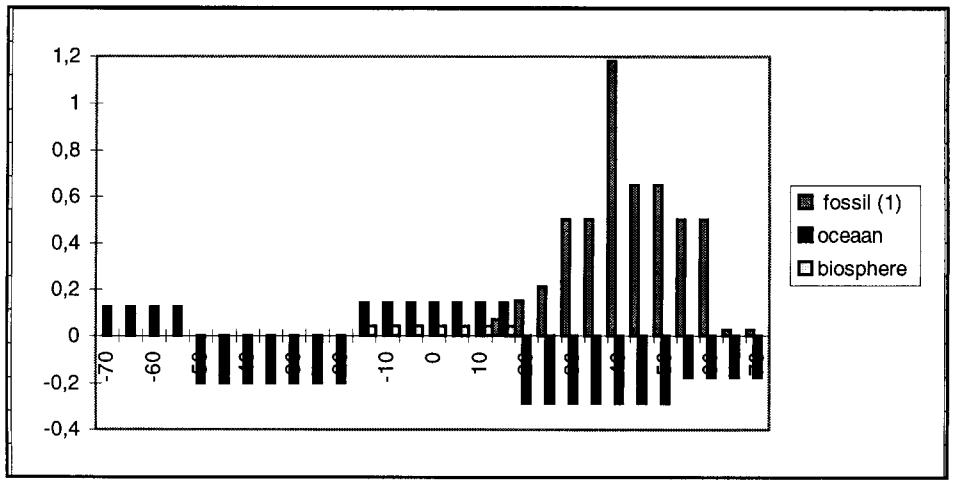


Figure 2.9 Surplus of fossil CO_2 is taken up by the oceans (Tans et al. 1990)

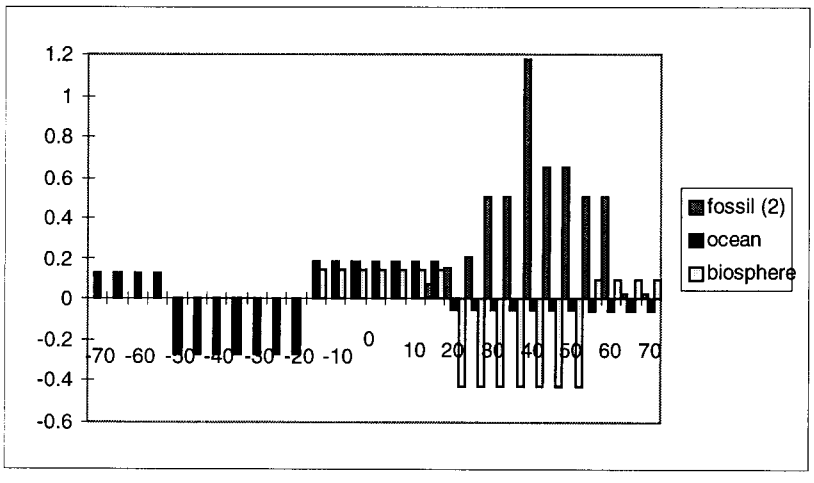


Figure 2.10 Surplus of fossil CO_2 is taken up by the biosphere (Tans et al. 1990).

3. Yearly variations

The following figures show the differences of the sources and sinks for 1990, 1991 and 1992 using data of Ciais *et al.* (1995). They show large variations over these years. Conclusion is that due to large year to year variations a comparison with bottom-up inventories for CO_2 for one year can not lead to validation of these bottom-up inventories.

Conway et al (1994) show an overview of interannual variations of CO_2 concentrations which can be derived from zonal variations in sources and sinks (Conway *et al.* 1994; Table 2.3).

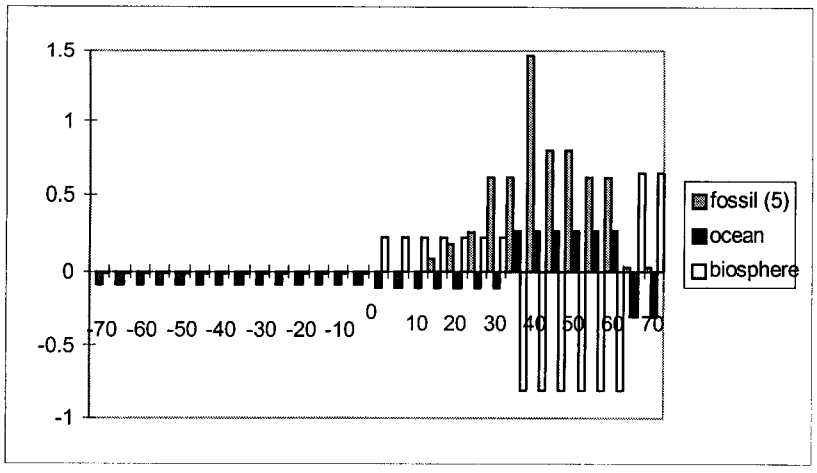


Figure 2.11 Sources and sinks of CO_2 for 1990. (Based on Ciais *et al.* 1995)

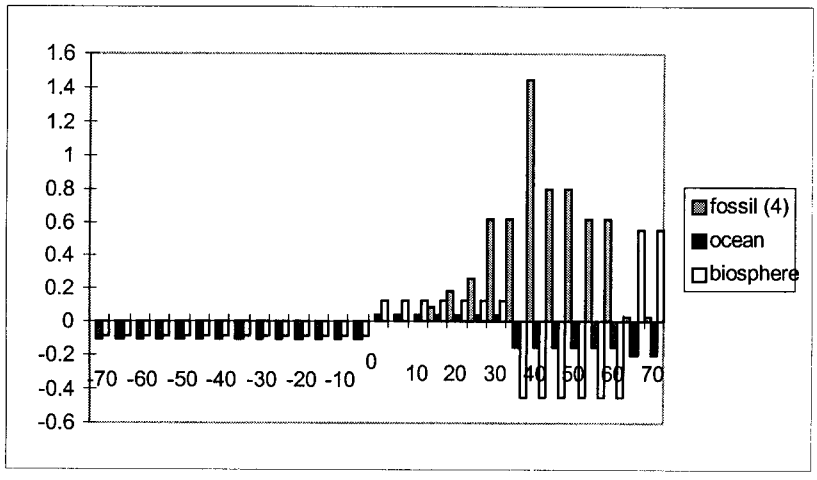


Figure 2.12 Sources and sinks of CO_2 for 1991. (Based on Ciais *et al.* 1995)

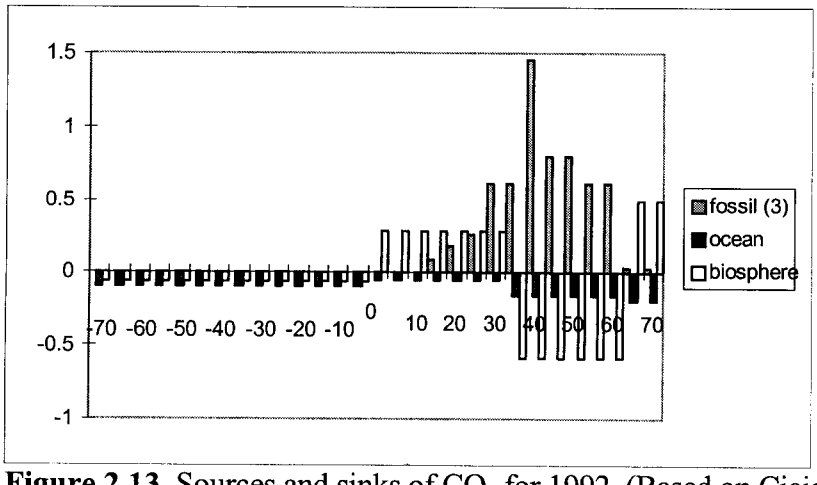


Figure 2.13 Sources and sinks of CO_2 for 1992. (Based on Ciais *et al.* 1995)

4. Different budget assumptions

The following figures show the differences in assumptions between Tans *et al.*: the biosphere is the dominant sink. Keeling *et al.*: the ocean is the dominant sink. Ciais *et al.*: both the ocean and biosphere are important sinks. This also explains the small CO_2 growth rates during the early 90's despite increasing fossil CO_2 emissions.

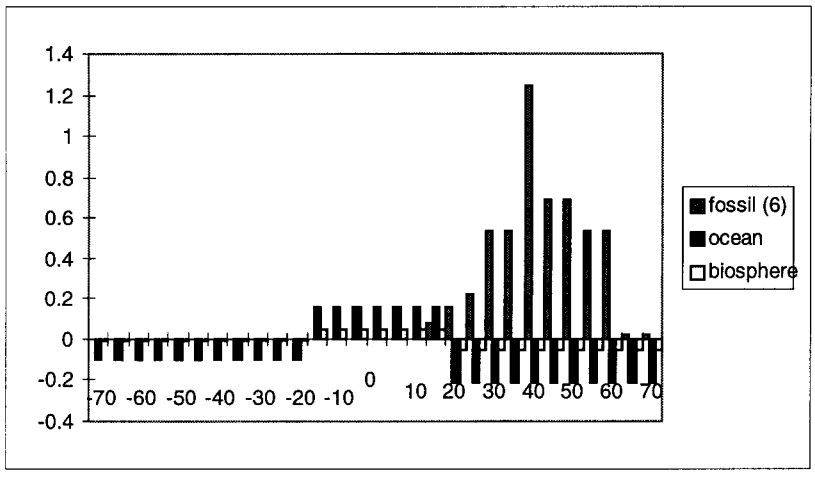


Figure 2.14 Keeling *et al.*: the ocean is the dominant sink

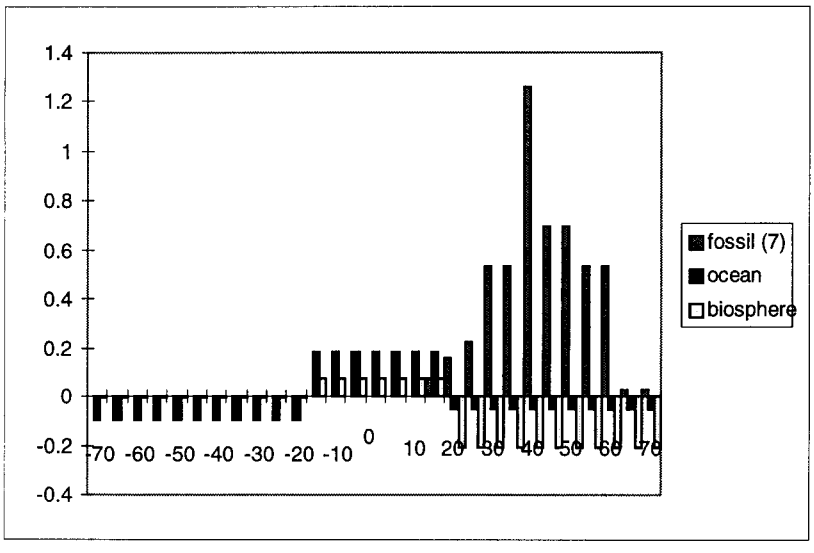


Figure 2.15 Tans *et al.*: the biosphere is the dominant sink

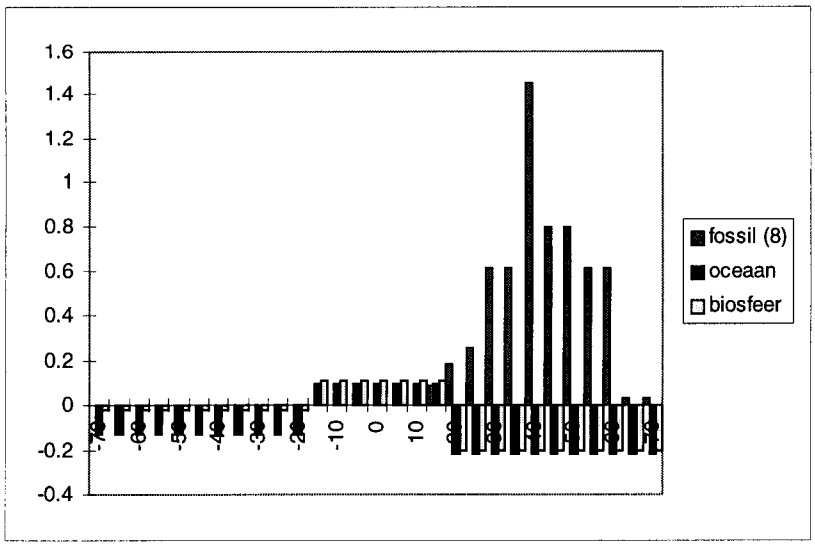


Figure 2.16 Ciais *et al.*: both the ocean and biosphere are important sinks.
This may explain the small CO_2 growth rates despite increasing fossil emissions during the early 90's.

2.3.5 Conclusions of global and zonal budget analysis

Yearly variations in the CO_2 budget are much larger than the yearly variations in the fossil fuel emissions.
Long term trends of the CO_2 concentration in the atmosphere are representative of the long term disturbance of the carbon balance due to antropgenic CO_2 emissions.
The uncertainty in the estimated CO_2 emission due anthropogenic activities derived from atmospheric measurements is 25-30%.

2.3.6 Trajectory analysis CO_2

No information is used in this first phase of the project concerning trajectory analysis of CO_2.

2.4. Evaluation of emissions inventories

2.4.1 Introduction

For this kind of evaluation to be successful, the documentation on the inventory must be complete and transparent (Van Amstel, 1993). The data must be detailed enough to reconstruct the inventory. Original calculation sheets and background reports to the National Communications should be provided and data should be referenced. A first evaluation or in-depth review on draft inventories was carried out by IPCC/OECD/IEA in 1993 to evaluate the draft IPCC methodology. For the Second Conference of the Parties the UN Climate Secretariat in 1996 published a second compilation and synthesis of 1990-1994 of official Annex-1 country inventories (UNFCCC/CP/1996/12/Add.2, 2 July 1996). An overview of emissions, emission factors and activity levels used in the inventories can be the basis for a thorough evaluation. Comparison with different databases is possible. In this preliminary study a comparison with EDGAR is made. In a second phase of the project a more detailed analysis will take place looking at emission factors and activity data. Here only emissions estimates are compared.

2.4.2 Comparison of national inventories with EDGAR data

A method is described to compare global inventories from EDGAR (Emissions Database for Global Atmospheric Research) with national inventories. National inventories are officially

submitted to the UNFCCC Climate Secretariat. Country studies from the US Country Studies Programme as summarised by Braatz *et al.* (1996) are also used. The latter are not considered official submissions but allow non-Annex 1 parties to use the IPCC Guidelines for later official submissions. Most of these country studies are made with financial support from the Global Environment Facility (GEF) fund. The comparison here is made to reduce uncertainties and to identify gaps in the existing methodology and source categories.

2.4.3 Precision, Accuracy, Uncertainties

Three sources of uncertainty can be defined: 1. Uncertainties in the national and international statistics (activity data); 2. Uncertainties arising from experimental research in the emission per unit product or unit area, (emission factors); 3. Uncertainties in the accuracy of the extrapolation methods developed to estimate emissions. The comparison is made by the sector split provided in the IPCC Guidelines (Vol. 1: Reporting instructions). EDGAR sectors are sometimes combined and sometimes split to make them comparable to the IPCC sectors. The level of detail for the comparison is chosen to be the summary tables 7A and 7B from the IPCC Guidelines Volume 1: Reporting Instructions (IPCC/OECD/IEA, 1995). The accuracy will be checked of both inventories. In some cases not all sectors are covered. This will lead to differences. The precision of the methods is related to the extrapolation from field level measurements to national level emission factors. This factor cannot be analysed in this preliminary study.

2.4.4 IPCC methodology for CO_2

The IPCC methodology for estimating CO_2 emissions from energy is based on the carbon content of the fuels and the fuel supply to the economy. Different sectors in the economy are distinguished. The main sectors for CO_2 emissions are combustion of fuels for heating, transport and industry. CO_2 process emissions from cement clinker production is a minor source. The main sink for CO_2 is sequestration by forests. This sink is decreased by deforestation in the tropics and seems to be increased by afforestation in higher latitudes. The method for calculating CO_2 emissions or sinks from the biosphere is based on the increase or decrease of the carbon content of above and belowground carbon from biomass and humus by the change in land use and vegetation by encroachment of agriculture or infrastructure. Essentially a number of ecosystems are defined, each with a related carbon content per hectare. At the moment the method is still in development. A five year average is taken to calculate carbon storage or release. Fellings and regrowth is taken into account. In table 1 in Appendix 1 the main IPCC sectors are given. In the next paragraphs the sector codes from IPCC will be used in the headings.

CO2 from combustion of fossil fuels. (IPCC sector 1A1-5)

The IPCC methodology for estimating emissions takes fuel statistics for apparent consumption and emission factors by fuel type. The IPCC Guidelines chose a tiered approach to provide for different levels of detail in the methodology, called Tier's. The level of detail chosen in the Tier 1 method is three types of fuel: solid, liquid and gaseous. In the Tier 2 method emission factors are used for each type of fuel used in a country. The default emission factors for both Tiers 1 and 2 are given in the IPCC Guidelines.

The IPCC method uses country fuel statistics for apparent consumption to calculate CO_2 from fuel combustion. A top down assessment is possible with IEA statistics. As EDGAR uses the IEA statistics EDGAR can be used for this assessment. Many European countries used energy statistics for actual consumption of fuels in the transport sector to calculate CO_2 from transportation. This way Luxembourg e.g. has a very high emission because fuel sales are high in

transport. Fuels are cheap, therefore most European transit traffic is filling up in Luxembourg. This leads to high emission estimates for CO_2 for a relatively small country.

2.4.5 EDGAR methodology for CO_2

EDGAR uses the TIER 1 approach for estimating CO_2 from fossil fuel combustion. The IEA data set is used for most of the calculations.

Fossil fuel use

Sectors where fossil fuels are used comprise energy production, transformation (i.e. production of secondary fuels such as coke and oil products) and combustion of fossil fuels (Table 2.5). Process (i.e. feedstock) emissions from coke ovens and refineries are also included here.

Table 2.5 EDGAR energy sectors for fuel combustion

EDGAR sector	IEA sector	Remark
F10. Industry	Industry	Excluding the energy sector (e.g. power generation, refineries)
F20. Power plants	Electricity generation	Public electricity, autoproducers of electricity, CHP plants
F30. Other transformation	Other fuel transformation	Refineries [1], coke ovens [1], blast furnaces, gas works, district heating, etc.
F40. RCO:	Other sector:	Sum of stationary non-industry sources (e.g. dwellings)
F41. - Commercial	- Commercial	= Commercial and public services
F42. - Residential	- Residential	= Household dwellings
F43. - Other	- Other end-use sectors	= IEA subsectors 'Agriculture' and 'Other/Non-Specified'
F50. Transport:	Transport:	Road, rail, air, and water transport, excluding marine bunkers
F51. - Road transport	- Road	Excluding off-road vehicles
F54. - RIO transport	- Rail,inland waterways,other	=Rail, inland shipping & other/non-specified transport [2]
F57. - Air transport	- Air transport	= Domestic & international air transport
F58. International shipping	Marine bunkers	International shipping as defined by bunker fuel consumption

[1] Emissions from refineries were calculated using emission factors based on refinery inputs (including combustion emissions from refineries), and for coke ovens using factors based on coke production (including combustion emissions), except for CO_2 and N_2O.

[2] Including pipeline transport.

The EDGAR energy production and consumption data sets have been constructed with some minor additions/modifications to have a better spatial distribution and a more complete estimate for major source categories. The IEA energy statistics 1971-1992 for 112 IEA countries were used, extended with 71 countries using IEA totals and country splits according to UN data. For another six countries, estimates by Samaras for road transport have been added, whereas for 37 IEA/UN countries, specific estimates for road transport have been added to existing country data sets, which did not specify fuel consumption in road transport (Samaras, 1993). More details on the construction of the fossil fuel data set is provided in Olivier *et al.* (1996).

The EDGAR emission factors for CO_2 essentially comply with the ones used in the GEIA inventory and the factors recommended by IPCC (1994). Globally uniform factors were used for combustion of coals, oil products and natural gas. In Table 2.6 these factors are summarised, including the conversion to other units. For CO_2, EDGAR treats the feedstock use of fuels and other non-energy use (such as for bitumen and lubricants) as a separate sector, with emission factors calculated as a percentage (depending on the fate of the substances) of the factor for combustion as was done in the GEIA inventory (Table 2.6). These percentages differ slightly from the defaults recommended by the IPCC. To calculate the net CO_2 emissions of the Other Transformation Sector (e.g. by coke ovens, blast furnaces, refineries) the same three values for coal, oil and gas were used, but now as negative factors, for the production of secondary fuels (coal products, gas works gas and oil products), to take into account that part of the carbon input which is not oxidised in the sector, but in other sectors using these secon-

dary fuels. If this net calculation was not made, there would have been a double counting or neglect of the losses in the transformation process itself.

The CO_2 emission factors in EDGAR for gas flaring in oil production were calculated on a per country basis from estimated emissions (Marland *et al.*, 1994) and oil production data.

Table 2.6 EDGAR emission factors for CO_2 from fossil fuel combustion (in kg CO_2-C/GJ)

Sector/fuel	Emission factor [1]	Remark (related factor used in GEIA inventory)	Reference
SOLID FUELS	25.50	Factor used for solids in GEIA CO_2 inventory is: 0.73257 kg CO_2-C/kg solid fuel (= 0.746*0.982) [3] Acc. IPCC, 1994, p. 1.22 this is equivalent to 25.5 kg C/GJ LHV	IPCC, 1994 [6]
LIQUID FUELS	19.26	This factor is average of gasoline and diesel factor (both are the fuels mostly used within the OECD) [4] Factor used for liquids in GEIA CO_2 inventory is: 0.7803 kg CO_2-C/kg crude cons. (= 0.85*0.918) (for bunker fuels 2% higher)	IPCC, 1994 [6]
GASEOUS FUELS	15.3	Factor used for gas in GEIA CO_2 inventory: 13.426 kg CO_2-C/GJ gas (= 13.7*0.98) [5] [13.7/0.9 = 15.3]	IPCC, 1994 [6]
Exceptions [2]:			
Gas-Chem. feedst.& NE	8.86	= 66% of EF gas	Marland&Rotty, 1984
LPG, ethane-Chem. f.&NE	11.76	= 60% of EF oil	Marland&Rotty, 1984
Naphtha	3.92	= 20% of EF oil	Marland&Rotty, 1984
Bitumen	0.	= 0% of EF oil	Marland&Rotty, 1984
Lubricants	9.80	= 50% of EF oil	Marland&Rotty, 1984
International marine bunkers (int. shipping)	19.65	Factor used for bunkers is 2% higher than for inland consumption.	Andres, 1994

[1] We have here ignored the unoxidised fraction from combustion, which compared to Marland and Rotty, 1984, is 1%, 1.5% and 1% for solids, liquids and gases, respectively (IPCC recommendations: 2%, 1% and 0.5%, respectively.) In addition, we assumed here that all oxidised carbon is converted into CO_2, neglecting fractions emitted as CO or other compounds.

[2] For feedstock use of fuels (in chemical industry) an emission factor of 0 has been assumed, except for gas, LPG and ethane as indicated here and except for white spirit, paraffin waxes, petroleum coke and 'other petroleum products' as well as for liquid fuel for electricity output, where we assume full oxidation (in contrast with LPG-feedstock/NE, ethane-feedstock/NE, naphtha, bitumen and lubricants).

[3] For coke production losses Marland and Rotty, 1984, assume 4.4% losses of coal throughput.

[4] Since our calculation is on a fuel-specific consumption basis, we converted from kg to GJ using the weighted average of the gasoline and diesel factor (both are the fuels mostly used within the OECD: 513 Mton and 428 Mton in 1990, respectively) [LHV] (acc. IPCC, 1994, p. 1.21): 44.8 GJ/ton gasoline and 43.33 GJ/ton diesel oil [LHV].
Average conversion factor: (44.8*513+43.33*428)/941 = 44.129 GJ/ton; subsequently: 0.85/44.129 = 1.9260e+04 and 1.965e+04 for bunker fuels (1.926 + 2%).

[5] According to IPCC, 1994, Marland and Pippin, 1990, used 15.3 kg C/GJ LHV. We used 13.7/0.9 = 1.522 and rounded it off to 15.3 C/GJ LHV to comply with Marland and Pippin, 1994; OECD, 1991; and with IPCC, 1994.

[6] Based on Marland and Rotty, 1984.

Biofuel combustion (IPCC sector 1A6)

Biofuels include wood, wood waste, charcoal, dung, crop residue, bagasse (a crop residue, but separately identified), ethanol and the IEA categories black liquor (an industrial waste, but separately identified), non-solid fuels (non-specified), industrial and municipal waste.

The activity levels for total biomass use per country are from Hall *et al.* (1994), except for some industrialised countries [IEA statistics and as secondary source the PHOXA report by Veldt (1994), and Leach (1988) for countries in the Middle East], resulting in a global consumption estimate of 50 EJ for 1990. The subdivision of the total biomass consumption in the different biofuels is based on country studies, IEA and OLADE statistics. Where no subdivision was available for a country, the subdivision of a neighbour with the same kind of vegetation was used. Total biomass consumption was also split in residential and industrial use based on a number of country studies and OLADE statistics. Again, when no subdivision was available, the division of a neighbouring country was used. Olivier *et al.* (1996) provides more details on the assumptions made for the sectoral and fuel split per country.

The emission factors for the biofuels are: for CO_2 from fuelwood 450 g C/kg fuel based on IPCC (1994) and 15 MJ/kg Lower Heating Value (air dry; 20% moisture), resulting in (rounded-off) 30 kg C/GJ (Hall *et al.*, 1994). For combustion of charcoal, which has a much higher carbon content per kg, the same factor is used, since the emission factor expressed as kg C/GJ is almost the same as the factor for fuelwood.

Of course net CO_2 emissions depend on the degree of sustainable production of fuelwood etc. As a first estimate it was assumed that the biofuel consumption represents a 100% extraction without any replacement. A second estimate was only 10% unsustainable production, which is extraction without replacement.

Industrial processes (IPCC sector code 2)
EDGAR categorises this source sector in about ten subsectors as shown in Table 2.7.

Table 2.7 EDGAR standard reporting sectors for industrial processes/solvent use

EDGAR sector	Code	Remark
I10. Iron & Steel	IRO	Production of pellets, pig iron, sinter, and steel (per process); hot and cold rolled steel; excluded is coke production (see fossil fuel sectors)
I20. Non-ferro metals	NFE	Production of copper, lead, zinc, aluminium, and molybdenum (primary and secondary)
I30. Chemicals	CHE	Production of organic and inorganic bulk chemicals (e.g. adipic acid, ammonia, nitric acid, sulphuric acid, N-fertilisers, polymers, monomers, etc.)
I40. Building materials	NME	Cement production
I50. Pulp & Paper	PAP	(not included in V2.0).
I60. Food	FOO	Bread, beer and wine production (bread not included in V2.0).
I70. Solvent use	SOL	Divided in 12 categories of solvent applications (chemical industry, paints, dry cleaning, degreasing, glues and adhesives, graphic arts (ink), leather, pesticides, rubber and plastics industry, vegetative oil extraction, household products, other solvent use)
I80. Transport evaporation	EVA	Evaporation of gasoline vehicles in road transport: in V2.0 included under combustion emissions
I90. Miscellaneous industry	MIS	Includes miscellaneous processes, not related to a specific type of industry; includes production and consumption of halocarbons and related compounds

Most industrial production data were taken from UN (1993/1995), since it provides a time series from 1970 to 1990, except for a few products in the iron & steel industry which were taken from IISI (1994). For many commodities the time series were not complete up to 1990. In those cases we extrapolated at maximum five years backward and forward in time in esti-

mating missing values, for 1990 mostly by assuming that the last known production level was kept constant in time. Also for solvent use and for the production of a number of chemical products no UN data were available. Instead, production data for 1990 were compiled from various sources.

The emission factor of CO_2 for cement production (136 kg CO_2-C/ton cement) is from Marland and Rotty (1984). Since cement production, rather then clinker production, is used as activity data, emissions of countries with high fractions of clinker import or export are uncertain.

Landuse (Agriculture and Deforestation, IPCC sectors 4 and 5A) and Waste Treatment (IPCC sector 6)
Landuse and waste treatment sources in EDGAR include rice paddies (CH_4), fertiliser use (N_2O), animals (CH_4 and N_2O), biomass burning (all gases), agricultural waste burning and landfills (CH_4). Waste water and sewage treatment, which are considered to be sources of methane, are not included because to date no representative spatial emission estimates exist.

Biomass burning consists of large-scale biomass burning (deforestation and savannah burning) and local fires of agricultural waste burning. Base levels for large-scale burning include the amount of carbon released in the tropics compiled by Hao *et al.* (1990) as distributions on a $5^{\circ}x5^{\circ}$ grid, based on FAO statistics for the period 1975-1980. These distributions were converted to the EDGAR $1^{\circ}x1^{\circ}$ grid and used as base level for calculation of 1990 emissions. Thus, in EDGAR V2.0 this source is not accounted for in OECD countries, Eastern Europe or the former USSR.

For CO_2 from large scale biomass burning only deforestation is accounted for. Carbon losses from savannah burning and agricultural waste burning do not contribute to net emissions, since the vegetation is regrown in an average time period of one to two years.

2.4.6 Global total comparison
Total CO_2 emissions from fossil fuel combustion in EDGAR amount to: 20694 Tg CO_2 in 1990. The EDGAR total from non-combustion (feedstocks) amounts to: 1206.8 Tg CO_2. CO_2 emissions from biofuel combustion is 5456.3 Tg if 100% unsustainable biofuel use is assumed (no regrowth or reafforestation for biofuel). It is 545.6 Tg CO_2 if only 10% unsustainable biofuel production is assumed. Industrial processes (cement) emits 570 Tg CO_2 and deforestation 1837.4 Tg CO_2. This brings the grand total to 29765.3 Tg CO_2 in 1990 if 100% unsustainable biofuel use is assumed, and 24854.6 Tg if only 10% unsustainable biofuel use is assumed. A grand total for country studies can not be made yet because only part of the world has submitted inventories to the Climate Convention. A partial comparison however can be made: 19502.8 Tg CO_2 is reported from the Climate Secretariat plus the US country studies (own calculations). In table 2.8 an overview of differences is given for OECD and Eastern European countries because only for these countries official data were available in the first phase of the study.

Table 2.8 Total anthropogenic CO_2 emissions in 1990 (10^6 kg CO_2) in OECD and Eastern European countries. Comparison of official National Communications and EDGAR data.

Million kg CO_2	Total official excl. Land use	Total EDGAR excl. Land use	Difference abs.	Difference %
Australia	288965	288994	29	0
Austria	59200	72665	13465	19
Belgium	114410	111233	-3177	-3
Bulgaria	82990	72899	-10091	-14
Canada	462643	520388	57745	11
Czech Republic	165792			
Slovak Republic	58278			
Czechoslovakia	224070	224176	106	0
Denmark	52025	63489	11464	18
Estonia	37797			
Finland	53900	81377	27477	34
France	366536	386865	20329	5
Germany	1014155	1007484	-6671	-1
Greece	82100	78374	-3726	-5
Hungary	71673	69511	-2162	-3
Iceland	2172	1905	-267	-14
Ireland	30719	35302	4583	13
Italy	428941	425342	-3599	-1
Japan	1155000	1072304	-82696	-8
Latvia	22976			
Liechtenstein	208			
Luxembourg	11343	10199	-1144	-11
Monaco	71			
Netherlands	167600	149938	-17662	-12
New Zealand	25476	32723	7247	22
Norway	35514	52890	17376	33
Poland	414930	372943	-41987	-11
Portugal	42148	46234	4086	9
Romania	171103	169030	-2073	-1
Russian Federation	2388720	3480756	1092036	31
Spain	227322	222146	-5176	-2
Sweden	61256	104897	43641	42
Switzerland	45070	46832	1762	4
United Kingdom	577012	581345	4333	1
United States of America	4957022	5295094	338072	6

2.4.7 Country comparison

Preliminary country comparisons show that differences between EDGAR and national inventories for carbon dioxide are generally large with some exceptions. Differences of more than 5% in carbon dioxide emissions in country or sectoral totals will be examined more closely. However an in-depth review of the differences is outside the scope of this study.

In figure 2.17 and table 2.9 an overview of differences is given for regions in the world. Differences between EDGAR and National Communications were small for these aggregated data. Differences were more than 5% in the following regions: Eastern Europe + former USSR, OECD excluding the European Union, and the Rest of the World Group 1, for which US country study inventories were available.

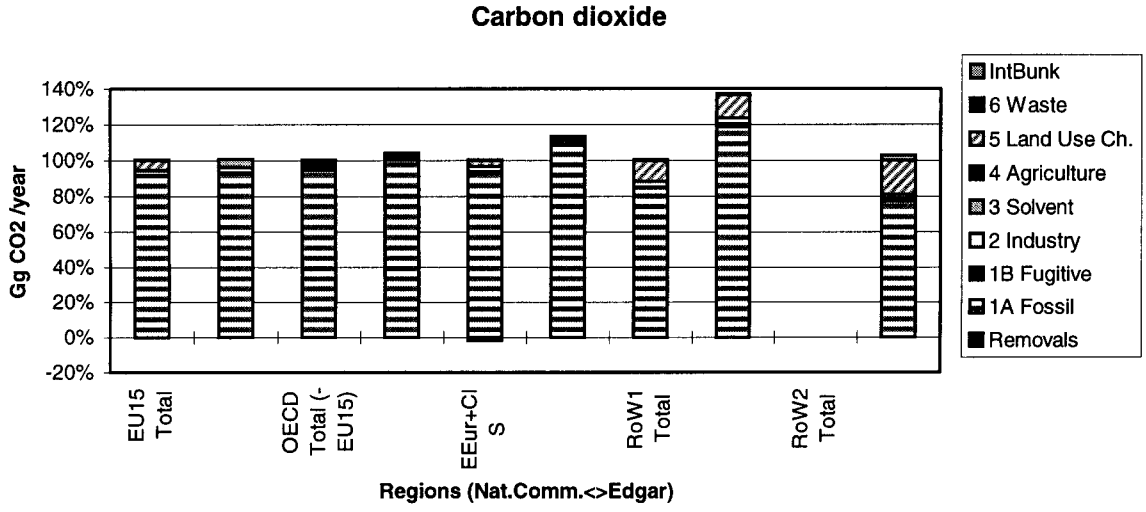


Figure 2.17 Comparison of CO_2 emissions from National Inventories with EDGAR for regions in the World.

In the above graph the first bar is the sum of anthropogenic CO_2 emissions from official National Inventories of 15 European Union countries. The second bar is the sum of anthropogenic CO_2 emissions from EDGAR data of the same countries. Third bar is rest of OECD. Fourth bar is EDGAR for the same countries, etc. Sum of national inventories of a region is set to 100%. The difference of EDGAR compared to National Inventories is given in percentages.
RoW1 is rest of the World 1. This is a group of countries for which data are available from the US country studies programme. RoW2 is rest of the World 2. This is a group of countries for which only EDGAR data are available.

Table 2.9 Comparison of CO_2 emissions for regions in the World as reported in National Communications and EDGAR. (Rest of World 1: results from US-CSP. Rest of World 2: only EDGAR results.)

		Categories	CO_2 Emissions in Gg/year						in percentages	Nat Comm is 100%							
Source	Group	of IPCC 1A CO2 from Fossil fuel combustion	1B CO2 Fugitive emissions	2 Industry emissions	4Agriculture	5 Land Use Change	6 Waste	International Bunkers	Total CO2 emission	1A CO2 from Fossil fuel combustion	1B Fugitive	2 Industry	3 Solvent use	4 Agriculture	5 Land use change	6 Waste	International Bunkers
Nat Comm	EU15 Total	3216069	6424	97738	3244	193823	2937	6424	3520528	91%	0%	3%	0%	0%	6%	0%	0%
Edgar	EU15 Total	3270636	9488	96765		0		164061	3376889	93%	0%	3%	0%	0%	0%	0%	5%
Nat Comm	OECD Total	6940705	27147	149322	7185	175220	46074	27147	7345791	94%	0%	2%	0%	0%	2%	1%	0%
Edgar	OECD Total	7377877	30465	102663		0		132180	7511004	100%	0%	1%	0%	0%	0%	0%	2%
Nat Comm	Eeur+fUSSR	3797001	18350	112089	0	135274	1715	18350	4064428	93%	0%	3%	0%	0%	3%	0%	0%
Edgar	Eeur+fUSSR	4411711	24843	93127	0	0	0	86956	4529680	109%	1%	2%	0%	0%	0%	0%	2%
Nat Comm	RoW1 Total	3811740	10059	147157	0	520133	581	10059	4489670	85%	0%	3%	0%	0%	12%	0%	0%
Edgar	RoW1 Total	5329144	73078	147049		566023		41127	6115295	119%	2%	3%	0%	0%	13%	0%	1%
Nat Comm	RoW2 Total								6665550	0%	0%	0%	0%	0%	0%	0%	0%
Edgar	RoW2 Total	5141281	122034	130489		1271747		195994	6665550	77%	2%	2%	0%	0%	19%	0%	3%

Eastern Europe and former USSR.

In EDGAR the quantity of fuels combusted is taken from IEA statistics. National statistics may differ and cause differences in emission estimates. In Eastern Europe and the Russian Federation the base year for the inventories was 1988. EDGAR estimates are higher for 1990 than the country estimates for 1988. After the collapse of the communist system in these countries, the economy collapsed also. National statistics bureaux are now changing their methods to establish comparability with western statistics. Apart from the different base year this may be the main reason for differences at this moment. Also, there is uncertainty about the energy content of solid fuels (e.g. the IEA conversion factor is 14% lower than the UN factor, see Von Hippel *et al.* 1993). In Figure 2.18 the results are given.

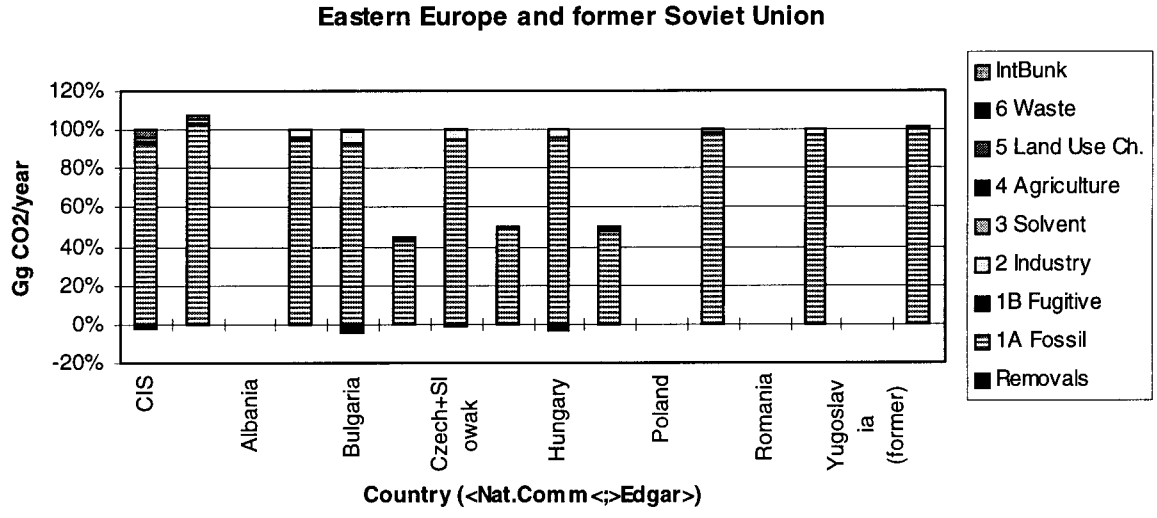


Figure 2.18 Comparison of CO_2 emissions from National Inventories with EDGAR for Eastern Europe.

First bar is National Inventory, second bar is EDGAR data for the same country. Missing bars mean the National Inventories are missing.

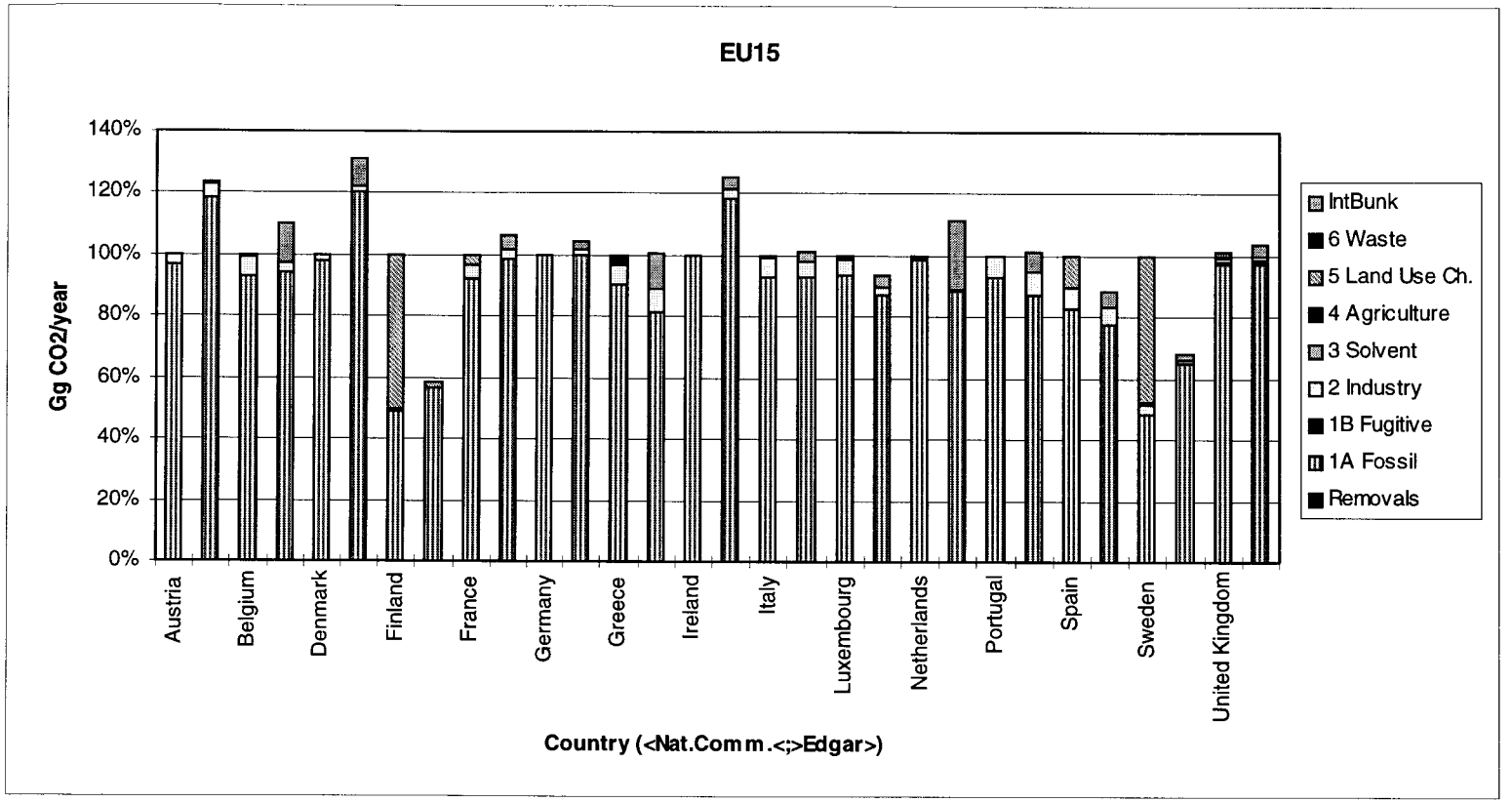


Figure 2.19 Comparison of CO_2 emissions from National Inventories with EDGAR data for EU-15 countries.

First bar is national inventory. Second bar is EDGAR data for the same country. National Inventory is 100%. Bunker emissions are not included in national totals. Sectors are: CO_2 emissions from international marine and aviation bunker fuels; CO_2 emissions from waste incineration; CO_2 emissions from land use change; CO_2 emissions from agriculture; CO_2 emissions from solvent use; CO_2 emissions from industrial processes like cement clinker production; CO_2 emissions from fugitive emissions from oil and gas wells; CO_2 emissions from fossil fuel combustion (largest source); CO_2 removal by storage in vegetation by increase in forest stock (not in this figure).

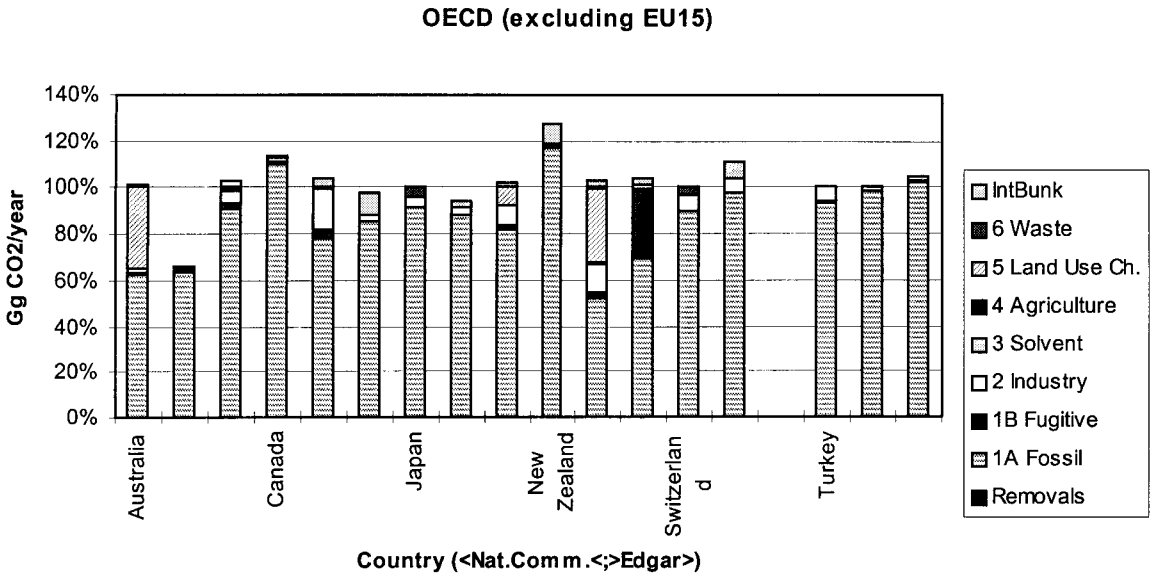


Figure 2.20 Comparison of National Inventory with EDGAR for the rest of OECD countries.

In the above graph the first bar is the national inventory. The second bar is the EDGAR data for the same country. The national Inventory is assumed to be 100%. Bunker emissions are not included in national totals. Sectors are: CO_2 emissions from international marine and aviation bunker fuels; CO_2 emissions from waste incineration; CO_2 emissions from land use change; CO_2 emissions from agriculture; CO_2 emissions from solvent use; CO_2 emissions from industrial processes like cement clinker production; CO_2 emissions from fugitive emissions from oil and gas wells; CO_2 emissions from fossil fuel combustion (largest source); CO_2 removal by storage in vegetation by increase in forest stock (not in this figure).

Comparison for the European Union.

As a whole the difference between National Communications and EDGAR in the EU15 is smaller than 5%. However in most individual countries within the EU15 differences are higher: between 5 and 10%. Countries with differences smaller than 5% are Belgium, Germany, Italy and the United Kingdom.

Comparison for the rest of the OECD countries excluding EU15.

Some countries show large differences in CO_2 emissions from combustion 1A when comparing National Communications with EDGAR data. Australia, Japan and the United States of America have differences smaller than 5%. Some countries show large differences in CO_2 from land use change.

Comparison for the Rest of the World.

For the countries that have participated in the US Country Studies Programme a comparison is possible with EDGAR data. Large differences are encountered in sector 1A CO_2 from combustion. Countries that show small differences are: Ghana, Mexico and Zimbabwe. Possible reasons for differences can only be analysed when detailed studies will become available on activity data and emission factors. A first guess is that CO_2 from biomass for energy is included in EDGAR where this should not have been done. In addition some inconsistencies are expected with the reporting of CO_2 emissions from waste incineration for energy purposes.

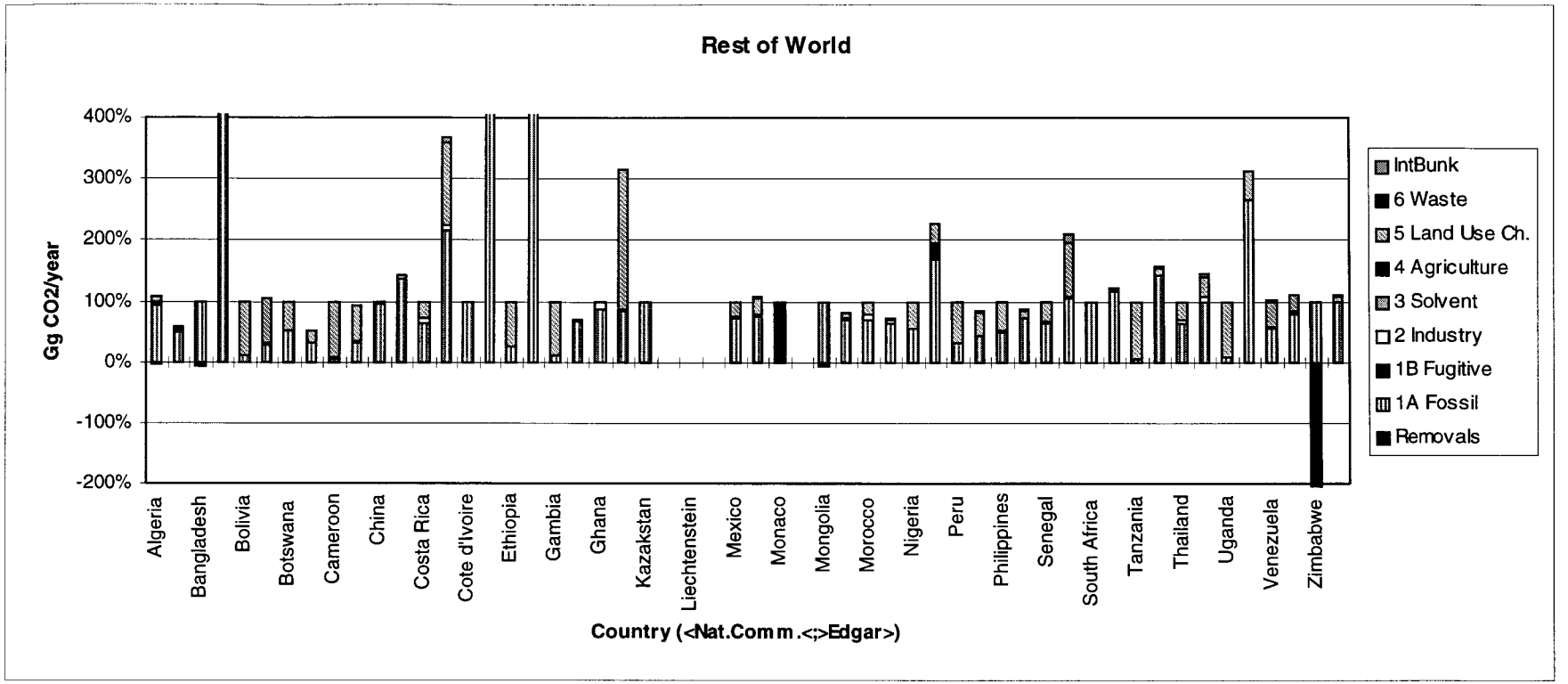


Figure 2.21 Comparison of CO_2 emissions from National Inventories with EDGAR for the Rest of the World-1.

In the above graph the first bar is the national inventory. The second bar is the EDGAR data for the same country. The National Inventory is assumed to be 100%. Bunker emissions are not included in the national totals. Sectors are: CO_2 emissions from international marine and aviation bunker fuels; CO_2 emissions from waste incineration; CO_2 emissions from land use change; CO_2 emissions from agriculture; CO_2 emissions from solvent use; CO_2 emissions from industrial processes like cement clinker production; CO_2 emissions from fugitive emissions from oil and gas wells; CO_2 emissions from fossil fuel combustion (largest source); CO_2 removal by storage in vegetation by increase in forest stock: Large in Zimbabwe in this graph.

CO_2 from fugitive emissions (IPCC sector 1B)
EDGAR reported CO_2 emissions for Nigeria and Norway. These emissions could not be found in National Communications. This CO_2 is stripped from natural gas during the production process.

CO_2 from industrial processes (IPCC sector 2)
Differences between EDGAR and National Communications were more than 5% in the following countries: Bulgaria, Iceland, New Zealand, Norway, Cote d'Ivoire and Ghana. With the exception of Cote d'Ivoire National Communications reported higher emissions. EDGAR is underestimating these emissions, because they were based on cement production (no other industrial processes with lime addition). EDGAR uses cement production rather than clinker production, thus neglecting international trade of clinker. No other industrial processes with lime were estimated in EDGAR.

CO_2 from land use change (IPCC sector 5)
The differences in this sector between EDGAR and the National Communications are caused by countries applying the IPCC methodology in different ways. In EDGAR no emissions are reported from this sector outside the tropics.

CO_2 from international bunkers
This is not reported in all National Communications. In EDGAR IEA data have been used to estimate emissions from international marine and aviation fuel bunkering.

Latitudinal distribution
For comparison with Top-down model results here a latitudinal distribution is given of EDGAR CO_2 emissions.

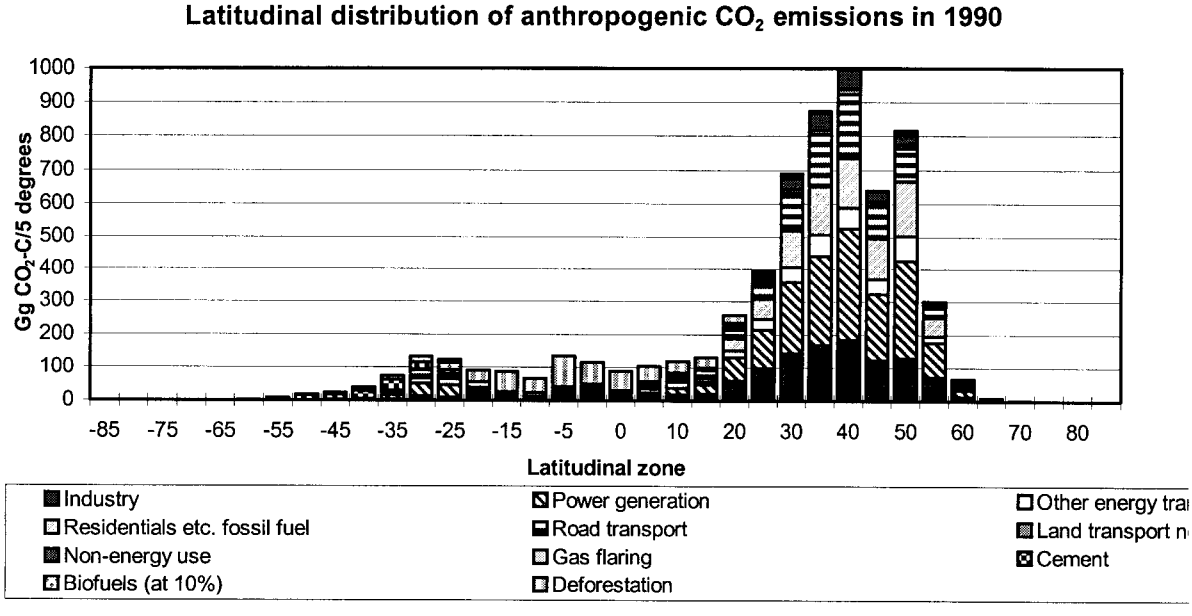


Figure 2.22 Latitudinal distribution of CO_2 emissions in 1990 according to EDGAR calculations.

2.5 Conclusions on the comparison of top-down with bottom-up emission inventories for carbon dioxide

The research questions as formulated in chapter 1.2 will be answered for CO_2 in the following:

Available inventories
For CO_2 different model studies are available. Here we have used the global model studies by Tans *et al.* (1989; 1990), Keeling *et al.* (1989), Ciais *et al.* (1995, 1997), and Conway *et al.* (1994). Two databases of national inventories have been used: the Climate Secretariat database and EDGAR. The US country studies results summarised by Braatz *et al.* (1996) were incorporated.

Presentation of inventory data
National inventory data are presented per sector per year according to the IPCC recommended summary table format. EDGAR data are presented per sector per year according to own sector definitions. These could however be translated easily to IPCC recommended format.

A method for comparison of emission inventories
Concerning the top-down bottom-up comparison of emission inventories for CO_2, it can be concluded that a global comparison is possible, but by doing this the uncertainty in national inventories can not be reduced. Because global carbon cycle models ignore short term perturbations due to fluctuations in the climate system they are not able to reproduce these shorter-term atmospheric CO_2 variations. It is therefore not possible to estimate fossil fuel emissions derived from atmospheric measurements and global CO_2 (Top-down) budget calculations for a year with more accuracy than based on Bottom-up emission inventories. The uncertainty in the estimated CO_2 emission due anthropogenic activities derived from atmospheric measurements is 25-30%. The uncertainty in the CO_2 emissions from fossil fuel combustion derived from national inventories is 10%.

Comparisons of bottom-up emission inventories like the comparison of national inventories with EDGAR are feasible and useful. Country results can be compared and groups of countries can be compared. Here the following groups were used: European Union, other OECD, Eastern Europe and former USSR, Rest of World 1 and Rest of World 2.

Uncertainties
The 4 main sources of uncertainty for CO_2 budgets are:
a) The measuring dataset and representativeness of the measuring data, because of poor coverage of the World.
b) Whether the ocean or the biosphere is the main sink.
c) Yearly variations in sources and sinks
d) Different budget assumptions between various authors

Due to large year to year variations in net fluxes a zonal comparison of top-down model results with bottom-up inventories for CO_2 for one year can not lead to validation of these bottom-up inventories. Yearly variations in the CO_2 budget are much larger than the yearly variations in the fossil fuel emissions. Long term trends of the CO_2 concentration in the atmosphere are representative of the long term disturbance of the carbon balance due to antropgenic CO_2 emissions.

Preliminary comparison
The country comparison with EDGAR showed that differences are large in all important sectors, even in Europe, although the totals match more closely. The differences were large especially in the land use change sector, the agriculture sector and the biofuels combustion sector.

Recommendations for methodology development
In this preliminary study we did not investigate the reasons for observed differences in detail. Clearly, a further in depth analysis is needed. . The recommended format for the comparison is the summary tables 7A and 7B of the IPCC Guidelines. The comparison of national inventories with EDGAR data has targeted some areas for potential improvement in the IPCC methodology for CO_2, namely the land use change sector, the agriculture sector and the biofuels combustion sector. The comparison of national inventories with EDGAR data will be completed in the second phase of the project when overviews of activity data and emission factor data are made.

3. Analysis for methane

3.1. Introduction

Methane is the second important greenhouse gas. Although the emissions are smaller than from CO_2, the greenhouse warming potential is high: 21 times CO_2 over 100 years. Methane inventories are numerous. A GEIA methane inventory is in preparation. This sectoral GEIA inventory will include the EDGAR results for fossil fuel combustion, biofuel use and industrial processes only. Other global inventories for 1985 were compiled by Fung *et al.* (1991). The comparison of Top-down with Bottom-up emission inventories is based on Bottom-up national inventories as summarised in data tables to the Second compilation and synthesis of the Climate Convention Secretariat (UNFCCC/CP/1996/12/Add. 2, 2 July 1996), the US country studies programme as summarised by Braatz *et al.* (1996) and the EDGAR database (Olivier *et al.*, 1996). Top-down modelling results have been used from Lelieveld and Crutzen (1993), Muller (1993), Fung *et al.* (1994), Hein *et al.* (1994; 1997), The and Beck (1995), Saeki *et al.* (1997). For local validation of national inventories in NW-Europe top-down model results have been used from Hollander and Vosbeek (1996), Veltkamp *et al.* (1995), Vermeulen *et al.* (1996, 1997), Janssen *et al.* (1997), Stijnen *et al.* (1997) and Van der Wal *et al.* (1997).

3.2. Monitoring concentrations in the atmosphere

The atmospheric distribution of greenhouse gas concentrations reflects the geography of its sources and sinks. This is especially the case for methane. Broad regions of elevated methane concentrations exist over strong source regions because the distribution is not completely homogenised by atmospheric circulation (Steele *et al.*, 1987). Mathematical models of tracer transport in the atmosphere have been used to deduce objectively, from spatial and temporal distributions of atmospheric concentrations, not only the total annual emissions and destruction of methane, but even rough seasonal and geographic distributions for its sources and sinks (Taylor *et al.*, 1991; Fung *et al.*, 1991; Brown, 1993). For north-west Europe this is done by Zhang (1996). Thus, precise measurements of atmospheric greenhouse gases at carefully selected sites over the globe can provide valuable information for verification of total emissions from a particular region.

While spatial variations of atmospheric concentrations of greenhouse gases are related to regional source strengths, there is a limit to the information about the distribution of sources that can be extracted from these variations. Atmospheric transport models yield greenhouse gas source/sink information that has been integrated over a few weeks and a few hundred or thousand kilometres. Individual sources and sinks cannot be differentiated. For example, once methane emitted in the United States from livestock, landfills, coal mining, oil and gas production and distribution systems mixes during transport to a monitoring site in the North Atlantic Ocean, concentration measurements at the monitoring site will provide information only about the sum of emissions from these sources. Therefore, the attribution of an observed concentration profile to a particular distribution of sources must make use of constraining data other than atmospheric concentrations, e.g. isotope studies (EPA, 1994).

3.2.1 Available measurements and techniques

Measurements of atmospheric methane concentrations are obtained by analysis of air samples collected at regular intervals from sites established for long-term monitoring. They are occasionally also obtained during transect measurements by aircraft or ships. Samples are usually transported to a laboratory, where they are analysed for greenhouse gas content. Gas

chromatography with flame ionisation detection is a widely accepted method and it yields a precision of 0.5% or better (Steele *et al.*, 1987). Infrared absorption methods can also be used, although they are less precise. The advantage is that it is a fast method so that a high frequency of measurements can be made, which makes this technique particularly useful for measuring methane concentrations using aircraft.

The existing network of long term monitoring sites established by the Climate Monitoring and Diagnostics Laboratory of the National Oceanic and Atmospheric Administration (about 30 sites of NOAA/CMDL) was designed to represent concentration levels in clean air (background concentrations, rather than sites with anthropogenic sources). This strategy was formulated in 1985 (Komhyr *et al.*, 1985). The purpose was to capture the broad scale hemispheric and latitudinal gradients in atmospheric trace gas concentrations. Recent efforts in the ALE/GAGE (atmospheric lifetime experiment/global atmospheric greenhouse gas experiment) network of the MIT (Ronald Prinn) have focused on the addition of sites to narrow the estimates of contributions from various sources and of removals by sinks. For example, data from around the Pacific rim will not quantify individual sources, such as methane from rice in China, but will put constraints on the total regional strength of all sources from the Asian continent (Dlugokencky *et al.*, 1993).

3.2.2 Measurements from space

Satellite measurements of methane in the troposphere are not available and measurements in the upper stratosphere have no meaning for global budget studies. An instrument named MOPITT (measurements of pollution in the troposphere) proposed for the Earth Observing System by Drummond (1992), is able to measure methane concentrations near the ground. However, this instrument will not be launched until 1998. MOPITT will provide repeated global coverage, reveal 'hot spots', and identify the scattered source regions. This information can then be used to test the accuracy of national emission inventories, as well as cross-check tracer model inferences of regional sources and sinks.

3.2.3 Maintenance and calibration of standards

Methane concentration measurement techniques, such as gas chromatography with flame ionisation detection, make use of 'standards' of known methane concentration. Infrared absorption methods frequently make use of a reference gas to improve the accuracy of the measurements. Usually working standards are prepared in individual laboratories from natural air and are calibrated against a primary set of standards maintained in the laboratory. There are no formal mechanisms for intercalibration of these standards among different laboratories, although informal comparison studies have been carried out. To improve the measurement accuracy and comparability it is important that an international set of primary standards of known methane concentrations covering the range expected near ambient levels be established and maintained (EPA, 1994). From these primary standards 'secondary' standards would be propagated for use throughout the international scientific community. The precision of the secondary standards should be ± 0.5 ppbv.

3.2.4 Sampling and reporting strategy

For atmospheric data to be useful for verifying annual methane emissions from a region, the measurements should be made with adequate temporal and spatial coverage. Measurements should cover at least a full seasonal cycle. Results so far have indicated that seasonal cycles differ by location. For example at Cape Grimm, Tasmania, the seasonal cycle consists of a winter maximum and a summer minimum. At Point Barrow, Alaska, however, there are two concentration peaks; one in February and one in October. A full year of data is necessary to determine the annual mean at each station. The annual means at all stations are used to calculate the latitudinal distribution of the atmospheric concentration, an important constraint

on source/sink locations in tracer models. During measurements, meteorological data have to be recorded as well. Trajectory analysis can only be carried out if detailed information is available on the mixing layer height and the origin of the air masses. Weekly measurements do not suffice for regional studies in the more polluted areas. Semi-continuous measurement strategies should be developed similar to the one used in Cabauw, the Netherlands (Vermeulen *et al.*, 1996).

Methane data can be collected and catalogued by the World Meteorological Organisation's archive in Japan, but regional centres should be chosen for ready availability of results to other researchers. The regional centres could maintain Web sites and FTP sites to improve the dissemination of results to interested parties (Janssen *et al.*, 1997).

3.2.5 Isotope studies

The isotopic ratio of greenhouse gases in the atmosphere is one constraint used to distinguish between source groups (Tyler, 1986; Quay *et al.*, 1991; Levin, 1996). Analysis of the radiocarbon content (^{14}C) of greenhouse gases provides a means of estimating fossil age versus present day carbon dioxide and methane, while its stable isotopic composition (the ratio of carbon ^{13} to carbon ^{12}: $^{13}C/^{12}C$) provides a means to separate biogenic from non-biogenic sources. ^{14}C is a radioactive isotope, with a half-life of 5700 years. As long as an organism is alive, it exchanges carbon with its surroundings, and its ^{14}C content equals that of atmospheric carbon dioxide. Once an organism dies, absorption of ^{14}C ceases, and the residual ^{14}C continues to decay. The longer an organism has been dead, the lower its ^{14}C content. The formation of fossil fuels occurs on geologic time scales, so that methane associated with these sources (venting and leaking from oil, gas, coal) contains no ^{14}C. The ^{14}C content of atmospheric methane can place bounds on the emissions associated with fossil fuel production and use (Wahlen *et al.*, 1989; Quay *et al.*, 1991; Vermeulen *et al.*, 1996).

According to Mook (1996) the standard marine limestone Vienna PeeDee Belemnite has the following isotopic ratio: ^{12}C = 98.9% and ^{13}C = 1.1%. $^{13}R = {}^{13}C/{}^{12}C = 0.01$. Instead of ^{13}R, isotope ratios are given relative to a reference: $^{13}\delta = {}^{13}R\text{sample}/{}^{13}R\text{reference} - 1$ in promille. The reference for stable carbon isotopes is a marine limestone: Vienna PeeDee Belemnite with ^{13}R=0.0112372. Measurement accuracy is ^{13}R = 0.0000003 and $^{13}\delta$ = 0.03 promille. Some $\delta^{13}C$ values are:

wetland methane	= -70 promille
fossil fuel	= -35±10
vegetation land	= -25
groundwater HCO_3^-	= -11
travertine	= -10
sugar cane/maize	= -10
atmospheric CO_2	= -8
marine CO_2	= -7
marine HCO_3^-	= +1
marine limestone/marble	= +2

Together, radiocarbon and stable isotope measurements of atmospheric methane can narrow uncertainties about source strengths. For example, if a methane sample has a relatively low $^{13}C/^{12}C$ ratio, but contains significant ^{14}C, it is almost certainly of biogenic origin. Methane from biomass burning would also have a significant ^{14}C content, but would have a relatively high $^{13}C/^{12}C$ ratio. If, however, a methane sample contains no ^{14}C, it is of fossil origin. Data on methane isotopes are thus a critical supplement to methane concentration measurements for identifying and verifying the location and magnitudes of individual methane sources or groups of sources (Braatz *et al.*, 1994).

Measurements of the $^{13}C/^{12}C$ isotopic ratio of atmospheric methane can place bounds on biogenic and non-biogenic source strengths (Tyler *et al.*, 1988; Vermeulen *et al.*, 1997). Sources of biogenic methane, produced by bacteria during anaerobic fermentation, include wetlands, wet rice cultivation, livestock, solid and liquid wastes and oceans and fresh waters. Non biogenic methane is derived from thermal alteration of buried organic matter (fossil sources) and from incomplete combustion during biomass burning. Methane from biogenic sources has the lowest, or most negative, $^{13}C/^{12}C$ values, while methane from thermogenic sources has mean values, and methane from biomass burning has the highest ratio (Quay *et al.*, 1991). Therefore, the $^{13}C/^{12}C$ ratio of atmospheric methane reflects the input from biogenic versus non-biogenic sources. The $^{13}C/^{12}C$ isotope ratio of atmospheric methane is also a function of sink strength because oxidation rates vary between $^{13}CH_4$ and $^{12}CH_4$. Next to the isotope ratio of carbon the isotope ratio of oxygen can provide information on the origin of carbon dioxide and water.

According to Mook (1996) the Vienna standard mean ocean water has the following isotopic ratio: $^{16}O = 99.8\%$, $^{17}O = 0.035\%$ and $^{18}O = 0.2\%$. The isotopic ratio $^{18}R = {}^{18}O/{}^{16}O = 0.002$. Instead of ^{18}R, $^{18}\delta$ is used as a measure. $^{18}\delta = {}^{18}Rsample/{}^{18}Rreference-1$. The reference for water is Vienna standard mean ocean water with $^{18}R = 0.0020052$. The reference for CO_2 and other gases as with methane is Vienna PeeDee Belemnite $^{18}R = 0.0020790$. The measurement accuracy of deviations of $^{18}R = 0.0000001$, or deviations of $^{18}\delta = 0.05\%$. Some delta isotopic ratios in promille are given below:

$\delta^{18}O$ ice Antarctica	= -50
$\delta^{18}O$ ice Greenland	= -35
$\delta^{18}O$ atmospheric oxygen and CO2 from fossil fuel	= -17
$\delta^{18}O$ marine water vapour	= -12
$\delta^{18}O$ precipitation the Netherlands	= -7.8
$\delta^{18}O$ marine CO2	= +1.7
$\delta^{18}O$ marine limestone	= +2
$\delta^{18}O$ sea water	= 0 by definition

3.3. Top down modelling

3.3.1 Introduction

In this paragraph inverse modelling will be evaluated as a method to compare the global budgets with national inventories for CH_4. Inverse modelling can be done on different scales: global average emission, emission per period per zonal band or emission per period per grid-cell. Periods can be anything between one hour and one year. Monthly averages are often used in these global models. Regional emissions (on isomaps or grids) derived from global atmospheric concentration fields are then compared with bottom-up estimates on the same spatial scale. Inverse modelling can be an independent check on emission inventories if no a priori emission estimates are used in the chemistry and transport models, however many more stations than the existing ones would then be needed to eliminate the uncertainty. In practice a priori emission profiles through the year are used for each source of emissions to reduce the uncertainty.

3.3.2 Methodology development for the comparison

Atmospheric concentrations of greenhouse gases are derived from global atmospheric chemistry and transport models (Lelieveld and Crutzen, 1993). The models start with "a priori" estimated global emissions of sources of greenhouse gases. These models then calculate the global concentration fields for these greenhouse gases, taking into account atmospheric loss, transport to the stratosphere and an eventual biosphere or soil sink. In the inverse mode of these models, concentration fields determined from measurement stations and interpolation techniques are the starting point and the models calculate the regional emissions. For methane a few inverse modelling attempts have been made and output is given as a zonal average be-

cause measurement locations are too few to give more detailed results. For methane in north-west Europe results from inverse modelling are available at a grid level of detail.

Existing measurement locations from the ALE/GAGE/AGAGE and from the NOAA/CMDL network are selected in remote areas to measure global background concentrations. For inverse modelling more measurement locations are needed, especially in the more polluted continental regions. Existing measurements from stations in Europe are available in a database made at RIVM (Janssen *et al.*, 1997). For methane inverse modelling results are published by Zhang (1996) and Zhang *et al.* (1997). In the second phase of the study methane emissions from zonal bands will be compared.

3.3.3 Global methane budget comparisons

The and Beck (1995) compared existing budgets with the outcomes of the Moguntia 3-dimensional atmospheric chemistry model, originally developed at the Max Planck Institute for Chemistry. Initial CH_4 emissions estimates were adjusted to fit the 1990 data, matching a globally averaged concentration of about 1712 ppbv at surface level and a 0.7% annual net increase of the global burden. The latter corresponds to an atmospheric increase of 12 ppbv per year at surface level. The adjustment was obtained by scaling all methane sources proportionally. The and Beck show in their report a comparison between the measured 1990 concentrations and the initial zonally averaged modelled concentrations. The 1990 concentration levels for methane could be reproduced by assuming a global emission of 539 Tg/yr. Using this initialisation The and Beck calculated a methane lifetime of 8.9 years and a tropospheric content of 4515 Tg for 1990. The resulting budget is compared with other budgets in Table 3.1 below.

Table 3.1 Comparison of global methane inventories in Tg CH_4 per year. (na = not available).

Source	The and Beck 1995	Muller 1993	Lelieveld and Crutzen 1993	Khalil 1992	IPCC 1994	EDGAR 1996	Hein *et al.* 1997
Waste	75	48	40	60	40	36	35 ± 15
Natural oil&gas	65	61	80	30	40	51	46 ± 23
Siberian gas	na	na	na	na	na	na	17 ± 14
Coal	17	16	35	46	30	38	35 ± 10
Animals	85	80	105	94	85	93	90 ± 20
Rice	75	89	95	65	60	60	88 ± 20
Biomass burning	62	58	30	52	40	37	40 ± 12
Wetlands	160	87	125	110	125	na	237 ± 20
Other	na	na	na	na	45	6	na
Total	540	460	560	510	445	na	587
Anthropogenic	380	370	435	400	320	320	350
Fossil methane	82	77	115	76	70	92	97 ± 15

3.3.4 Uncertainty in global budgets for methane

Khalil (1992) developed a statistical method for estimating uncertainties in the total global budgets of atmospheric trace gases, especially methane. Here a short description will be given of his method. The global budgets of trace gases are the balance between the measured concentrations, the emission rates from various man-made and natural sources, and the global removal rates from chemical processes in the atmosphere or deposition on the soils, vegetation, and oceans. Often for each source an emission range is calculated that is consistent with experimental observations. When these ranges are added together to produce a total

annual emission rate from all sources, a range is usually calculated by adding up lower limit emissions and upper limit emissions. This process often leads to an unrealistically large range for the total source even though the uncertainties in emissions from individual sources are justified (Khalil, 1992). When a statistical method is used to evaluate the total uncertainty, the 95% confidence interval of total global emissions from a trace gas, the uncertainty range is often reduced to half the value calculated by adding up ranges. For the methane budgets, in spite of the large uncertainties in emissions from individual sources, the global total emission rates have an uncertainty of only 10 to 20% (95% confidence limit), depending on the budget. When a range and a median value of emissions from a source are quoted, there are three broad possibilities for associating a probabilistic interpretation. The first is that the median is the most likely value and that the extremes of the range are less likely. This is the intuitive notion of ranges as extreme values based on extreme extrapolations or uncommon experimental observations. The second and more conservative assumption is that any value within the specified range is equally likely. Finally, the third possibility is that the middle value of the range is less likely than the extremes. This can be safely abandoned as unrealistic. Therefore Khalil started with the assumption that any value within the specified range is equally likely to occur (uniform distribution). On the basis of this assumption Khalil worked out the probability distributions for the total emissions when uniformly distributed emissions from different authors from literature sources are added. For methane 11 different budgets were found in literature and represented in a figure to illustrate the probability distribution functions of these 11 estimates. No quantitative conclusion on the global budget itself was made.

3.3.5 Temporal and spatial variability in atmospheric CH_4

Measurements and model results are described here to illustrate the temporal and spatial variability of atmospheric CH_4. Global budgets are derived from measurements and model results. The dominant sink of CH_4 is chemical conversion in the troposphere by the reaction with the OH^- radical. Yearly variations in CH_4 concentration will not only be caused by variations in the strength of the OH^- sink but also by variations in the (natural and anthropogenic) sources. Dlugokencky *et al.* (1994) give some descriptive statistics of CH_4 concentrations measured at 37 sites from the NOAA/CMDL network over the period 1983-1992.

Table 3.2 Descriptive statistics of CH_4 concentrations measured at 37 sites from the NOAA/CMDL network over the period 1983-1992 (Dlugokencky *et al.*, 1994).

CH_4	
North-South gradient	140 ppb (pole to pole)
Amplitude seasonal cycle	30 ppb (NH)
Trend	11.1 ppb/yr
Variations in the trend	0 ppb/yr (1992) - 13.5 ppb/yr (1983)

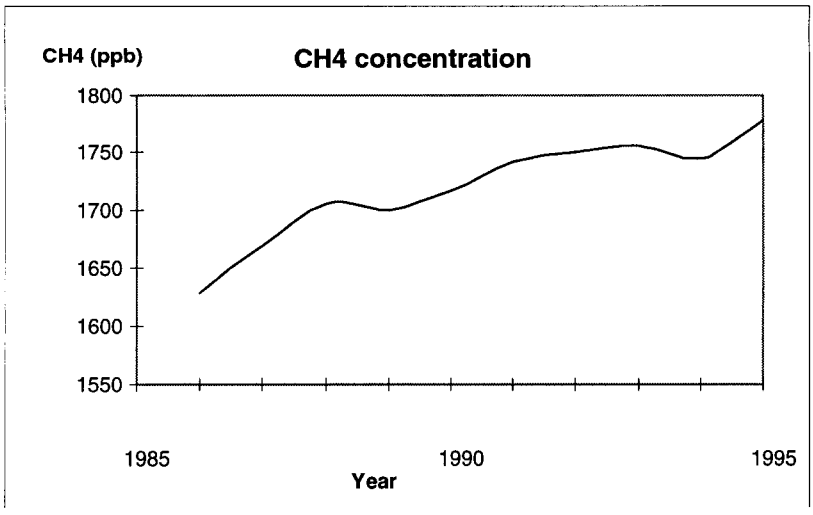


Figure 3.1 Time series of yearly averaged CH_4 concentrations at Barbados 1985-1995 (Dlugokencky *et al.*, 1994)

Short term variations

Contrary to the stratospheric sink for N_2O the tropospheric OH^- sink for CH_4 is not assumed to be constant in time because the OH^- radical concentration varies in space and time. The CH_4 concentration at background stations therefore shows a seasonal variation depending on latitude see figures 3.2 and 3.3. Yearly and monthly variations in the trend are shown in figures 3.4 and 3.5. Yearly Variations of 15 ppb CH4 mean differences in the atmospheric CH_4 content of about 42 Tg CH_4 which is in the order of the atmospheric increase (37 Tg CH_4, IPCC 1995).

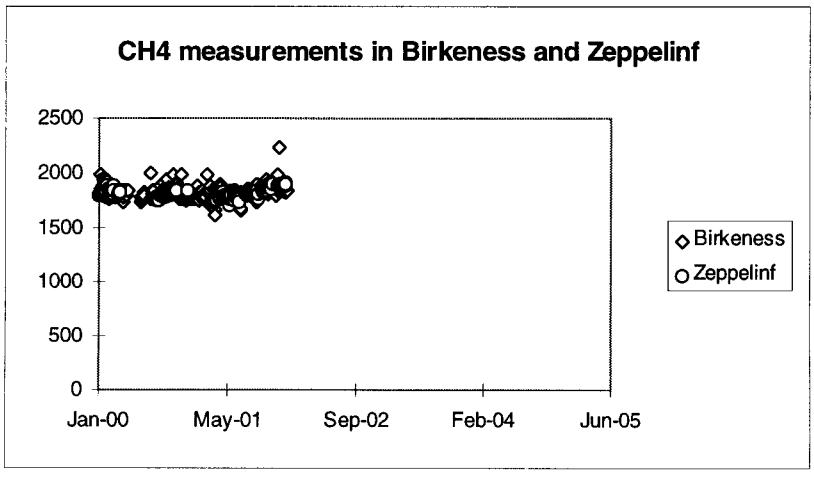


Figure 3.2 Time series of methane concentrations (ppb) measured at Birkeness and Zeppelin-fjellet in 1990-1991.

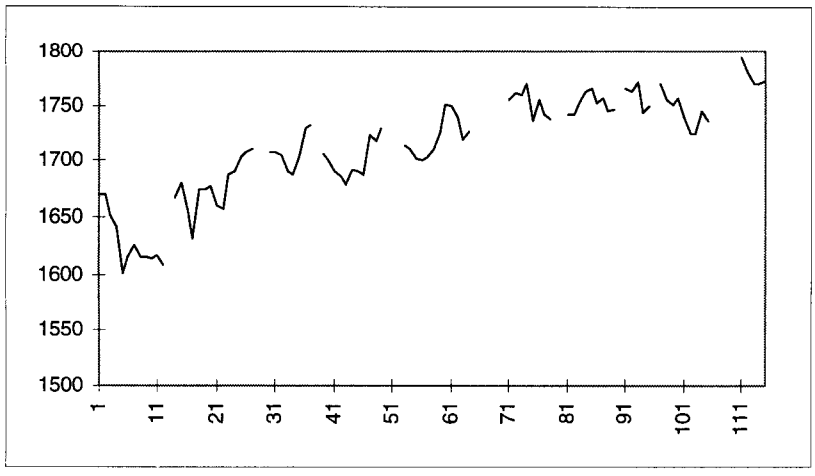


Figure 3.3 Time series of montly averaged methane concentrations (ppbv) at Barbados 1986-1995.

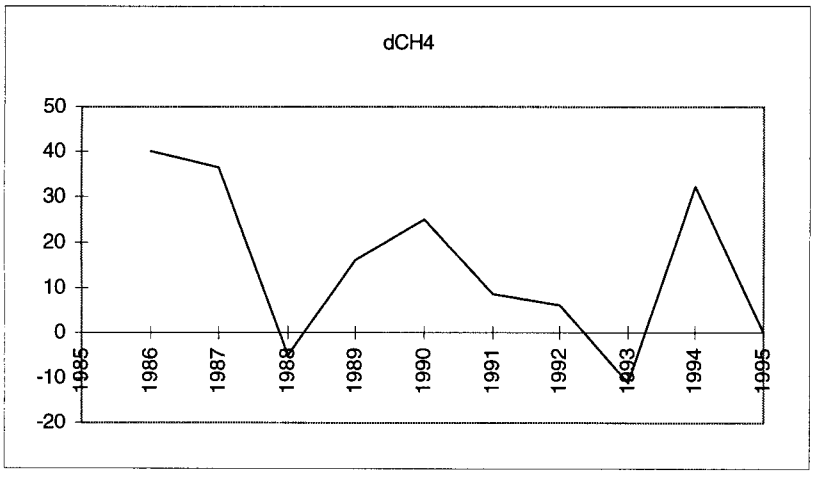


Figure 3.4 Yearly variations in the growth rate of methane (ppb) at Barbados

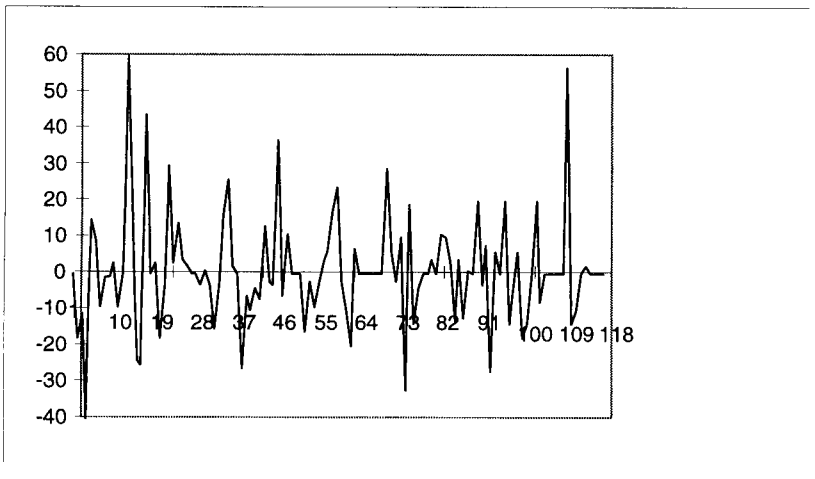


Figure 3.5 Monthly variations in the CH_4 concentration (ppb) . Monthly variations are substantially larger than the variations in the yearly averaged growth rate.

The OH sink and the atmospheric lifetime of CH_4

The reaction rate of CH_4 with the OH radical is not accurately known, this means that the tropospheric lifetime of CH_4 will not be accurately known and the emissions (source strengths) of CH_4 cannot directly be derived from atmospheric measurements. The global emission of CH_4 has recently been adjusted to an updated OH reaction rate and reduced lifetime of CH_4 with an increased global emission strength as result. This difference between the 1990 and 1995 estimate (IPCC 1990 and IPCC 1995) amounted up to 40 Tg CH_4. This can be seen as an uncertainty range in the atmospheric sink term.

3.3.6 Models and measurements used to analyse the atmospheric CH_4 budget

Fung *et al.* (1991), Hein *et al.* (1994; 1997), Taylor *et al.* (1991), Seaki *et al.* (1997), The and Beck (1995) have published analyses of the CH_4 budget based on measurements and model calculations, see table 3.3.

Table 3.3 Models used in the analysis

Model type	spatial resolution	temporal resolution	period	reference
3-D	4^0x5^0 and 9 layers	4-hourly winds	1 year in 1984-1987	Fung *et al.* (1991)
3-D	8^0x10^0 9 layers	12-hourly winds	1987	Hein *et al.* (1994)
3-D	2.5^0x2.5^0 2.5^0 latitudinal bands and 7 layers	12-hourly winds	1984	Taylor (1991)
2-D	20 latitudinal bands of equal area 9 layers	10 days	1984-1994	Saeki *et al.* (1997)
3-D	10^0x10^0	monthly	1987	The *et al.* (1995)

3.3.7 The global budget

Fung *et al.* (1991) modelled seven possible source/sink distributions of CH_4 which compare well with the observations, both in terms of:
- the annual mean north south gradients and
- the seasonal cycle at the stations.

The budget assumptions are shown in table 3.4.

Table 3.4 Different global budgets for CH_4 (in Tg CH_4/yr) that are in agreement with atmospheric measurements (from Fung *et al.*, 1991).

Sources	**Budgets**						
	1	**2**	**3**	**4**	**5**	**6**	**7**
Ruminants	80	80	80	80	80	80	80
Bogs/tundra	80	35	35	35	35	35	35
Swamps	35	70	75	60	55	45	80
Rice	110	100	110	100	100	50	100
Landfills	40	20	20	15	20	40	40
Gas vents	20	80	65	20	80	25	10
Gas leaks	25	40	20	25	15	35	30
Coal	35	35	25	35	40	35	35
Biomass	55	60	50	55	50	100	55
Termites	40	40	40	100	15	40	20
Hydrates	5	5	5	5	0	5	5
Oceans	15	15	15	10	10	10	10
Sinks							
OH	500	500	500	500	450	450	450
Soil	0	40	0	0	10	10	10

N.B. The difference of the total emission strength in the different budgets is 40 Tg CH_4. Differences between OH sink are 50 Tg CH_4.

In table 3.5 budgets are given from various authors to illustrate the problem that from measurements and model results still different budgets can be constructed. This is related to the uncertainties in all terms of the budgets. Recently Hein *et al.* (1997) claim that they have reduced the overall uncertainty in the budget with 10%. Their results are given with the uncertainty ranges in the last column of table 3.5. Their increase in the total emissions compared to earlier budget studies is mainly attributed to an increase in natural wetland swamp emissions. Compared to the early budget of Fung *et al.* (1991) total wetland emissions have increased with 120 Tg/yr to 237 Tg/yr. Other emission estimates have changed as well. Compared to the estimates of Fung *et al.* (1991) ruminant emissions have increased with 10 Tg/yr to 90 Tg/yr, rice emissions are reduced with 20 Tg/yr to 88 Tg/yr, and landfill emissions are reduced with 5 Tg/yr to 35 Tg/yr. Overall total bacterial emissions are increased from 335 to 450 Tg/yr. Total fossil methane emissions are increased from 75 to 100 Tg/yr, the increase is from Siberian gas leaks, estimated at 17 Tg/yr. Total minor sources are decreased from 80 to 40 Tg/yr.

Table 3.5 Differences between the global CH_4 budgets of various authors (in Tg CH_4/yr).

Source	Fung *et al.* (1991)	Saeki *et al.* (1997)	Hein *et al.* (1994)	The and Beck (1995)	Lelieveld and Crutzen (1993)	IPCC 1994	Olivier *et al.* (1996) EDGAR	Hein *et al.* (1997)
Wetlands Bogs/tundra	35		22	160	125	115		44 ± 7
Wetlands Swamps	80		67					192 ± 19
Total natural	**115**		**90**	**160**	**125**	**115**	-	**237 ± 20**
Ruminants	80		70	85	80	85	93	90 ± 20
Rice	100		67	75	70	60	60	88 ± 20
Landfills	40		66	75	40	40	36	35 ± 15
Total bacterial	**335**	**336**	**293**	**395**	**315**	**300**	-	**450**
Oil and gas vents	10		15	65	45	40	26	
Gas leaks	30		58 (incl. Siberia)				26	46 ± 23
Siberian gas								17 ± 14
Coal	35		43	17	35	30	38	35 ± 10
Total fossil	**75**	**79.5**	**115 (incl. oil)**	**82**	**80**	**70**	**90**	**97 ± 15**
Biomass	55	47.5	48	60	30	40	32	40 ± 12
Termites	10				30	20		small
Hydrates	5				10	15		small
Oceans	10			11	10	10		small
Total Other	**80**	**47.5**	**48**	**71**	**80**	**85**	**32**	**40 ± 12**
Total Emissions	**480**	**463**	**456**	**550**	**475**	**455**	-	**587**
OH	500		375	460-485	455	445		489
Soil	10		29	30	30	30		?
Stratospheric loss			16					46
Total Sinks	**510**		**420**	**500**	**485**	**475**		**525**

3.3.8 The zonal budget

Fung *et al.* (1991), Saeki *et al.* (1997) and The and Beck (1995) modelled the CH_4 interhemispheric gradient and derived a zonal budget based on measurements and model calculations. The figures 3.6 to 3.13 show their zonal budgets, interpreted by Janssen (1997). For comparison the zonal distribution of emissions of EDGAR sources is also given in Figure 3.24. In the second phase of the project a more detailed analysis of the zonal budgets will be carried out. The uncertainty can be reduced by taking national inventories and process them in EDGAR to constrain each zonal band result from global modelling.

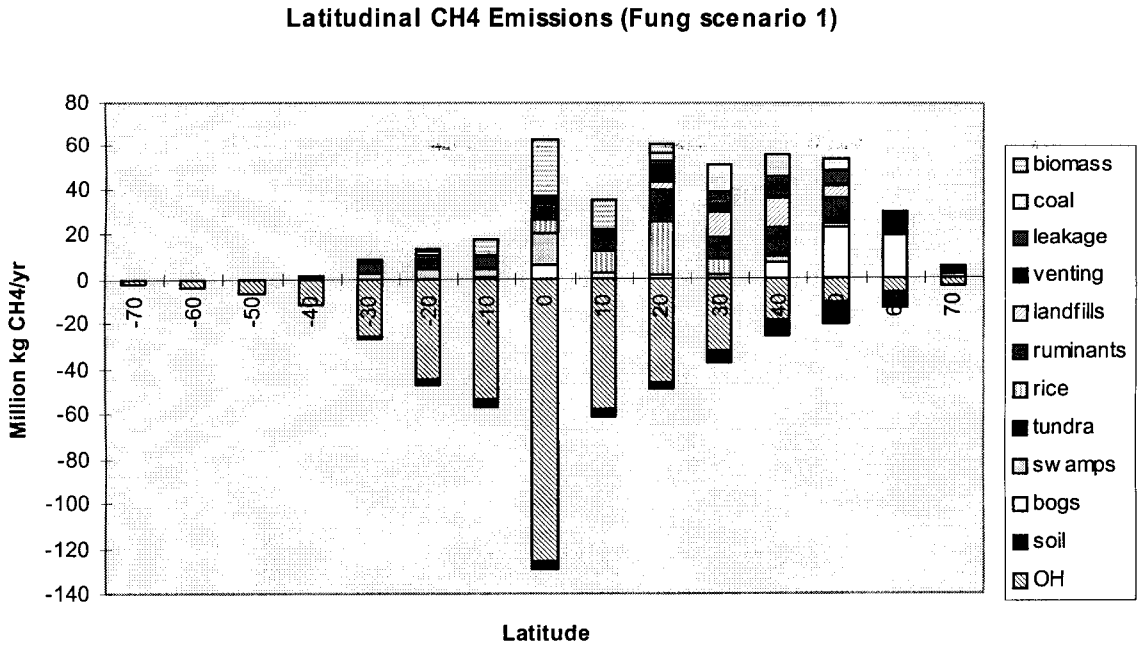


Figure 3.6 Fung *et al.* (1991) the zonal budget 1 (high tundra; global total 540 Tg CH_4)

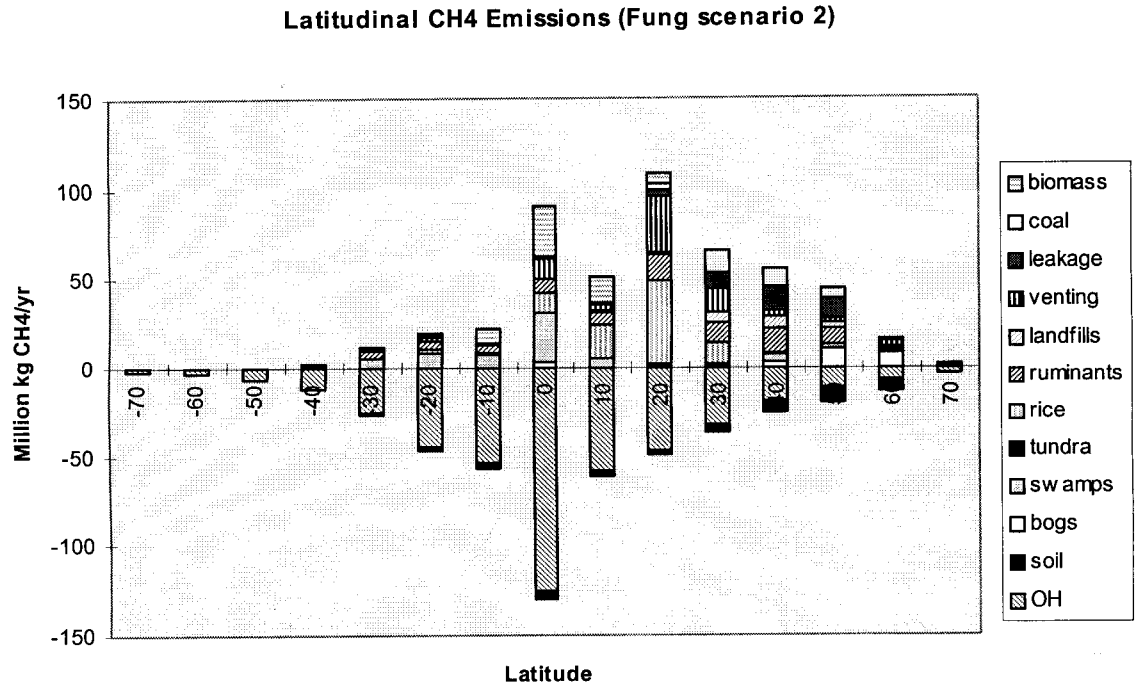


Figure 3.7 Fung *et al.* (1991) the zonal budget 2 (high fossil and high global total; 580 Tg CH_4)

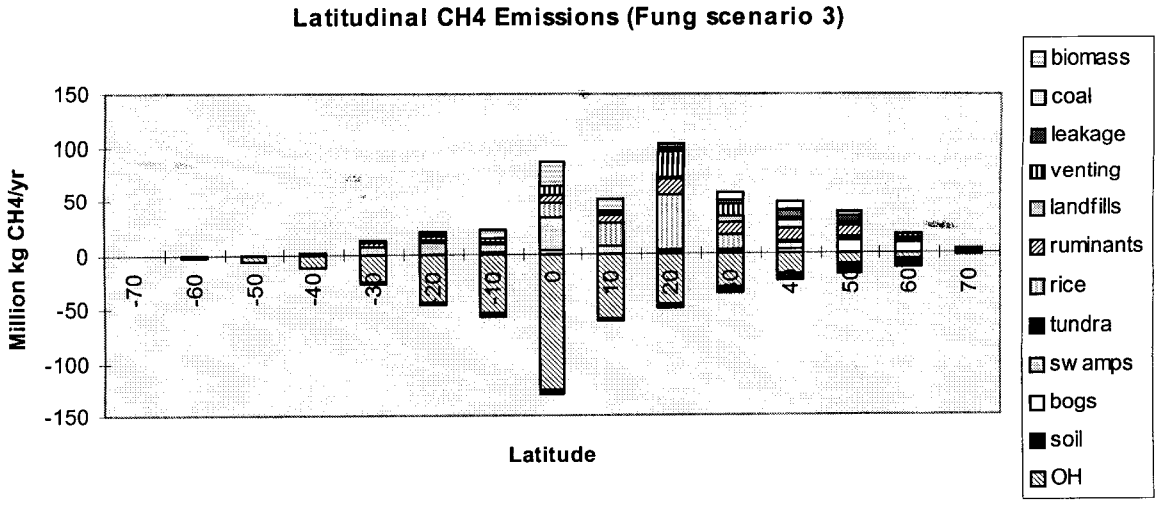


Figure 3.8 Fung *et al.* (1991) the zonal budget 3 (high bogs; global total 540 Tg CH_4)

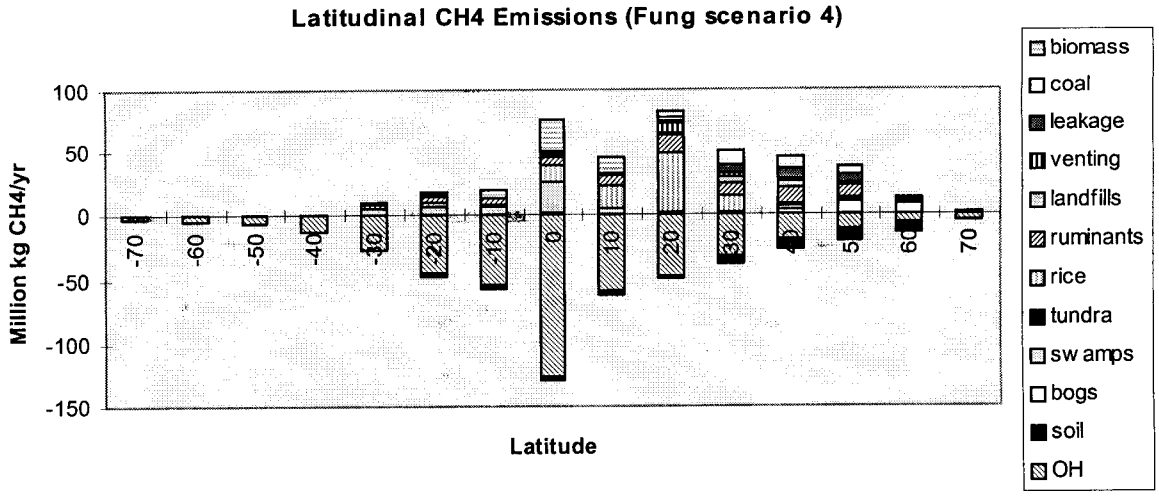


Figure 3.9 Fung *et al.* (1991) the zonal budget 4 (high ruminants; global total 540 Tg CH_4)

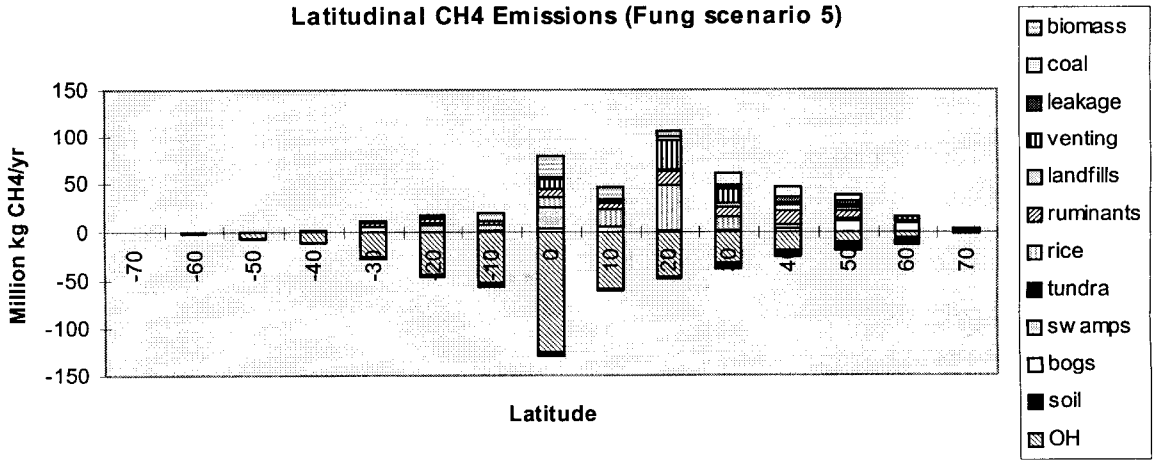


Figure 3.10 Fung *et al.* (1991) the zonal budget 5 (high fossil; global total 500 Tg CH_4)

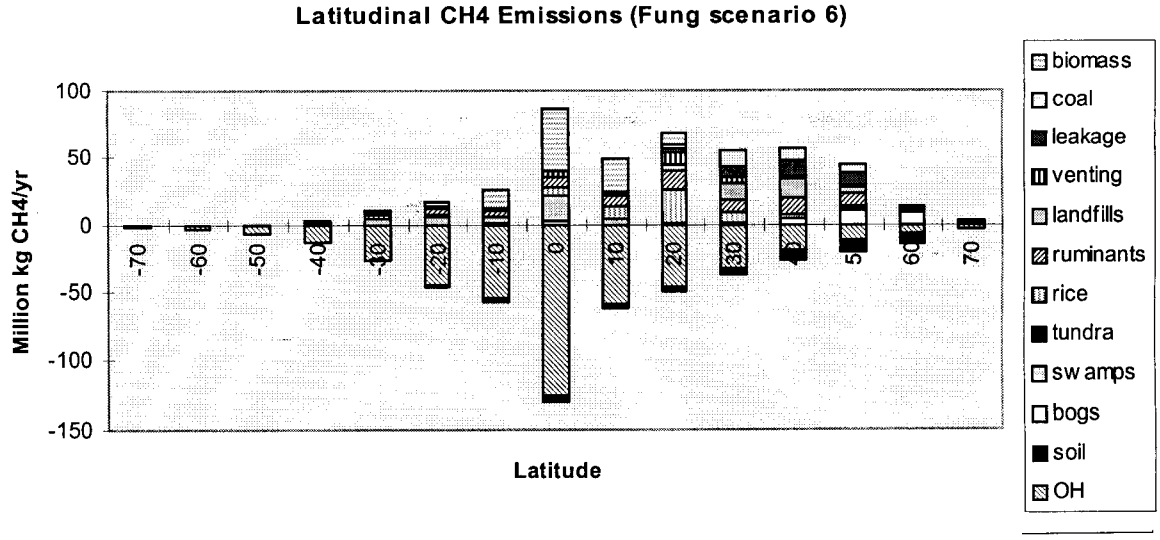


Figure 3.11 Fung *et al.* (1991) the zonal budget 6 (low rice; global total 500 Tg CH_4)

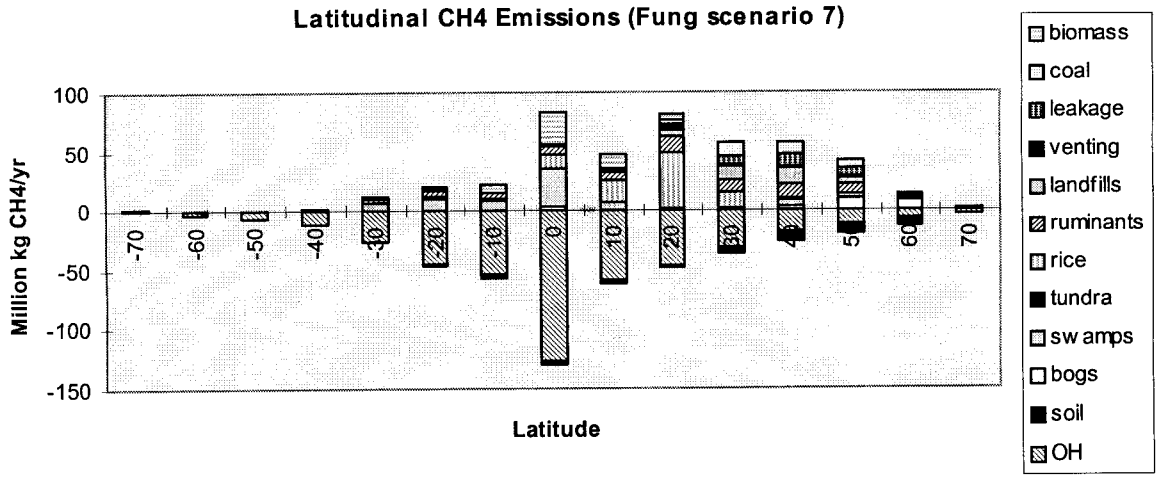


Figure 3.12 Fung *et al.* (1991) the zonal budget 7 (preferable scenario; low fossil, high wetlands, global total 500 Tg CH_4)

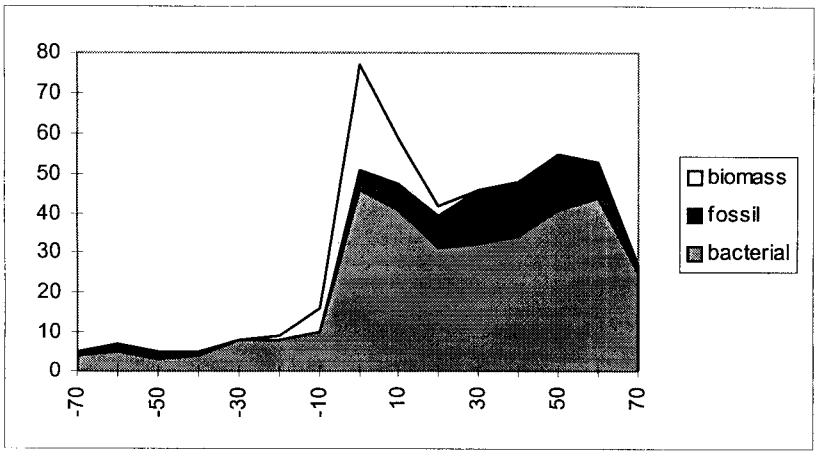


Figure 3.13 Saeki *et al.* (1997) the zonal budget.

3.3.9 The regional and local budget.

CH_4 differs from CO_2 and N_2O in that measuring data with high spatial and temporal resolution are available in source areas (Janssen *et al.*, 1997). Because we also have models available for Europe with high temporal (up to one hour) and spatial (5 x 5 km) resolution the emission of countries within a radius of 500 km from measuring stations can be deduced from measurements as shown by Vermeulen *et al.* (1997), Stijnen *et al.* (1997) and Van der Wal *et al.* (1997).

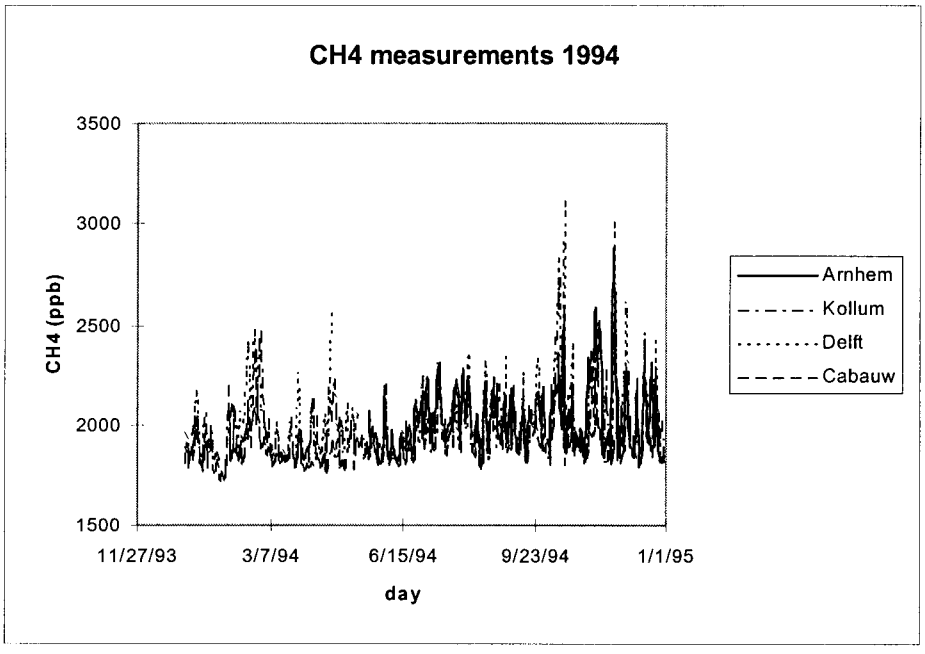


Figure 3.14 High resolution (hourly) measurements in a source area (The Netherlands).

3.3.10 Conclusions on budgets

The main conclusions are:
The short term variations in the atmospheric increase of the CH_4 concentration are in the order of magnitude of the yearly increase.
The long term atmospheric increase of CH_4 concentrations is representative of CH_4 emissions by human activities.
The uncertainty of total CH_4 emissions due to human activities derived from atmospheric measurements is in the order of 35% (derived from the uncertainty of emissions 40-50 Tg CH_4 and uncertainty of the OH sinks which is about the same)

3.3.11 Methodology of the Kalman filter for the evaluation of the methane budget for north-west Europe

Kalman filtering is used as a statistical technique for interpolation between measurement stations. Kalman filtering is applied in inverse modelling of nitrous oxide emissions by Hartley and Prinn (1993) and for methane by Zhang (1996) and Janssen *et al.* (1997). For north-west Europe the first promising results will be demonstrated in this preliminary study report. These results will also be used as a lesson for methodology development in this field.

Different simulation models on methane concentrations in north-west Europe are available in the Netherlands. The Eulerian dispersion model EUROS and the OPS model were developed at RIVM. The LOTOS model was developed by TNO. The Lagrangian transport model COMET was developed by ECN based on the KNMI-RIVM MPA model. These models

provide a reasonable picture of the transport dynamics and of the methane distribution for a specific region on a longer time frame.

However, since knowledge on physical transport processes is far from complete, and because deterministic models are an approximation and simplification of the real transport dynamics, model results will never be perfectly consistent with observations. Measurements seldom yield a complete picture of the process of methane emissions from the surface as well as chemistry and transport through the atmosphere, because sampling is limited in space and time. Therefore measurements and model results have to be integrated to obtain a more complete picture. The integration of measurement results with atmospheric model results is called data assimilation. With data assimilation maximum use is made of measurements and process knowledge. In figure 3.15 an example is given of data assimilation for one station Kollumerwaard for February 1994, using a Kalman filter. The filter enhances the deterministic model results.

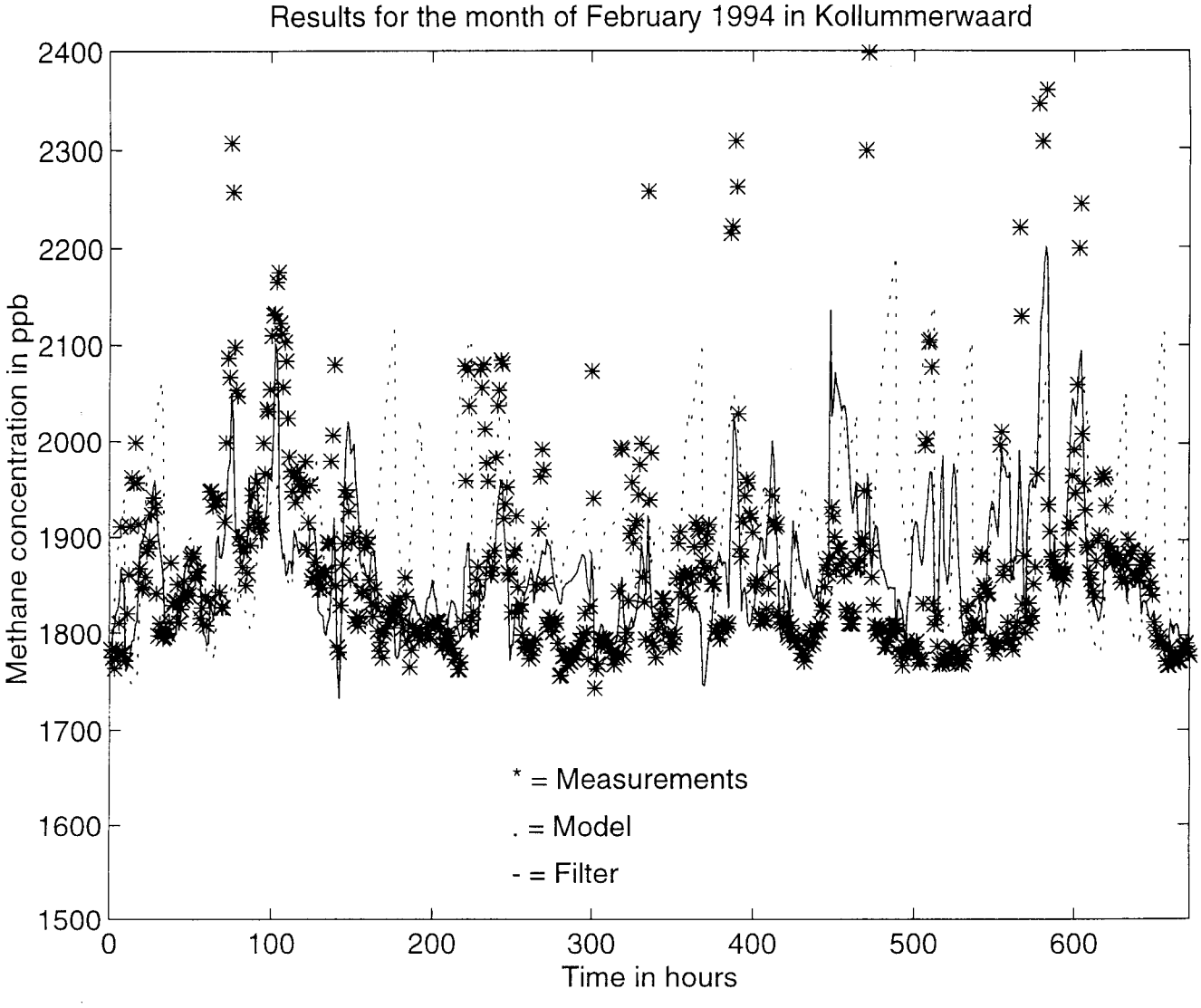


Figure 3.15 EUROS with Kalman filter results for February 1994 at Kollommerwaard, the Netherlands.

Among the different data assimilation methods the Kalman filtering approach has become more and more important and attractive, as the processing power of computers is rapidly increasing. The Kalman filter is an optimal sequential filtering technique based on a linear state-space time domain formulation. It is used to improve the modelled prediction by assimilation of measurement results. It was introduced by Kalman in 1960. In the Netherlands it has been used by Verlaan and Heemink (1995) for storm surge prediction and by Zhang (1996) for methane concentration prediction. It is an optimal state estimator that minimises the estimation error covariance under the assumption that complete specification of both dynamical and statistical model parameters are known. In 1994 an extended Kalman filtering (EKF) technique was used to handle non-linear models for weather forecasts (Bouttier, 1994). The advantage of the extended Kalman filtering compared to the other data assimilation methods such as optimal interpolation or the adjoint method is that the approximate covariance matrices of the error are provided at each time step. It was shown by Bouttier that even an extremely simplified version of the EKF could be useful for data assimilation in weather forecasting.

The EUROS model was used by Zhang (1996) to evaluate methane emissions in north-west Europe. Using a Kalman filter approach, a more complete picture using the measurement results of three stations in the Netherlands: Kollumerwaard, Delft and Cabauw, was obtained. A smoother approach was used to identify the emissions input uncertainty.

The EUROS model makes use of a Eulerian model grid. It contains 2860 cells in a grid of 55 by 52, each cell representing an area of 0.55 by 0.55 degrees. In the model the vertical dynamic transport of air pollutants (in this case methane) is assumed to pass through four layers: the surface layer, the mixing layer, the reservoir layer and the top layer. Advection, horizontal diffusion, vertical diffusion, fumigation and emissions are used to model the methane concentration in each layer and each cell of the grid. A wind field is used as the driving force for the diffusion.

In order to reduce the calculation burden related with Kalman filtering a reduced grid is used. The number of noise grid points is chosen to be 25 evenly spread in the model grid of 2860 grid points. The measuring stations are assumed to be on the nearest noise grid point. The model input of methane emissions per gridcell is calculated from measurement data using a smoother function for extrapolation. See figure 3.16 for forward calculations of methane concentration in the atmosphere on the 7th of February 1994. The EUROS model is run with average windfields over six hour periods. In order to reduce the calculation burden, average wind fields over longer time periods such as a day, or a week can be calculated and used. Average windfields over a month could be used for comparison with national emissions inventories. Emissions inventories are usually totals per year per source. These yearly totals could be distributed according to theoretical distribution functions for the seasonal variation.

3.3.12 Example of Inverse Modelling with a KALMAN filtering technique

Janssen *et al.* (1997) have validated the European methane emissions using the three dimensional EUROS model. With the KALMAN filtering technique measurement results can be used to enhance the results of the model. In the EUROS model concentrations of methane are calculated from emissions data. Using the inverse mode measured concentrations are used to calculate the expected emissions. Windfields are used to calculate concentrations at a specific day for Europe. Using this technique, a validation of national inventories is possible. In Table 3.6 the results are given. The Kalman results indicate that the "a priori" estimates from the LOTOS database were too low for Germany and the Netherlands, that they were comparable for France, and that the LOTOS inventory of the UK was too low compared with the model outcomes. See also figure 3.17. The south-west point of the grid is the south tip of Portugal

and the lowest grid point in the figure. It gives results for a month. So results are very preliminary. The large rise in this figure indicates that the "a priori" LOTOS emission estimate was far too low for that month. The national inventory of the UK was higher, so another run will be done with the national inventory as the "a priori" estimate in the model.

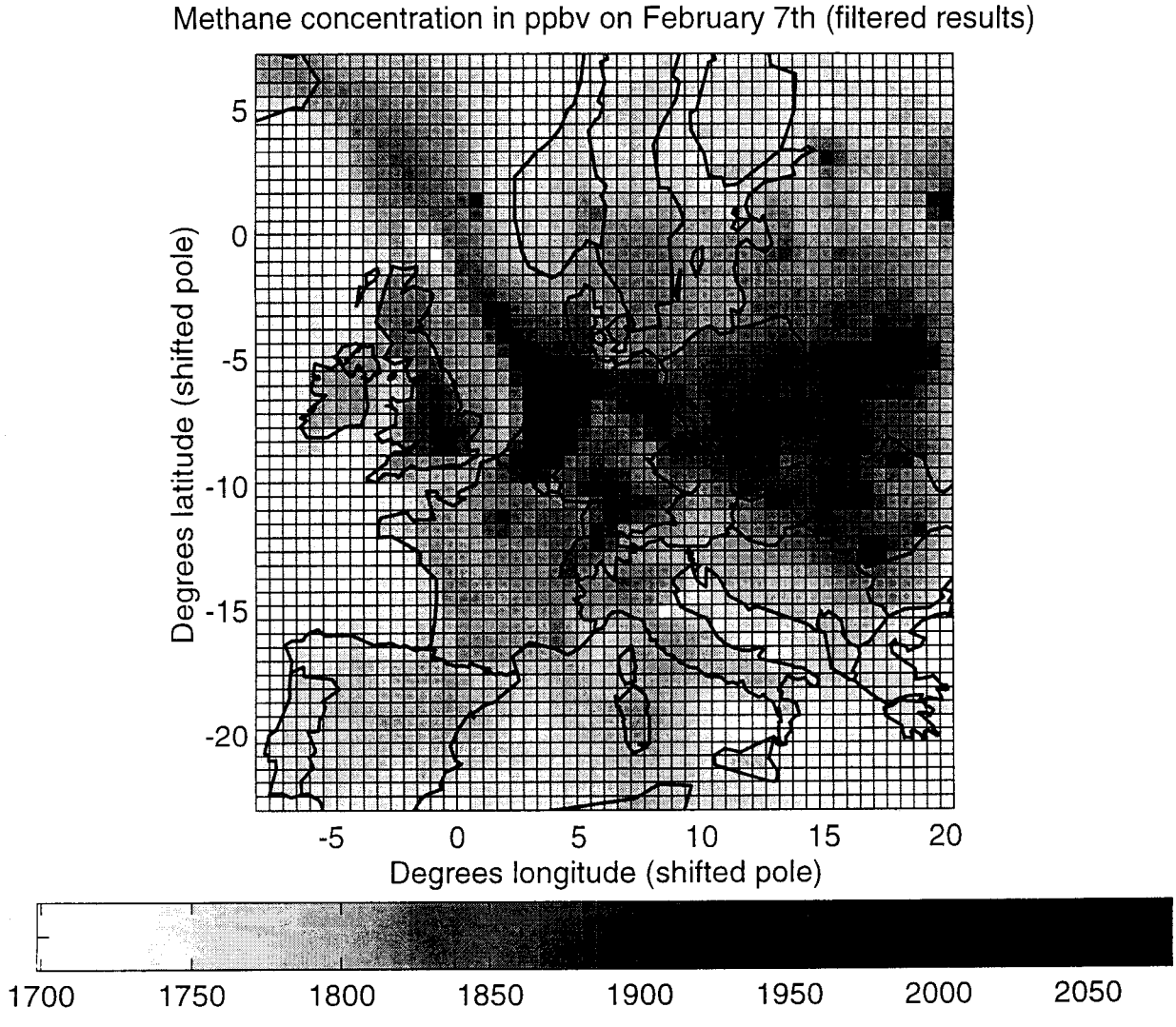


Figure 3.16 Forward calculation of methane concentration in the atmosphere on the 7th of February 1994, using the EUROS model for Europe and average wind fields.

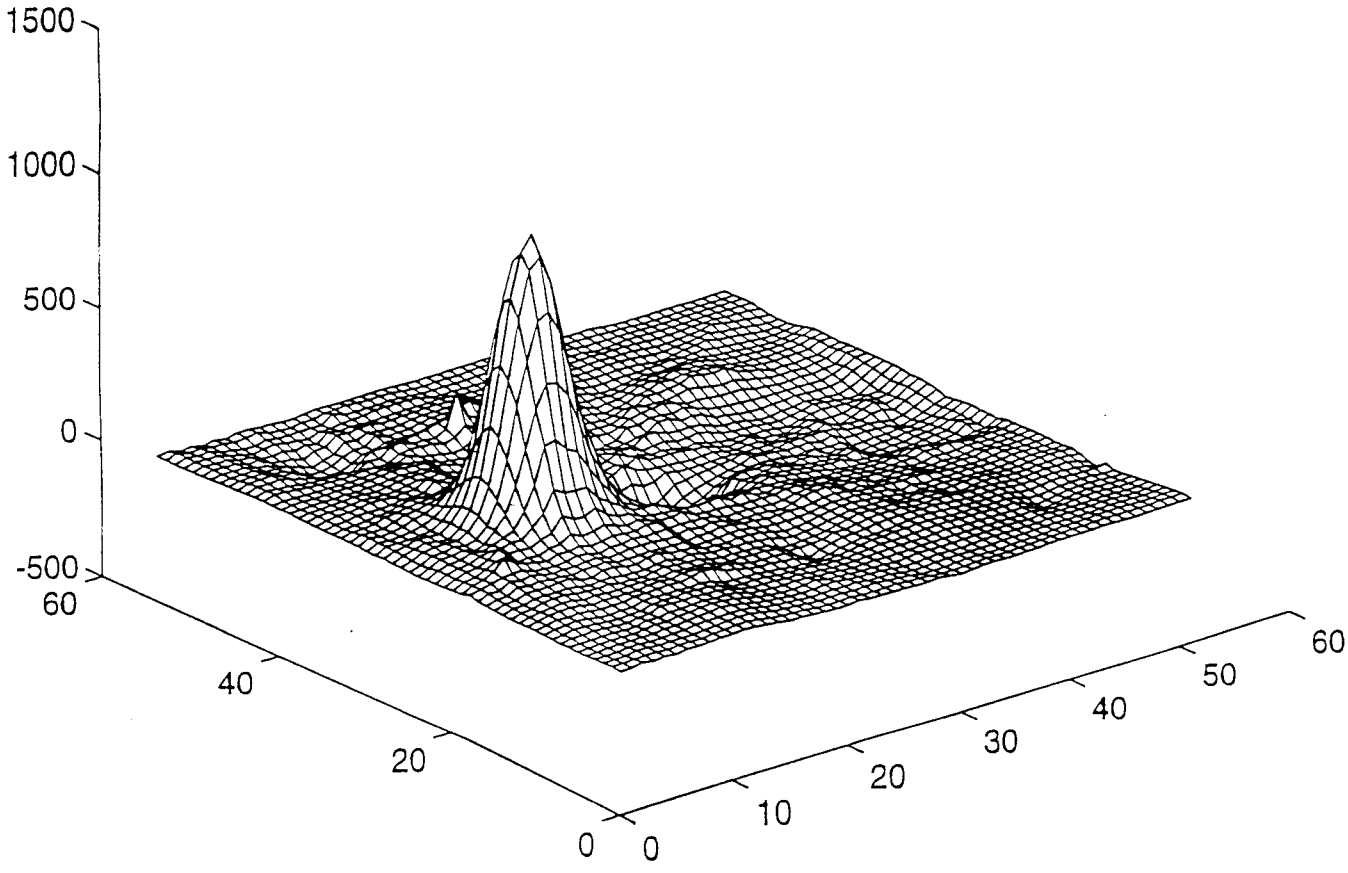


Figure 3.17 Difference (ppb) between an "a priori" calculation of CH_4 concentrations using the LOTOS emission database and the EUROS model and results of the EUROS model using real CH_4 measurements and the Kalmanfilter as a data-assimilation tool for February 1994. The south-west point of the grid is the south tip of Portugal. The high dome in the figure is over the UK.

Table 3.6 Preliminary comparison of National inventories with EDGAR, LOTOS and EUROS results.

Country	National inventory (mln. Kg.)	EDGAR (mln. Kg.)	LOTOS (mln. Kg.)	EUROS Kalman
Germany	5682	6153	3773	~-
France	2896	3765	3339	~0
United Kingdom	4531	3877	2306	++
Italy	3902	2396	1873	?
Netherlands	1060	1007	778	~-

Natural sources are not included in National inventories and EDGAR, but are included in LOTOS and EUROS models.
This comparison will be improved in the second phase of the project. ~- = LOTOS is a bit too low. ++ = LOTOS is too low. ~0 = LOTOS is comparable to model outcome. ? = no conclusion possible.

3.3.13 Trajectory analysis for methane.

Another method for validation of greenhouse gas emissions on a regional scale is trajectory analysis (Vermeulen *et al.*, 1996). In trajectory analysis Lagrangian, as opposed to Eulerian models are used. Eulerian models use a fixed grid. In Lagrangian models the origin of pollution is analysed by back tracking of the moving air masses passing over a measuring station. A validation of methane emissions for north-west Europe has been executed by the Netherlands Energy Research Foundation (ECN) through the use of this trajectory method

(Vermeulen *et al.*, 1996). In this method measured concentrations of methane at one location (Cabauw, the Netherlands) are related to the emissions upwind. Using wind-speed and direction, the origin of the air is calculated. If the height of the mixing layer is known from meteorological data, an emission at a remote location can be related to the concentration in the air measured at the fixed station. No distinction can be made between a large source at a remote location or a smaller source nearby unless more information is collected from nearby monitoring stations. One possibility is to use the direction of the wind and information on known sources in each direction. Isotope measurements have also been used at Cabauw to distinguish between methane from fossil fuel burning or venting and biogenic methane from waste, wetlands and animals. Of course more information is needed to distinguish the different sources of emissions at the level of detail which is used in national inventories. In the following a validation of the national inventory of the Netherlands is given using this trajectory methodology.

Cabauw
At the Cabauw measuring station in the Netherlands measurements of the atmospheric methane concentrations have been carried out since 1993. Also the ^{13}C and ^{14}C isotopic composition of methane has been measured in the same period by Vermeulen *et al.* (1996). They have conducted a trajectory analysis of the data. The Cabauw station measured methane concentrations at 200 m. above sea level in the mixing layer. Influence of local sources in the data were therefore minimised. The Cabauw methane concentrations record during 1993-1994 was used to calculate an emissions map of north-west Europe. The COMET transport model, specifically developed for this exercise, was used. The calculated source distribution for about 30 European districts matches reasonably well with the expected spatial pattern calculated from emissions inventories. The estimates for emissions per region are equal to or higher than the values reported in the national emissions inventories. For the Netherlands the COMET inverse calculation yields an emission of 1500 Million kg (Gg) methane per year, while the emission inventory for 1990 is 1200 Million kg (Van Amstel ed., 1995). A different emission estimate based on measurements resulted in a methane emission of 1200 Million kg for the Netherlands (Hollander and Vosbeek, 1996). This is in exact agreement with the official inventory (Van Amstel ed., 1995).

What is trajectory analysis? A Lagrangian transport model COMET is used to analyse methane concentration measurements at one location in relation to the wind direction and a 36-hours backward wind trajectory. For methane the atmospheric chemistry is negligible over this short period of time because the atmospheric lifetime is over 10 years. In the forward mode, per time step, the model retrieves the emissions for the grid cells under the current circular source area and calculates the concentration changes and isotopic composition for the modelled components in a column of air with the current mixing layer height. In the inverse mode the transport model is used to determine the contribution of each area to the measured concentration and ^{14}C content of each sample. The model uses the mixing layer height and the trajectory path to calculate the emission related to a measured concentration. The problem consists then of M linear equations, with N unknowns. M is the number of measurements and N the number of parameters. The solution is taken to be the least squares fit through the data. The matrix T contains the coefficients which have been calculated with the transport model. T_{ij} gives the contribution of the j^{th} source to the CH_4 concentration and the ^{14}C content of the i^{th} sample. This gives a set of equations:

$$m_{ij} = \sum T_{ij} \cdot e_{ij}$$

with m_{ij} (i=1...M) the series of measurements, and e_j (j=1...N) the emission factors to be determined. This problem is then solved by inversion of the matrix T through singular value

decomposition. This decomposition of the matrix T makes it possible to identify ill defined components of the problem, and to remove those terms.

Results
To arrive at reliable emission estimates, the number of concentration measurements should be much larger than the number of areas for which emissions are calculated. In the inverse model the emissions are calculated for aggregate areas on the LOTOS grid. Close to Cabauw a finer resolution is obtained. To test the validity of the inverse model outcomes, a comparison was made with the outcomes in the forward mode. In Table 3.7 the comparison of results is given. In the mean time recalculations have been done with better meteorological data from the DNMI for 1993-1995 about the height of the boundary layer. These results are presented in the Dutch workshop of 29 June 1997 (Vermeulen *et al.* 1997). Another conclusion from the trajectory study came from $\delta^{13}C$ analysis of samples: The non-biogenic methane emission in the Netherlands is 15% of the total.

Table 3.7 Comparison of COMET 'a priori' methane inventory with COMET in the inverse mode.

Country	COMET inventory	COMET inverse mode	Uncertainty
	Gg CH_4/yr	Gg CH_4/yr	%
Germany	6900	7650	0.7
France	3150	4150	0.6
Netherlands	1150	1150	2.9
Belgium	600	500	4.0
Luxembourg	35	55	5.6
United Kingdom	3850	3550	2.1
Ireland	650	800	2.7
Denmark	375	450	5.5
North Sea	550	975	6.8
Czech+Slovak Rep.	1150	950	2.1
Bulgaria	500	400	3.6
Romania	275	925	2.3
Hungary	525	700	2.6
Poland	5025	6250	0.5

Conclusion
Trajectory analysis based on only one measuring station is capable of assessing the national inventories for a number of surrounding countries in Europe. Although the sectoral detail of the trajectory analysis results is less than the bottom-up national inventories, the country totals can at least be validated.

3.4. Evaluation of emission inventories

3.4.1 Introduction
In this paragraph national inventories as reported to the Climate Convention Secretariat will be compared to the emission estimates made in EDGAR. For this purpose first a general description will be given of the methodologies used to estimate emissions in the IPCC Guidelines (3.4.3) and EDGAR (3.4.4). Then the country emissions will be compared (3.4.5). For this kind of evaluation to be successful, the documentation of the inventories must be complete and transparent (Van Amstel, 1993). The data must be detailed enough to reconstruct the inventory. Original calculation sheets and background reports to the National Inventory should be available and data should be referenced. A first evaluation or in-depth review on draft inventories was carried out by IPCC/OECD/IEA in 1993 to evaluate draft IPCC meth-

odology. For the Second Conference of the Parties to the Climate Convention the Climate Secretariat in 1996 published a second compilation and synthesis of 1990-1994 data of OECD and Eastern European countries (UNFCCC/CP/1996/12/Add.2, 2 July 1996). An overview of emissions, emission factors and activity data used in the inventories can be the basis for a thorough evaluation. Here a comparison with EDGAR is made. Only emissions will be compared to search for big differences or omissions. In a second phase of the project a more detailed analysis will take place using also information on emission factors and activity data. A comparison with GEIA is in preparation.

3.4.2 Precision, Accuracy, Uncertainties

Three sources of uncertainty can be defined: 1. Uncertainties in the national and international statistics; 2. Uncertainties arising from experimental research in the emission per unit product or unit area, called emission factors; 3. Uncertainty in the accuracy of the extrapolation methods for the upscaling from the field level to the national level. The precision of both IPCC and EDGAR estimation methods for methane is dependent on the underlying field research and the methods for upscaling to derive emission factors per unit of product or area. This will not be analysed here. More can be found in the IPCC Guidelines Reference manual and in the EDGAR documentation (Olivier *et al.* 1996). The accuracy of the inventories can be evaluated using top-down model results. This was described in an earlier section. Uncertainties in the estimates for methane are often cited as 20-30%. This is a high uncertainty and is related to the highly variable nature of the methane formation processes in time and space. Methanogenesis in soils is especially variable.

3.4.3 IPCC methodology for methane

The IPCC methodology for estimating methane emissions is documented in the IPCC Guidelines (IPCC/OECD/IEA, 1995; 1997). The main sources for methane emissions are leaks in the oil and gas systems, venting from oil and gas production sites, methane formation in ruminants (cattle mainly), methane from rice paddies and methane from waste treatment (landfills and waste water treatment). The main sink for methane is destruction in the atmosphere by OH radicals. Natural methane is formed in wetlands and natural seepage from oil and gas fields. Methodology of IPCC is based on experimental research to find emission factors for all these sources. Improvements can be expected as deterministic models are developed that explain methane formation from variables like soil type, temperature, moisture and oxygen content.

3.4.4 EDGAR methodology for methane

Fossil fuel

Fossil fuel use comprises production, transformation (i.e. production of secondary fuels such as coke and oil products) and combustion of fossil fuels. Process (i.e. non-combustion) emissions from coke ovens and refineries are also included here. The construction of the EDGAR energy production and consumption data sets has been discussed in section 2.4. For coal production, the 1990 data has been split into underground and surface mining, using separate country specific assumptions for hard coal and brown coal. More details can be found in Olivier *et al.* (1996). EDGAR data for fossil fuel, biofuel, and industrial processes will be incorporated in the GEIA inventory for methane.

Emission factors for fossil fuel production, transmission and handling

1) Coal production.
   The emission factors used in EDGAR to estimate CH_4 emissions due to brown coal and hard coal mining have been taken from a literature study by Smith and Sloss (1992). When

specified, emission factors for a given country, type of coal and mining depth were applied; in all other cases a global default values given in this reference were used. The emission factors used in Version 2.0 are listed in Table 3.8.

2) Crude oil production.
For crude oil production, the emission factors for CH_4 have been calculated from emission estimates by Arthur D. Little (1989). The emissions have a geographical distribution according to the Oil and Gas Journal energy database (OGJ, 1991).

3) Crude oil loading into marine tankers.
A minor part of the total crude oil production emission comprises emissions from oil loading into marine tankers. The magnitude of this part has been estimated with emission factors for oil loading, taken from OLF (1993). The crude oil loading emissions have been distributed according to an inventory of oil loading facilities in OGJ (1989), International Petroleum Encyclopedia. For this emission source category the general crude oil production profile has been assumed.

4) Natural gas production and natural gas transport.
The CH_4 emission factors for natural gas production and transport have been taken from Ebert *et al.* (1993).

5) Crude oil refining and storage at refinery site.
The emission factors for petroleum refining for CH_4, have been proposed by Veldt and Berdowski for the LOTOS database at TNO-MEP, Delft (TNO-MW, 1990). These emission factors account for all combustion and fugitive emissions occurring at the refinery site.

Table 3.8 Emission factors for methane emissions from underground and surface coal production in 1990 as used in EDGAR (g CH_4/GJ coal).

Hard coal Underground		Hard coal Surface		Brown coal Underground		Brown coal Surface	
World	835	World	77	World	24	World	24
USA	815	USA	75	Afghanistan	39	Afghanistan	39
Czechoslov.	785	Australia	60	Romania	39	Brazil	39
Germany	675	UK	15	Vietnam	39	Colombia	39
Hungary	585			Australia	0.26	Czechoslo.	39
Poland	585			Thailand	0.26	Norway	39
Romania	585					Philippines	39
Former USSR	585					Portugal	39
UK	455					Romania	39
Australia	335					Vietnam	39
South Africa	270					Australia	0.26
China	325					Germany	0.26
Rest of World	325					Algeria	0.26
						Spain	0.26
						UK	0.26
						Ireland	0.26

For fossil fuel combustion a subdivision was made for industrial use, use for utilities and residential use; each category and detailed fuel type has its own emission factor. EDGAR emission factors are taken from the LOTOS database, which are assumed to be globally applicable. The reference for the emission profile for coke production is the PHOXA report by Veldt (1994). For road transport and the evaporation of gasoline from automobiles the emission factors for CH_4 have been taken from Samaras (1991). The emission factor for CH_4 in air transport has been taken from Olivier (1995), in which the factor for methane had been derived from the total VOC factor for the LTO cycle and a percentage of 10% in total VOC

emissions in the LTO cycle. Above 1 kilometre methane emissions are assumed to be negligible. These emission factors are the global aggregate of all air traffic, including military aircraft and general aviation; aggregated emission factors which apply for scheduled civil air traffic only may differ 10 to 50%. All other means of transport have been regarded as small industrial combustion units for which the appropriate emission factors have been taken from the TNO-MW LOTOS database (TNO-MW, 1990). Olivier *et al.* (1996) show the calculated globally and regionally aggregated emission factors for fossil fuel combustion, both per main sector and fuel type.

Biofuel combustion
The sources of activity levels for total biofuel use have been described in Section 2.4. The methane emission factors for biofuels are from Veldt and Berdowski (1995). For the USA, the emission estimates from the US-EPA inventory for biofuel combustion have been converted into the EDGAR processes and entered directly into the EDGAR database. For all other countries the approach is described in section 2.4.5.

Industrial processes
Activity data were described in Section 2.4. For all countries the default emission factors for CH_4 from the LOTOS database have been used in EDGAR (TNO-MW, 1990) (see Table 3.9)

Table 3.9: EDGAR emission factors for CH_4 from industrial processes in 1990

Sector/product factor	Emission factor	Unit	Region	Reference
Ethene (ethylene)	247.5	g CH_4-C/ton	World	TNO, 1990 (LOTOS)
Styrene	22.5	g CH_4-C/ton	World	TNO, 1990 (LOTOS)
Coke production	13.171	kg CH_4-C/TJ	World	TNO, 1990 (LOTOS)
Sinter production	375.0	g CH_4-C/ton	World	TNO, 1990 (LOTOS)
Pig iron production (blast furnace)	675.0	g CH_4-C/ton	World	TNO, 1990 (LOTOS)

Land use and waste treatment
Land use and waste treatment sources in EDGAR include rice paddies (CH_4), fertiliser use (N_2O), animals (CH_4 and N_2O), biomass burning (all gases), agricultural waste burning and landfills (CH_4). Waste water and sewage treatment, which are considered to be sources of methane, are not included because to date no representative spatial emission estimates exist.

Rice production levels and the area of arable land per country were taken from FAO (1991), combined with country-specific corrections for all arable land grid cells. For emissions from animals we used animal populations per country from FAO (1991), except for caribous which were defined as numbers per grid cell (Lerner *et al.*, 1988). For agricultural waste burning we used estimates of carbon released per grid cell based on regional estimates of Andreae (1991) combined with the distribution of arable land according to Olson *et al.* (1983). For more details can be found in Bouwman *et al.* (1995). The landfilled amounts of waste are based on per country estimates of waste production per capita and the fraction which is disposed of by landfilling as specified for the 13 regions in RIVM's climate model IMAGE 2 with data from SEI (1992) as described in Kreileman and Bouwman (1994).

For rice cultivation and landfills the emission factor for CH_4 was taken from Kreileman and Bouwman (1994), with factors for landfills derived from Subak *et al.* (1992). Factors for methane from enteric fermentation by ruminants were taken from Gibbs and Leng (1993). For

biomass burning and agricultural waste burning, the CH_4 emission factors have been taken from Veldt and Berdowski (1995).

3.4.5 Global comparison

Total anthropogenic methane emissions are 320 Tg according to EDGAR (see table 3.12). The total anthropogenic methane emissions according to the National Inventories and US country studies are 170 Tg (see table 3.11). Clearly, not all countries have reported yet, which explains the big difference. The global total according to the IPCC assessment (1994) is 335 Tg. The EDGAR estimate is somewhat lower. In table 3.13 the differences between EDGAR and National Inventories is given for 12 world regions. The differences can be explained mainly because countries have not yet reported to the Climate Secretariat. The European Union (15 countries) figure for EDGAR and national inventories is comparable. Clearly reporting in the EU15 is complete.

3.4.6 Country comparison.

For methane, differences of more than 10% between EDGAR and National data will be looked into in more detail. In figure 3.18 and table 3.10 an overview of differences is given. The differences between EDGAR and National Communications in total national methane emissions were more than 10% in many countries, see table 3.10.

EDGAR uses TIER 1 methods to estimate methane from existing sources. National methods are generally IPCC TIER 2 or more detailed. This is the main reason for differences between EDGAR and country estimates. Differences in methane inventories are sometimes large and can vary by country.

In the following tables 3.11, 3.12, 3.13 the methane emissions per source are given according to the official National Communications (as of April 1997), according to the US country studies programme and according to EDGAR. In table 3.13 a comparison is made between official data and EDGAR.

Table 3.10 Total methane emissions in 1990 in OECD and Eastern European countries. Comparison of EDGAR data and National Communications.

Million kg CH4 per year (1990)	Total Official excl. Biomass[1)] excl. Bunkers	Total EDGAR excl. Biomass[1)] excl. Bunkers	Difference absolute	Difference %
	Gg methane	Gg methane	Gg methane	
Australia	6243	4476	-1770	-28%
Austria	603	452	-153	-25%
Belgium	?	599		
Bulgaria	1370	515	-856	-63%
Canada	3088	3851	+763	+25%
Czech Republic	942	1654		
Slovak Republic	347			
Czechoslovakia	1289	1654	+365	+28%
Denmark	407	423	+16	+4%
Estonia	323			
Finland	252	264	+12	+5%
France	2896	3757	+861	+30%
Federal Republic Germany		4809		
Former DDR		1327		
Germany	5682	6136	+454	+8%
Greece	343	426	+83	+24%
Hungary	545	708	+163	+30%
Iceland	23	19	-4	-19%
Ireland	796	657	-139	-17%
Italy	3901	2388	-1513	-39%
Japan	1382	3336	+1954	+141%
Latvia	159			
Liechtenstein	1	1	0	0%
Luxembourg	24			
Monaco		0,5		
Netherlands	1060	1002	-58	-5%
New Zealand	1986	1032	-954	-48%
Norway	290	265	-25	-8%
Poland	6100	4619	-1481	-24%
Portugal	226	421	+195	+86%
Romania	1954	2202	+248	+13%
Russian Federation	27000	47092[2)]	+20092	+74%
Spain	2151	2027	-124	-6%
Sweden	329	364	+35	+11%
Switzerland	332	313	-19	-6%
United Kingdom	4531	3868	-663	-15%
United States of America	27000	41512	+14512	+54%

[1)] Because of large uncertainty in activity level of biofuel use, this source is excluded here.
[2)] Total for former USSR.

Table 3.11 Methane emissions in 1990 for regions in the World based on National Communications and US country studies.

CH4 Gg/yr

Methane emissions according to National Communications and US country studies

	Fossil Fuels	Biomass Burning	Fugitive Oil,Gas	Fugitive Coal	Industry	Solvents	Enteric	Manure	Rice	Soils	Agr.Waste. Burning	Savanna Burning	Land.Use Change	Waste	Total (incl. Biomass for energy, excl. Int. Bunkers)	
IPCC code	1A1-5	1A6	1B1	1B2	2	3	4A	4B	4C	4D	4E	4F		5 6		
EU15	734	86	2854	1499	23	0	6732	2626	102	299	0	146	14	8052	23169	Total EU15
Rest OECD Europe	22	3	5	23	1	0	224	93	0	25	0	0	0	247	643	Total OECD-Eur.
E-Eur+Russia	102	23	6495	21686	6	0	6278	350	186	0	0	3	8	4572	39709	Total E-Eur+Russia
OECD-N.Am.	655	772	4491	4472	0	0	6460	2520	429	0	0	79	38	10945	30861	Total OECD-NA
OECD-Pacific	59	2	854	296	25	0	4768	271	271	0	370	22	379	2294	9611	Total OECD-Pacific
Latin-Am.	260	75	73	2820	0	0	3523	109	267	0	238	16	921	908	9210	Total Latin-Am.
Africa	594	1374	1440	217	0	0	3275	301	854	0	1822	349	221	2562	13009	Total Africa
India Plus	0	0	0	6	0	0	453	0	439	0	0	0	0	76	974	Total India Plus
China Plus	73	2686	10656	387	0	0	5850	2850	11800	0	0	0	0	14	34316	Total China Plus
E-Asia	12	240	7	1	0	0	732	109	6657	0	0	48	223	0	8029	Total E-Asia
Total of countries	2511	5261	26875	31407	56	0	38295	9228	21005	323	2430	663	1805	29670	169530	Total of countries
Total of world IPCC 1994		40000	55000	30000			80000	25000	60000					55000	375000	Total of world IPCC 1994
Ratio		0.13	0.49	1.13			0.48	0.37	0.35					0.54	0.45	Ratio

Note: Part of the countries have not reported, therefore the ratio: world total/total of countries, is small in most source sectors.

Table 3.12 Methane emissions in 1990 for regions in the World based on EDGAR.

METHANE EMISSIONS 1990 EDGAR															
All countries are considered			Fugitives	Fugitives			Agriculture								
Gg CH4 /year (full molecular)	Fossil fuels	Biofuels	Coal	Oil gas	Industry	Solvents	Enteric	Manure	Rice	Soils	Agr.waste	Savannah burning	Land use	Waste	Total (incl. biofuels, excl. Int.Bunk)
EDGAR REGIONS	1A1-5	1A6	1B2	1B1	2	3	4A	4B	4C	4D	4E	4F		5	6	
EU15	620	55	3586	1138	151	0	6996	3069	169	0	0	969	0	6101	22854 Total EU15
Other OECD Europe	60	129	100	95	32	0	939	174	21	0	0	383	0	724	2655 Total Rest OECD Europe
East Eur + former USSR	1283	36	8924	27402	218	0	11420	2540	311	0	0	1376	0	4573	58083 Total Eastern Europe +fUSSR
OECD North America	555	395	12217	10804	65	0	6353	2289	520	0	0	1230	0	11053	45482 Total OECD North America
OECD Pacific	137	25	1123	765	130	0	3159	773	997	0	0	172	0	1754	9034 Total Pacific
Latin America	126	1077	417	2279	38	0	17537	858	2398	0	1384	1022	2617	2683	32436 Total Latin America
Africa	81	3769	1324	1590	7	0	9014	393	1409	0	4293	1090	1728	1724	26422 Total Africa
Middle East	63	177	39	4309	0	0	1203	147	391	0	0	521	1	1388	8238 Total Middle East
India Plus	68	4021	865	479	17	0	13547	1100	24627	0	23	2821	403	1966	49938 Total India Plus
China Plus	1552	2776	9176	644	106	0	7125	2099	18922	0	48	1461	252	2571	46732 Total China Plus
East Asia	188	1600	194	2091	28	0	1288	564	10037	0	55	780	570	1144	18540 Total East Asia
Total Rest of World	6	10	0	7	0	0	6	1	3	0	0	0	0	13	46 Total Rest of World
Total World EDGAR	4739	14070	37965	51604	791	0	78586	14006	59805	0	5803	11826	5571	35694	320462 Total World EDGAR
Total World IPCC 1994		40000	55000	50000			80000	25000	60000					55000	375000 Total World IPCC 1994
Share EDGAR		0,.5	0.69	1.03			0.98	0.56	1.00					0.65	0..85 Share EDGAR

Note: Because in principle all countries are reported in EDGAR the share gives us information on the accuracy of the Total World estimates from IPCC compared to EDGAR.

Table 3.13 Methane emissions per region compared: National Communications and Emissions Database for Global Atmospheric Research (EDGAR).

Methane emissions Million kg per year (1990)																		
NC + USCS compared with EDGAR			Fossil Fuels	Biomass Burning	Fugitive Oil,Gas	Fugitive Coal	Industry	Solvents	Enteric	Manure	Rice	Soils	Agr.Waste. Burning	Savannah Burning	Land.Use Change	Waste		Total (incl. Biomass for energy, excl. Int.Bunk)
Source	Sort.Code	Group	1A1-5	1A6	1B1	1B2	2	3	4A	4B	4C	4D	4E	4F		5	6	
NatComm	1	EU15	734	86	2854	1499	23	0	6732	2626	102	299	0	146	14	8052	23169	Total EU15
Edgar	1	EU15	620	55	3586	1139	151	0	6996	3070	169	0	0	967	0	6101	22854	Total EU15
NatComm	2	OECD-Eur.	22	3	5	23	1	0	224	93	0	25	0	0	0	247	643	Total OECD-Eur.
Edgar	2	OECD-Eur	60	129	100	95	32	0	936	172	21	0	0	383	0	714	2641	Total OECD-Eur.
NatComm	4	E-Eur+fUSSR	102	23	6495	21686	6	0	6278	350	186	0	0	3	8	4572	39709	Total E-Eur+fUSSR
Edgar	4	E-Eur+fUSSR	1281	35	8924	27394	218	0	11357	2527	310	0	0	1373	0	4534	57952	Total E-Eur+fUSSR
NatComm	5	OECD-NA	655	772	4491	4472	0	0	6460	2520	429	0	0	79	38	10945	30861	Total OECD-NA
Edgar	5	OECD-NA	555	395	12217	10804	65	0	6353	2289	520	0	0	1230	0	11053	45482	Total OECD-NA
NatComm	6	OECD-Pacific	59	2	854	296	25	0	4768	271	271	0	370	22	379	2294	9611	Total OECD-Pacific
Edgar	6	OECD-Pacific	136	15	1123	765	130	0	3132	759	991	0	0	172	0	1653	8875	Total OECD-Pacific
NatComm	7	Latin-Am.	260	75	73	2820	0	0	3523	109	267	0	238	16	921	908	9210	Total Latin-Am.
Edgar	7	Latin-Am.	59	354	151	1518	4	0	3239	215	225	0	135	236	794	840	7770	Total Latin-Am.
NatComm	8	Africa	594	1374	1440	217	0	0	3275	301	854	0	1822	349	221	2562	13009	Total Africa
Edgar	8	Africa	64	2062	1316	853	7	0	4239	178	567	0	1267	645	671	924	12794	Total Africa
NatComm	10	India Plus	0	0	0	6	0	0	453	0	439	0	0	0	0	76	974	Total India Plus
Edgar	10	India Plus	1	312	0	50	0	0	806	76	4131	0		121	6	194	5696	Total India Plus
NatComm	11	China Plus	73	2686	10656	387	0	0	5850	2850	11800	0	0	0	0	14	34316	Total China Plus
Edgar	11	China Plus	1531	2398	9057	635	100	0	6529	1986	14869	0	11	1291	67	2351	40824	Total China Plus
NatComm	12	E-Asia	12	240	7	1	0	0	732	109	6657	0	0	48	223	0	8029	Total E-Asia
Edgar	12	E-Asia	16	321	11	71	2	0	609	195	5134	0	12	397	172	364	7306	Total E-Asia

Note: Only the countries with information from National Communications or USCSP are aggregated in the regional totals and compared with the same countries in EDGAR. So these regional totals are not the final estimates because more countries in the future will report their emissions estimates.

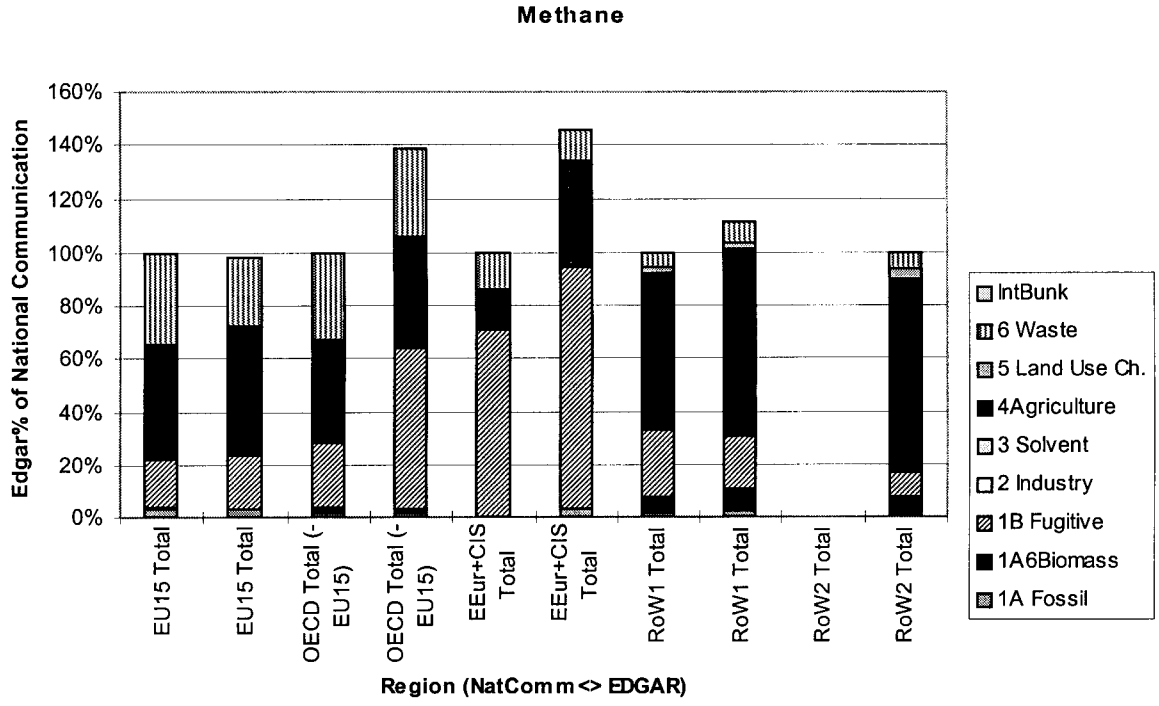


Figure 3.18 Comparison of National Inventories with EDGAR data for different regions in the World: European Union 15, Rest of OECD countries, Eastern Europe and former Soviet Union, Rest of the World 1 for which US country studies were available and Rest of the World 2 where only EDGAR data were available.

(In the above graph the first bar is the sum of national inventories. The second bar is the sum of EDGAR data for the same countries. National Inventories total for a region is assumed 100% here. Emissions of methane from international marine and aviation bunker fuels are not included in national totals. Sectors according to legend are: CH_4 emissions from international marine and aviation bunker fuels; CH_4 emissions from landfills and waste water treatment; CH_4 from land use change; CH_4 emissions from agriculture such as cattle and manure; CH_4 emissions from solvent use; CH_4 emissions from industrial processes; CH_4 emissions from oil and gas production and distribution (gas vents and leaks); CH_4 emissions from fossil fuel combustion. RoW1 = Rest of World for which USCSP data are available. RoW2 = Rest of World for which only EDGAR data are available).

In figure 3.18 a comparison is made of the data from the previous tables. From this figure it is clear that the regional official total for the European Union 15 is about the same as the EDGAR data, but within sectors differences may be large, such as agriculture and waste. The difference in the rest of OECD Europe (smaller countries) is large but in absolute terms this is not so important. The difference in Eastern Europe is large, in the total as well as the sectors. For the rest of the World 1 relative differences are not large, but in absolute terms this may be important. In the following IPCC sector codes are used in headings.

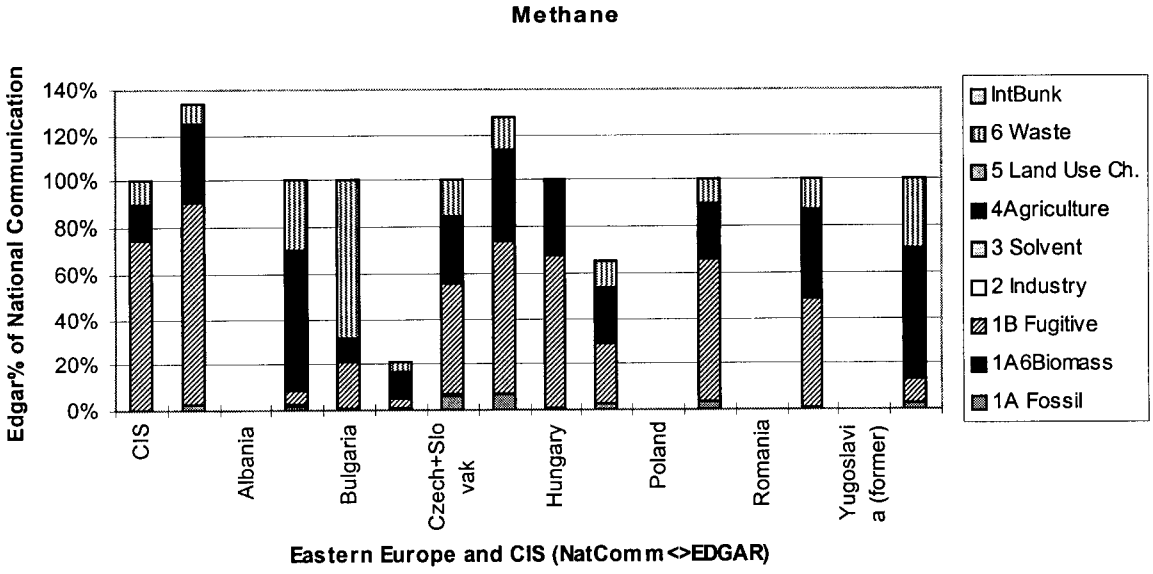


Figure 3.19 Comparison of National Inventories with EDGAR data for Eastern Europe and Russian Federation.

(In the above graph the first bar is the national inventory of the Russian Federation. The second bar is the EDGAR data for the same countries. National Inventories total for a country is assumed 100% here. Emissions of methane from international marine and aviation bunker fuels are not included in national totals. Sectors according to legend are: CH_4 emissions from international marine and aviation bunker fuels; CH_4 emissions from landfills and waste water treatment; CH_4 from land use change; CH_4 emissions from agriculture (such as cattle and manure); CH_4 emissions from solvent use; CH_4 emissions from industrial processes; CH_4 emissions from oil and gas production and distribution (gas venting and leaks); CH_4 emissions from fossil fuel combustion. For some countries only EDGAR data are available).

Methane from fossil fuel combustion (IPCC sector 1A1-5)
No differences are found over 10% between EDGAR and National data, with the exception of Algeria and Botswana. In Algeria methane from fossil fuel burning seems to be reported incorrectly. It is the same amount as fugitive emissions in EDGAR so maybe it should have been reported under fugitive emissions. Methane from biomass burning 1A6 shows some large differences in Bangladesh, Botswana and Nigeria, but in absolute terms the quantities are small.

Fugitive Methane emissions (IPCC sector 1B)
Differences between EDGAR and national estimates are over 10% in the following regions: Eastern Europe + former Soviet Union and in the rest of OECD (excluding EU15). The following countries show differences of more than 20%: Hungary, Spain, Japan, Norway, United States, Algeria, and Venezuela. The most important differences are in the Russian Federation where the EDGAR estimate is 13% higher, in the USA where the EDGAR estimate is 50% higher, and in Germany where the EDGAR estimate in 13% higher.

Possible reasons for the differences are:
1. EDGAR overestimates leaks from the distribution network by using gross gas consumption. In reality part of this gas is distributed directly to the industry and to power plants by high pressure transport mains that are less leaky than the low pressure distribution network.

2. EDGAR uses its own detailed methodology for methane from coal mines, based on basin specific methane emission factors.

Methane from agriculture (IPCC sector 4)
Methane from agriculture is emitted by different sub sectors. Methane from animals and from manure are the larger contributors. Methane from agricultural waste burning is more important in the tropics. A difference between national inventories and EDGAR data of more than 20% was found in the following countries in Europe: Finland, France, Greece, Portugal; in the rest of OECD: Japan, New Zealand; in the rest of the world: Bangladesh, Botswana, Cameroon, Ethiopia, Gambia, Ghana, Mongolia, Peru, Philippines, Senegal, Thailand, Uganda, Zimbabwe. EDGAR estimates are higher than national estimates with the exception of Ireland, Italy, Australia, New Zealand, Switzerland, Peru, Thailand and Uganda. The differences are primarily a consequence of the more detailed calculations used in national estimates. The animal statistics used by countries may differ somewhat from the FAO statistics used in EDGAR. Another possible explanation is that in EDGAR estimates were used for the amount of agricultural residues burned from Hall *et al.* (1994). These are different from country statistics.

Methane from land use change and forestry (IPCC sector 5)
Methane from land use and forestry is a minor source with 5.6 Tg according to EDGAR.

Methane from waste (IPCC sector 6)
Differences are high and primarily the result of more detailed estimates used in national inventories. When looking at totals per region, differences are below 10%, so it seems that EDGAR comes closer to regional totals. For the following countries differences of more than 20% were found between EDGAR and national data: Bulgaria, Austria, Finland, Portugal, United Kingdom, Iceland, Norway, Algeria, Botswana, Côte d'Ivoire, Gambia, Mongolia and Senegal.

Possible reasons for differences are:
1. In EDGAR an estimate is made using per capita waste generation and the methane potential from landfilled waste, whereas in countries more detailed statistics are used on the amounts produced and landfilled.
2. In some countries a more detailed time dependent method is used to calculate methane from landfills, the Scholl Canyon Model. This is the case in the Netherlands and the United Kingdom. Differences are about 20%.
3. In national estimates methane from liquid waste is included.

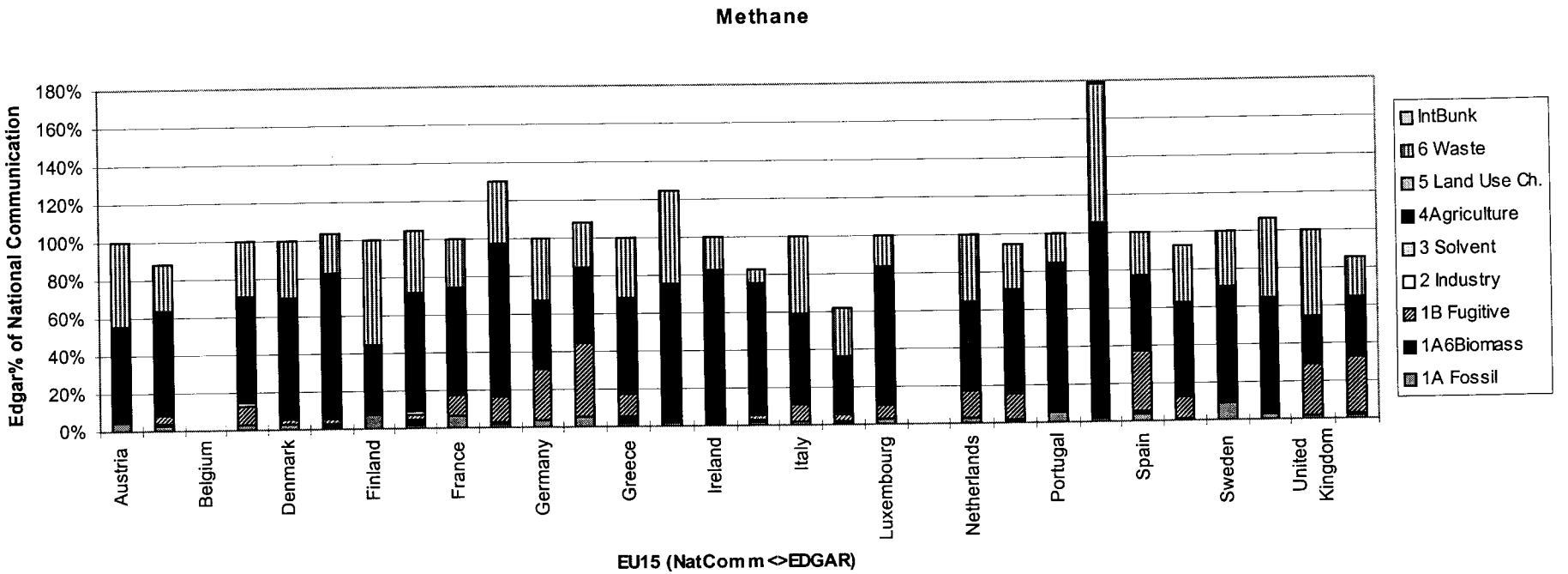


Figure 3.20 Comparison of methane emissions in 1990 of National inventories of European Union countries with EDGAR data.

(In the above graph the first bar is the national inventory of Austria. The second bar is the EDGAR data for the same country. National Inventory total for a country is assumed 100% here. Emissions of methane from international marine and aviation bunker fuels are not included in national totals. Sectors according to legend are: CH_4 emissions from international marine and aviation bunker fuels; CH_4 emissions from landfills and waste water treatment; CH_4 from land use change; CH_4 emissions from agriculture (such as cattle and manure); CH_4 emissions from solvent use; CH_4 emissions from industrial processes; CH_4 emissions from oil and gas production and distribution (gas venting and leaks); CH_4 emissions from fossil fuel combustion. For Belgium only EDGAR data are available, for Luxembourg only national inventory is available).

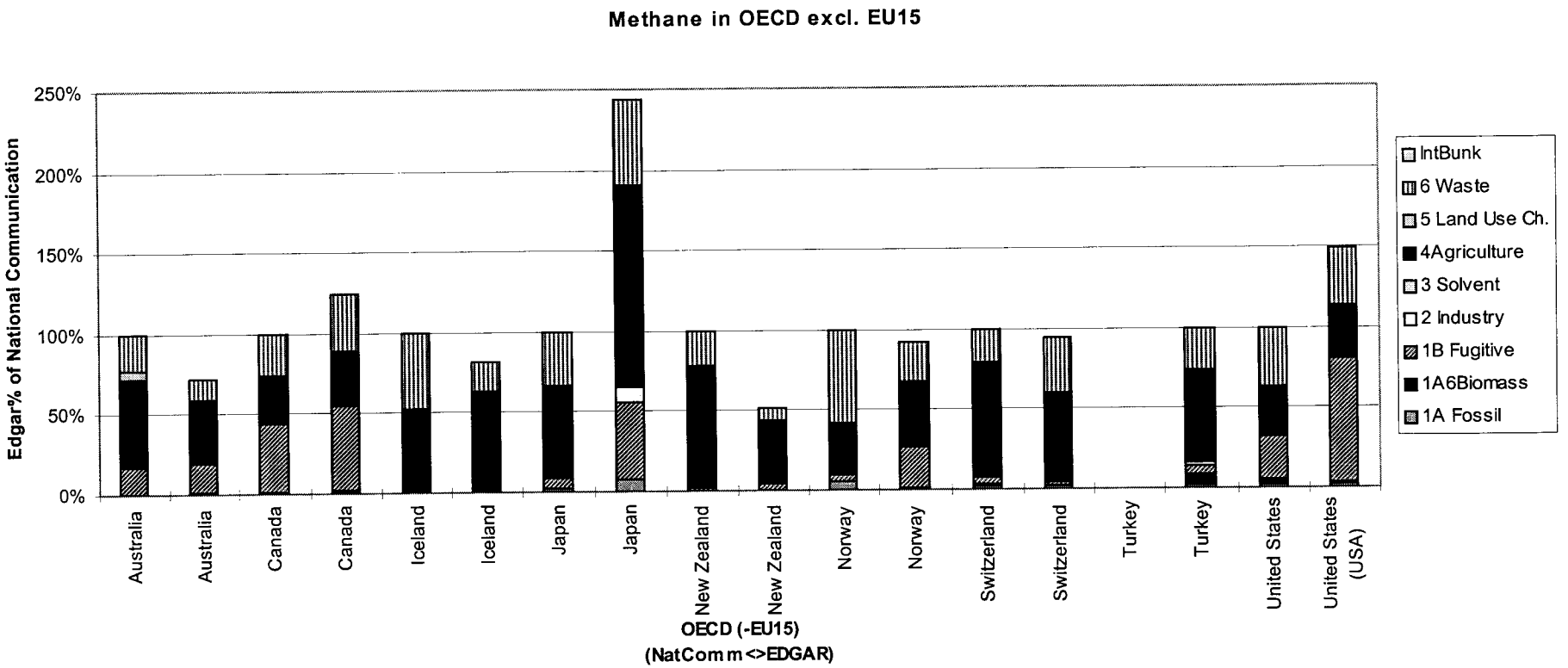


Figure 3.21 Comparison of methane emissions in 1990 of National inventories of other OECD countries with EDGAR data.

(In the above graph the first bar is the national inventory of Australia. The second bar is the EDGAR data for the same country. National Inventory total for a country is assumed 100% here. Emissions of methane from international marine and aviation bunker fuels are not included in national totals. Sectors according to legend are: CH_4 emissions from international marine and aviation bunker fuels; CH_4 emissions from landfills and waste water treatment; CH_4 from land use change; CH_4 emissions from agriculture (such as cattle and manure); CH_4 emissions from solvent use; CH_4 emissions from industrial processes; CH_4 emissions from oil and gas production and distribution (gas venting and leaks); CH_4 emissions from fossil fuel combustion. For Turkey only EDGAR data are available).

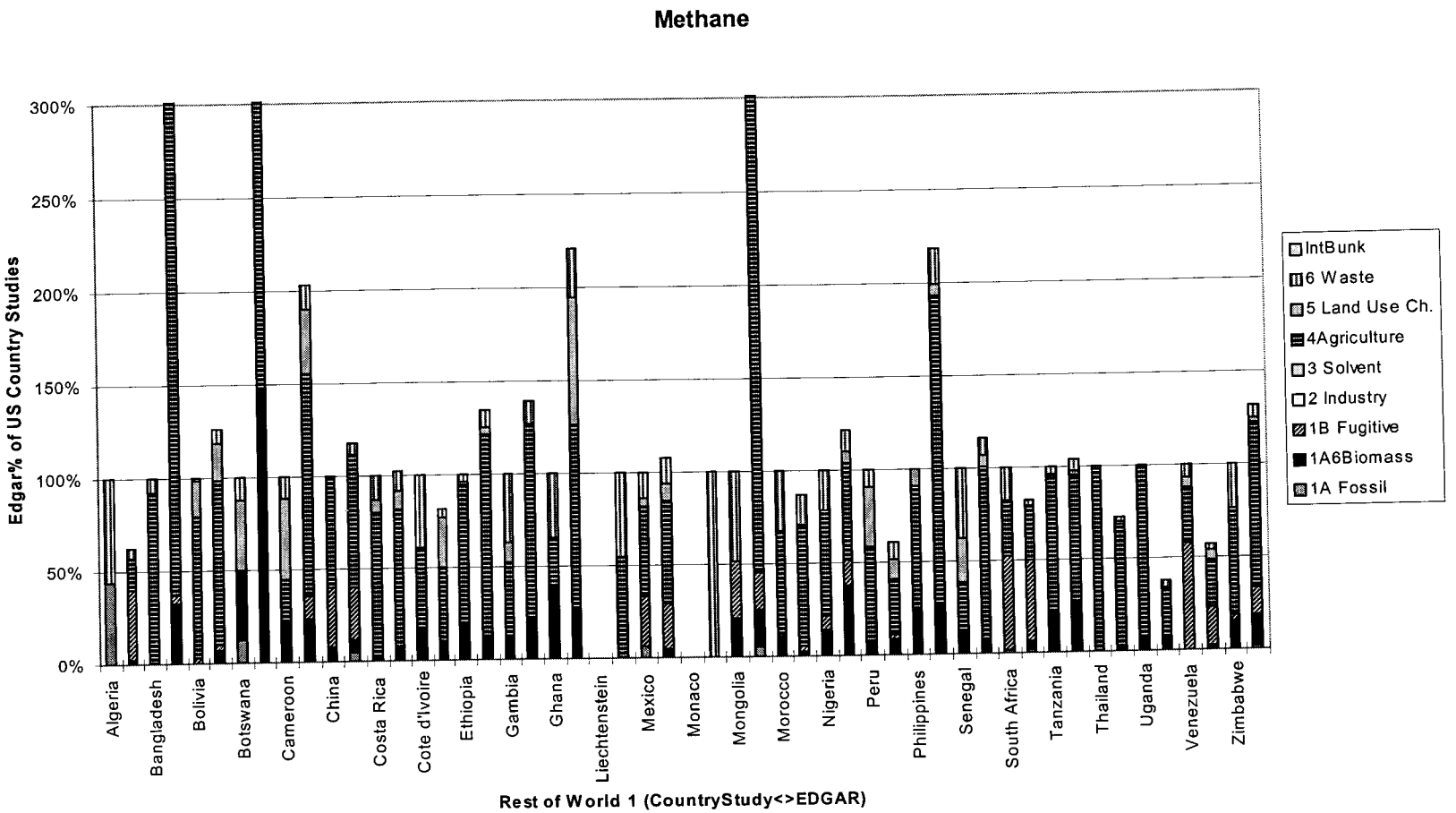


Figure 3.22 Comparison of methane emissions in 1990 of US country studies with EDGAR data.

(In the above graph the first bar is the country study of Algeria. The second bar is the EDGAR data for the same country. US country study total for a country is assumed 100% here. Emissions of methane from international marine and aviation bunker fuels are not included in national totals. Sectors according to legend are: CH_4 emissions from international marine and aviation bunker fuels; CH_4 emissions from landfills and waste water treatment; CH_4 from land use change; CH_4 emissions from agriculture (such as cattle and manure); CH_4 emissions from solvent use; CH_4 emissions from industrial processes; CH_4 emissions from oil and gas production and distribution (gas venting and leaks); CH_4 emissions from fossil fuel combustion. For some countries only EDGAR data are available).

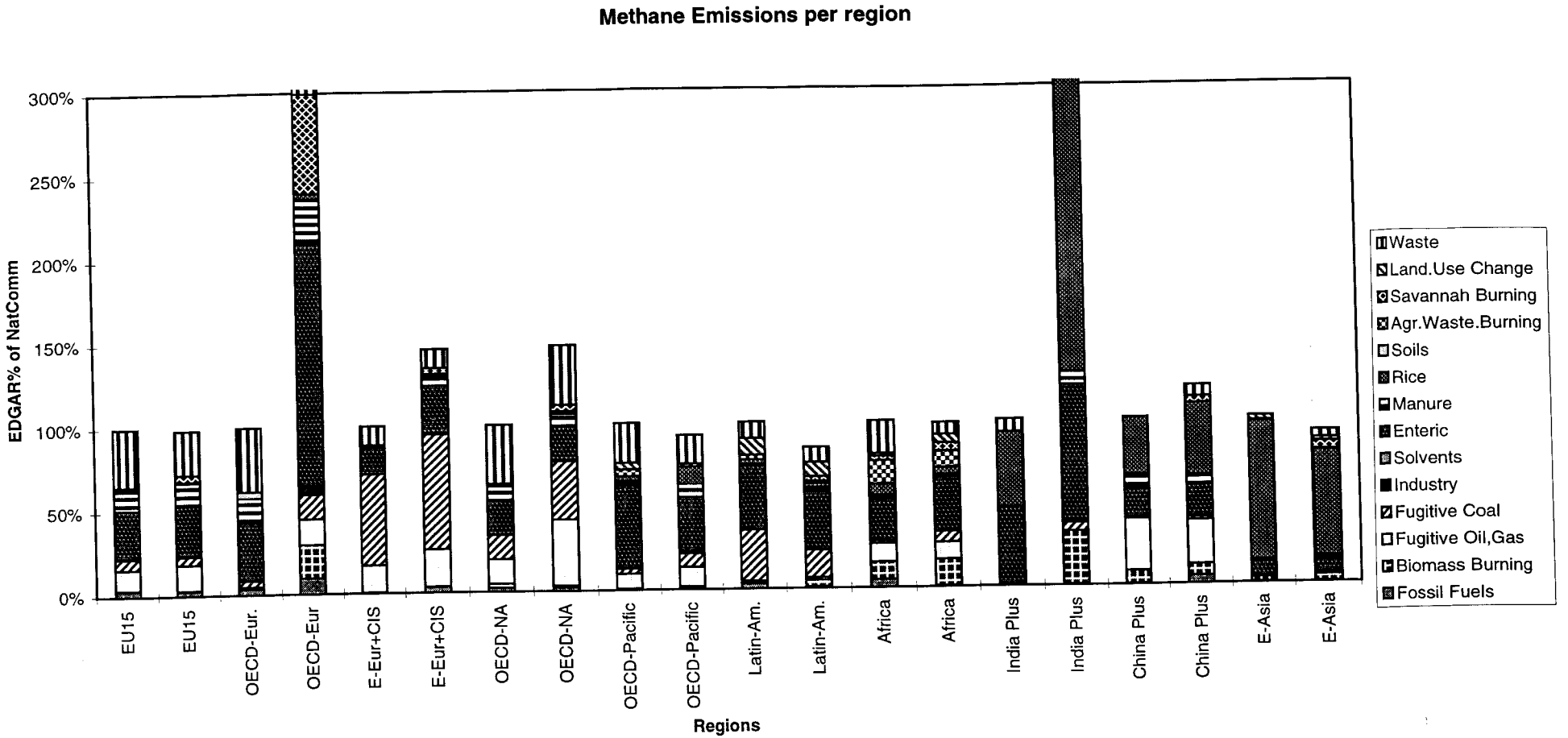


Figure 3.23 Comparison of CH_4 emissions from national inventories with EDGAR for IMAGE regions in the World.

(In the above graph the first bar is the sum of national inventories and US country studies. The second bar is the sum of EDGAR data for the same countries. National Inventories total for a region is assumed 100% here. Emissions of methane from international marine and aviation bunker fuels are not included in national totals. Sectors according to legend are: CH_4 emissions from waste; CH_4 from land use change; CH_4 emissions from savannah burning; CH_4 emissions from agricultural waste burning; CH_4 emissions from soils; CH_4 emissions from rice; CH_4 emissions from livestock; CH_4 emissions from manure; CH_4 emissions from enteric fermentation; CH_4 emissions from solvent use; CH_4 emissions from industrial processes; CH_4 emissions from coal production; CH_4 emissions from oil and gas production and distribution (gas vents and leaks); CH_4 emissions from biomass burning; CH_4 emissions from fossil fuel combustion. Countries are compared only if for both US Country Study and EDGAR data were available).

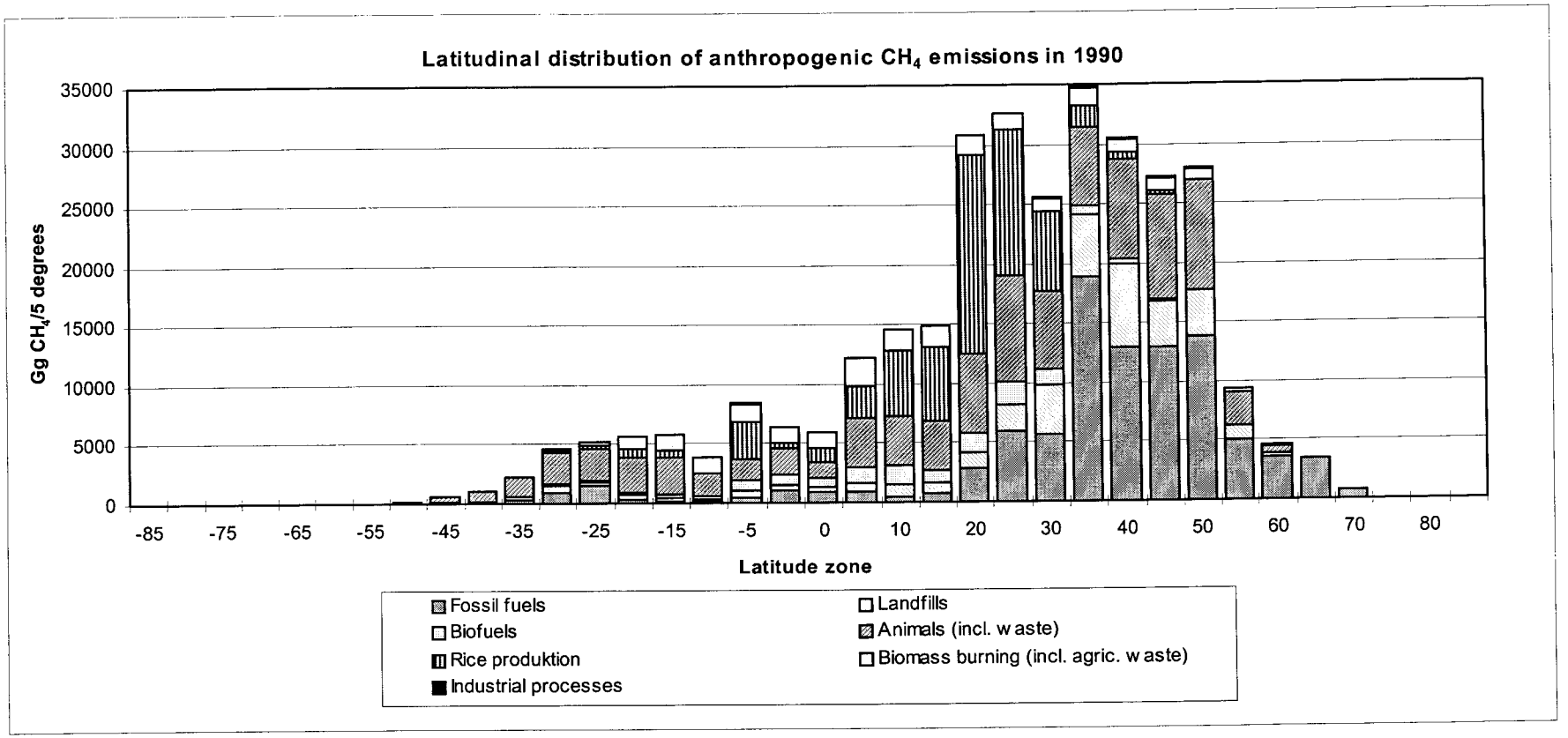


Figure 3.24 Latitudinal distribution of methane according to EDGAR. (over 5 degree bands, 90 = N)
In the second phase of the study this distribution will be compared with top-down model results for zonal bands.

3.5 Conclusions for methane

The research questions as formulated in chapter 1.2 will be answered for CH_4 in the following:

Available inventories
For methane many different model studies are available. Here we have used the global model synthesis by Fung *et al.* from 1991, Lelieveld and Crutzen 1993, Muller 1993, Hein *et al.* 1994 and 1997, The and Beck 1995, and Saeki 1997. For local validation studies for NW Europe we have used various studies in the Netherlands based on four measurement sites (Hollander and Vosbeek, 1996; Vermeulen *et al.* 1996; Janssen *et al.* 1997). Two databases of national inventories have been used: the Climate Secretariat database of national inventories and the EDGAR database. The summary of US country studies by Braatz *et al.* 1996 has been incorporated.

Presentation of inventory data
National inventory data are presented per sector per year according to the IPCC recommended summary table format. EDGAR data are presented per sector per year according to own sector definitions. These could however be translated easily to IPCC recommended format.

A method for comparison of emission inventories
Concerning the top-down bottom-up comparison of emission inventories for methane it can be concluded that global, regional and gridded comparisons are possible, but by doing this the uncertainty can not be reduced while both model results and national inventories have the same overall uncertainty of about 20-35%. Gridded comparisons using data assimilation with a Kalman filter, are only feasible in Europe and North America at this moment, while long term high frequency measurements are still lacking in the other regions.

Comparisons of bottom-up inventories like the comparison of national inventories with EDGAR are feasible and useful. The recommended format for the comparison is the summary tables 7A and 7B of the IPCC Guidelines. Comparisons can be done by country and by groups of countries. Here the following groups were used: EU15, other OECD Europe, Eastern Europe and Russia, OECD North America, OECD Pacific, Latin America, Africa, Middle East, India region, China region, East Asia, Rest of the World.

Uncertainties
The uncertainties are seldom quantified in the inventories. From some experimental studies we know that uncertainties are in the range of 20-35% for all sources. The uncertainties in the national inventories can not be reduced by validation with model studies, while these are from the same magnitude. Uncertainties can be reduced by deterministic studies and model development to get a grip on the processes of methane formation in soils. To reduce uncertainties more measurements of the type of the Cabauw site (at 200 m height) are needed in regions other than Europe and North America.

Preliminary comparison
The country comparison with EDGAR showed that differences are large in all important sectors even in Europe, although the totals match more closely. In a second phase the reasons for these differences have to be investigated.

Recommendations for methodology development
In this preliminary study we did not investigate the reasons for observed differences in detail. Clearly, a further in-depth analysis is needed. A database of emission factors is needed to carry out this in-depth study. IPCC methodology can be improved in the following sectors: agriculture, land use, biofuel combustion, landfills and waste water treatment.

Other conclusions
1. The short term variations in the atmospheric increase of the CH4 concentration are in the order of magnitude of the yearly increase.
2. The long term atmospheric increase of CH4 concentrations is representative of CH4 emissions from human activities.
3. The uncertainty of total CH4 emissions due to human activities derived from atmospheric measurements is in the order of 35% (derived from the uncertainty of emissions estimates of 40-50 Tg CH4 and uncertainty of the OH sinks which is about the same).
4. The comparison of Top-down with Bottom-up emission inventories is possible on a local scale for NW-Europe because long term measurement results are available.
5. Trajectory analysis with one measuring station like Cabauw is capable of assessing the national inventories for surrounding countries up to a distance of 500 km. The sectoral detail is less than the inventories but the totals can at least be validated.
6. The comparison of inventories with models for zonal bands is possible but the uncertainty in the models is the same or higher as national emission inventories, around 30%.
7. The EDGAR database could be used for a zonal comparison. EDGAR can load country totals, distribute them on the grid and calculate zonal totals.
8. The comparison of national inventories with EDGAR data indicate some areas for possible improvement of the IPCC methodology. Especially methodology for estimating methane from agriculture, methane from biofuel combustion, methane from land use change, methane from landfills and methane from waste water treatment could be improved.
9. For comparison of national and global inventories: use IPCC standard data tables.
10. For comparison of national inventories with global Top down model results three levels of detail are possible for methane: global totals, zonal averages and local comparisons with geographical detail (gridded).
11. For Africa, Asia, and Latin America long term measurements in polluted areas are lacking for this kind of local comparisons.
12. For gridded comparisons the national inventories could be disaggregated to a grid and to monthly totals for groups of sources to compare with model results. The distribution functions used in the EDGAR database could be used for this.

4. Analysis for nitrous oxide

4.1 Introduction

Nitrous oxide (N_2O) is, like CO_2 and CH_4, one of the natural components of the earth's atmosphere. It is a greenhouse gas and it plays an important role in the chemistry of the stratosphere. As a result of human activities, its concentrations have been increasing since the beginning of the century. Nitrous oxide is responsible for about 4-6% of the present radiative forcing. The direct Global Warming Potential (over 100 years) amounts to 310 (when $CO_2 = 1$) (IPCC, 1995).

Nitrous oxide in the atmosphere is largely of biogenic origin. Bacteria in soils and oceans release N_2O during nitrification and denitrification. Human activities tend to enhance biogenic production of N_2O. Agriculture, land use change, deforestation and nitrogen fixing processes all stimulate bacterial production of N_2O. In addition, several anthropogenic abiogenic sources can be distinguished, including industrial and combustion processes.

During the 1980s and 1990s several emission inventories have been published. In this chapter, these inventories will be analyzed. First, an overview will be given of available material that can be used for top-down/bottom-up comparisons (section 4.2). This overview will be focused on the recent inventories as well as on monitoring of concentrations in the atmosphere. Next, the inventories will be analyzed in more detail in section 4.3 (top-down analysis) and 4.4 (bottom-up analysis). This analysis will focus on the way in which data are presented and the uncertainties involved, and may lead to a standard presentation of emission data to enable inventory comparisons. Conclusions will be drawn in section 4.5.

4.2. Availability of information

4.2.1 Emission inventories

A top-down with bottom-up emission inventory comparison is based on available information. National Communications as summarized in the UNFCCC Second review and synthesis report for the Second Conference of the Parties in Geneva (UNFCCC/CP/1996/12/Add. 2) and US country study results as summarized by Braatz *et al.* (1996) will be compared with EDGAR data (Olivier *et al.*, 1996). Top-down models for N_2O will be described and a method for comparison will be developed. Available models of atmospheric budgets of nitrous oxide are from Hartley and Prinn (1983) and Bouwman and Taylor (1996).

Global inventories: During the last fifteen years or so several global inventories have been published and compared to global budgets derived from atmospheric measurements. Natural emissions are from soils (Bouwman *et al.*, 1993) and oceans (Nevison *et al.*, 1994) mainly. Anthropogenic sources include agriculture, energy use, industry, waste and some other minor sources (e.g. Bouwman *et al.*, 1995; Nevison *et al.*, 1994; Kroeze, 1994; and IPCC 1990, 1992, 1995). Some of these are analyzed in more detail below.

EDGAR and GEIA: The EDGAR inventory is described by Olivier *et al.* (1996). It is a database with most important sources of emissions of greenhouse gases and other pollutants like NO_x, SO_2, VOC with sectoral detail for all countries in the world. EDGAR results are presented and

on a grid of 1 x 1 degree longitude and latitude. The GEIA database is also on a grid of 1 x 1 degree. It has no sectoral detail and no information on emission factors and activity levels. The EDGAR inventory is identical to the GEIA inventory for N_2O and includes natural and anthropogenic sources. These are described in Olivier *et al.* (1996) and Bouwman *et al.* (1995). For a description of EDGAR and GEIA see paragraph 2.1.1 and 2.1.2 and for a description of the methodology of EDGAR for N_2O see paragraph 4.4.3.

National inventories: A number of countries published their National Communications, as presented in section 4.4. Most of these used the 1995 IPCC Guidelines for National Greenhouse Gas Inventories for estimating their emissions. Some other countries used in-country or alternative methods for estimating their emissions, e.g. the Netherlands (Kroeze, 1994; Van Amstel *et al.*, 1994). In addition, some countries calculated their emissions based on the revised IPCC Guidelines for National Greenhouse Gas Inventories (IPCC/OECD, 1997) or new in-country measurements, including China, Belgium, Bulgaria (Kroeze and Bogdanov, 1997) and India (Mitra ed, 1994).

4.2.2 Monitoring concentrations in the atmosphere

Analysis of ancient air in Antarctic ice reveals that in pre-industrial times, the atmospheric concentration of N_2O was in the range of 270 - 290 ppbv (Weiss 1981b; Pearman *et al.*, 1986; Brasseur and De Rudder, 1987; Wigley 1987; Khalil and Rasmussen 1988). Around 1900 the concentration amounted to 293 ppbv. The present-day concentration of 311 ppbv has not been experienced during 45,000 years (Leuenberger and Siegenthaler, 1992).

Systematic measurements of atmospheric concentrations of N_2O on a global scale have been carried out since the mid-seventies at the ALE/GAGE and CMDL networks (Trends '93; Figure 4.2 and Table 4.1). Concentrations are measured in weekly flask samples at 6 CMDL ground-based stations: Alert, North Pole (90° N); Barrow, Alaska (71° N); Niwot Ridge, Colorado (40 °N); Mauna Loa, Hawai (20° N); Samoa (14° S) and South Pole (90° S); and several times per day at five ALE/GAGE monitoring sites: Adrigole, Ireland (52° N); Cape Meares, Oregon (45° N); Ragged Point, Barbados (13° N); Point Matalula, American Samoa (14° S) and Cape Grim, Tasmania (45 °S). All stations are global background stations and are located at remote areas (notably coastal sites) far away from source areas.

These measurements indicate that the nitrous oxide concentrations have been increasing at about 0.2 - 0.3% per year (about 0.8 ppbv/y) during the last decades (Figure 4.1). Trends over the last decade were found to be relatively variable: over 3-year periods the trends have ranged from 0.5 ppbv/yr to 1.2 ppbv/yr (Khalil and Rasmussen, 1992).

Moreover, the observations show that a North-South gradient is not very well established. This is a result of a long atmospheric lifetime (110 - 180 years) and sources evenly distributed over the earth. Nevertheless, a small interhemispheric difference is observed: concentrations on the Northern Hemisphere exceed those on the Southern by about 1 ppbv (0.3%) (Weiss, 1981a,b; Khalil and Rasmussen, 1983a; Prinn *et al.*, 1990). There are some indications that in pre-industrial times the interhemispheric difference was the opposite, with Southern Hemispheric concentrations exceeding Northern Hemisperic by 1-2 ppbv (Khalil and Rasmussen, 1988). This is in line with an increase in anthropogenic emissions, located mainly at the Northern Hemisphere (e.g. Bouwman *et al.*, 1995).

Atmospheric concentrations show relatively little spatial and temporal variation. Although biogenic production of N_2O is affected by temperature and precipitation changes and thus by seasonal and diurnal cycles, no clear trends have been observed. Nitrous oxide seems to be well-mixed throughout the troposphere. In the stratosphere, however, concentrations decrease with height to negligible levels from 45 km altitude (Ko and Sze, 1977; Roy, 1979).

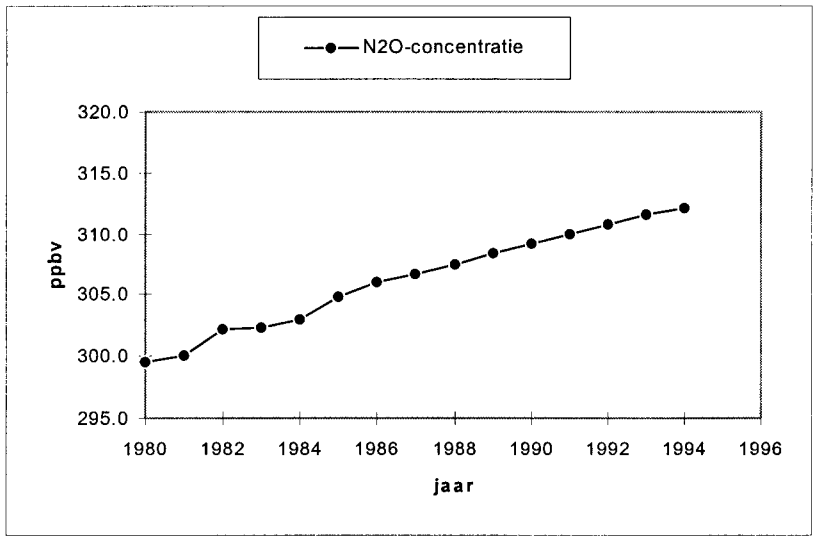
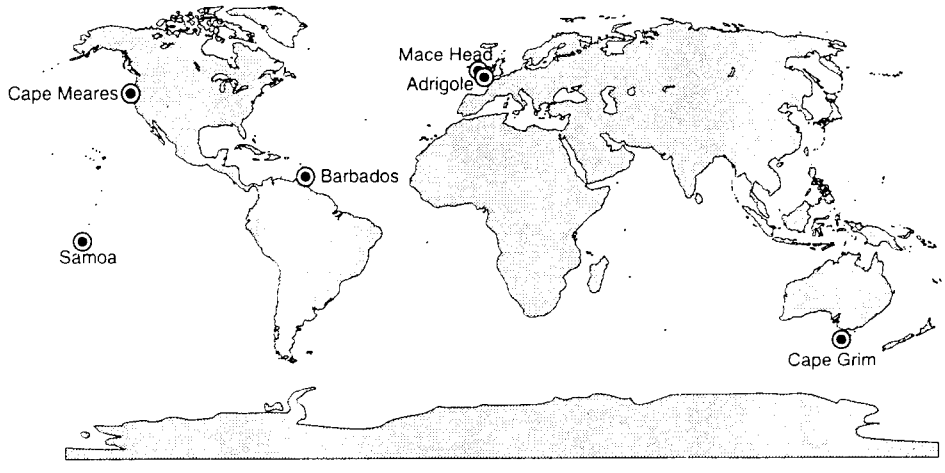


Figure 4.1 Trend of the N_2O concentration as measured at Barbados

Systematic measurements of atmospheric concentrations of N_2O on a global scale are carried out in the ALE/GAGE and CMDL network (Trends '93), see figure 4.2 and table 4.1.



ALE/GAGE monitoring sites.

Figure 4.2 ALE/GAGE and NOAA/CMDL network sites.

Table 4.1 Overview of the N_2O monitoring networks ALE/GAGE and CMDL

Reference	network	number of stations
Prinn *et al.* (1990)	ALE/GAGE	6
Bodhaine and Rosson (1988)	NOAA/CMDL	5

4.3 Top down modelling

4.3.1 Inverse modelling for N_2O

The analysis for N_2O differs from the analysis of CO_2 and CH_4 in the sense that

1) virtually all sources are located at the earth's surface and N_2O can be considered chemically inert in the troposphere,
2) there is only one major relatively constant sink for N_2O: the stratosphere, where N_2O is removed by photolysis and reaction with excited oxygen atoms, and
3) as a result of which the annual variation in concentrations are caused by variations in emissions (natural and anthropogenic) rather than in the sinks (as with CO_2).

Inverse modelling of N_2O at a regional level is hampered by the small spatial variation in atmospheric concentrations (see above) and the limited number of atmospheric measurement stations. Nevertheless, inverse modelling studies have been performed at the global scale (Khalil and Rasmussen, 1992) and for four zonal bands (Prinn *et al.*, 1990). In addition, a 3-dimensional atmospheric transport models were used by Bouwman and Taylor (1996) and Nevison (1994) to simulate atmospheric concentrations. These studies are discussed in more detail in the following sections.

4.3.2 Analysis of the global budgets of N_2O

One-box atmospheric model

Khalil and Rasmussen (1992) discussed the uncertainties in inverse modelling on a global scale. Part of the uncertainty in emission estimates is caused by the annual variations in concentrations that are not further discussed. Khalil and Rasmussen (1992) argue that the uncertainties in source estimated follow from the uncertainty in (i) the atmospheric lifetime and (ii) the absolute concentrations and trends. However, they also show that these uncertainties mainly affect the estimates for *total* and *natural* emissions, while the estimated *anthropogenic* emissions are relatively insensitive to these parameters, except the trend. This can be illustrated by a 0-dimensional atmospheric box model (equations 1 and 2; modified from Khalil and Rasmussen, 1992):

$$S_n * F = C_n / T \quad [1]$$

$$S_a * F = dC_a/dt - C_a/T \quad [2]$$

where

C_n = concentration due to natural sources (ppbv)
C_a = concentration change due to human activities (ppbv)
S_n = natural emissions (Tg N)
S_a = anthropogenic emissions (Tg N)

τ = atmospheric lifetime (years)
t = time (years)
F = conversion factor (Tg/ppbv): Khalil and Rasmussen (1992) assume that 1 Tg N_2O/y from the earth's surface equals to about 6.35 ppbv/y in the troposphere (here we assume that 1 ppbv = 4-5 Tg N/y).

This simple model indicates that natural emissions amount to 6 - 13 Tg N/y, assuming that that pre-industrial concentrations (270 - 290 ppbv, see above) were not affected by human activities and that the atmospheric lifetime is within the range of 110 to 170 years (Prinn *et al.*, 1992; Minschwaner *et al.*, 1993; IPCC, 1995). The uncertainty range is largely affected by the uncertainty in the atmospheric lifetime, which is deduced from the troposphere-stratosphere flux and the strength of the stratospheric sink are known (observations and model calculations).

Equation 2 indicates that anthropogenic emissions (S_a) are within the range of 2 - 8 Tg N/y. This estimate largely depend on the annual change in atmospheric N_2O (dC_a/dt) which amounts to 2 - 6 Tg N/y (three-year averages of 0.5 - 1.2 ppbv/y; Khalil and Rasmussen, 1992). The value of C_a/τ ranges between 0.5 and 1.9 Tg N/y, as calculated from a 41 - 21 ppbv increase in atmospheric N_2O (C_a) to 311 ppbv and a 110 - 170 year atmospheric lifetime (see above).

Using the longer-term averaged concentration increase of 0.8 ppbv/y, Khalil and Rasmussen (1992) conclude that the average anthropogenic emissions amounted to 4.5 (3.9 - 5.1) Tg N/y. The 4 - 5 Tg N is a remarkebly small uncertainty range, considering the large uncertainties in bottom-up estimates, as shown in Table 4.4. For instance, the latest IPCC Scientific Assessment estimates total anthropogenic sources in the range of 3.7 - 7.7 Tg N/year (IPCC, 1995).

Until the late 1980s coal combustion was considered to be the main source of anthropogenic N_2O, as reflected in Khalil and Rasmussen's a priori estimate of 2.3 Tg N/y (Table 4.4). Their analysis indicated that the fossil fuel source could be considerably lower if emissions from land use change and agriculture were higher than previously estimated. Presently the generel view is that indeed agriculture is the most important anthropogenic source of N_2O, emitting about 3.5 Tg N/y (IPCC, 1995), while coal combustion is considered a very small source of N_2O. The 1.3 Tg N reported by IPCC (1995) is mainly from traffic and industry (adipic acid and nitric acid production).

1-D, 2-D and 3-D Models

Tables 4.2 and 4.3 overview the 2-D model of Prinn *et al.* (1990) and the 1-D model of Khalil and Rasmussen (1992) that were used in inverse mode for calculation of emissions from atmospheric concentrations. Total N_2O emissions, atmsopheric increase and stratospheric destruction are matched to close the global N_2O budget (Table 4.2).

The models of Prinn *et al.* (1990) and Khalil and Rasmussen (1992) have been run in the inverse mode and emissions have been estimated to fit the overal concentration field, the averaged yearly increase (0.8 ppb) and/or the zonal concentration gradient (1 ppb). These global N_2O models assume a troposphere-stratosphere exchange rate and a stratospheric destruction rate which both determine the effective atmospheric lifetime t_e. The effective atmospheric lifetime is a measure of the strenght of the N_2O sink. Prinn *et al.* (1990) assume an effective lifetime

between 150 and 182 years derived from 2-D and 3-D model calculations. Khalil and Rasmussen (1992) assume a lifetime of 150 years. These lifetimes are relatively long if compared to more recent estimates of 120 years as published by Minschwaner *et al.* (1992) and IPCC (1995).

Bouwman and Taylor (1996) and Nevison (1994) use 3-D atmospheric transport models to calculate atmospheric concentration fields using a gridded emission database as input to the model. In both studies temporal trends in emissions were assumed. The stratospheric loss rates were chosed such that the fit present-day atmospheric concentrations.

Table 4.2 One-, two- and three-dimensional models used to analyze the N_2O budget

model type	spatial resolution	temporal resolution	period	Reference
2-D model	8 tropospheric boxes and 1 stratospheric box	monthly	1978-1988	Prinn *et al.* (1990)
1-D model	1 troposphere and 1 stratosphere box	yearly	1976-1987	Khalil and Rasmussen (1992)
3-D model	R15 (about 4.5° latitude x 7.5° longitude and 12 vertical layers	monthly	1990	Nevison (1994)
3-D model	2.5° latitude x 2.5° longidtude and 7 vertical layers	monthly	1987-1991	Bouwman and Taylor (1996)

Table 4.3 Summary of N_2O budget as published in several studies (see Table 4.2 for details on the models); "-" means not available

Reference	lifetime (years)	sink (Tg N/y)	source (Tg N/y)			atmospheric increase (Tg N/y or ppbv/y)
			natural	anthropogenic	total	
			calculated from atmospheric concentrations			
Composite (see text)	110-170	-	6 - 13	2 - 8 [1)]	8 - 21	0.5 - 1.2 ppbv/y [1)]
Prinn *et al.* (1990)	166	-	-	-	13 (11.5-14.5) [2)]	-
Kahlil *et al.* (1992)	150	-	9.5	4.5 (3.9-5.1) [3)]	-	0.8 ppbv/y
IPCC (1995)	120	12.3 (9-16) [4)]	-	-	16.2 (13-20)	3.9 (3.1-4.7)
			bottom-up inventories			
Nevison (1994)	-	11.0	9.4	6.0	15.4	4.4
Bouwman and Taylor (1996)	-	10.5 [5)]	9.3 [5)]	4.3 [5)]	13.6 [5)]	3.1 [5)]
IPCC (1995)	120	-	9 (6-12) [4)]	5.7 (3.7-7.7) [4)]		3.9 [4)]

[1)] 3-years average

[2)] 10 year average

[3)] Derived from the tropospheric trend 1977-1987

[4)] Last decade (1984-1994)

[5)] 1976-1987

Table 4.4 Global inventories of N_2O emission for major source categories as published by several authors, based on inverse modelling (adjusted) or bottom-up inventories (*a priori*)

	anthropogenic sources			natural sources		
	combustion + industry ($Tg N_2O$)	land use change ($Tg N_2O$)	agriculture ($Tg N_2O$)	temperate soils ($Tg N_2O$)	tropical soils ($Tg N_2O$)	ocean ($Tg N_2O$)
Prinn *et al.* (1990) *a priori*	1.5	1.6	1.0	1	4	2.5
Prinn *et al.* (1990) *adjusted*	<1	2-4	>1	1	4	2.5
Khalil *et al.* (1992) *bottom-up inventory*	1.0	1.7	2.4	-	8 [1)]	2
Nevison (1994) *a priori*	0.7	0.6	4.7	-	5.6 [1)]	3.8
Bouwman and Taylor (1996) *a priori* [2)]	0.8	0.6	3	-	5.7 [1)]	3.6
IPCC (1995) *bottom-up inventory*	1.3 (0.7-1.8)	0.5 (0.2-1.0)	3.9 (2-5.8)	2 (0.6-4)	4 (2.7-5.7)	3 (1-5)

[1)] including temperate soils

[2)] based on the EDGAR/GEIA annual inventory

Interannual variations

The atmospheric concentrations indicate that there is some interannual variability in N_2O emissions (Figure 4.3; Prinn *et al.* 1990; Khalil and Rasmussen, 1992). This could have several reasons. Assuming that stratospheric removal is a relatively constant process, the variability is most probably due to changes in annual emissions. As mentioned earlier, both natural and anthropogenic emissions are of biogenic (bacterial) origin. Bacterial activity is affected by local temperature and precipitation. Thus interannual differences in weather conditions may affect annual emissions. Another reason may be interannual differences in human activities.

Figure 4.3 shows the yearly variations of the total N2O emissions derived by Prinn *et al.* (1990). Note that units are in Tg N_2O (to be multiplied by 28/44 for conversion to Tg N).

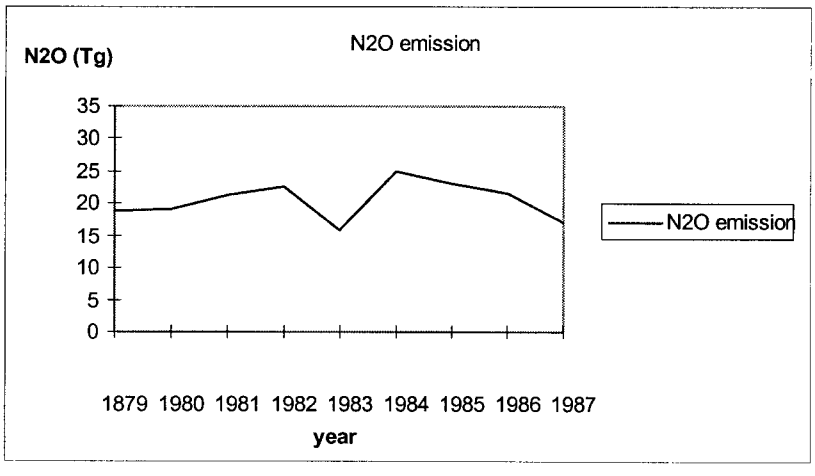


Figure 4.3 Total N_2O emissions for the period 1979 - 1987 as derived by Prinn *et al.* (1990).

4.3.3 Analysis of the zonal budgets of N_2O

Prinn *et al.* (1990) used inverse modelling techniques to estimate N_2O emission for four latitudinal belts. They calculate emissions for the regions 30-90° N (22-34% of total emissions), 0-30° N (32-39%), 0-30° S (20-29%) and 30-90° S (11-15%). Both Nevison (1994) and Bouwman and Taylor (1996) use these distributions to evaluate their assumed emissions, and both are in general agreement with Prinn *et al.* (1990). Nevison (1994) and Bouwman and Taylor (1996) were able to realistically simulate the interhemispheric 1 ppbv difference in N_2O concentrations.

Prinn *et al.* (1990) used a priori and adjusted emission inventories for their calculations. These were used to derive a latitudinal distribution (10 degree bands) of emissions per source as reflected in figures 4.4 and 4.5. These figures show the importance of natural emissions from tropical soils in the total emissions. Anthropogenic emissions are found to be mainly located at the Northern Hemisphere. For comparison the zonal distribution of EDGAR sources is also given in Figure 4.11.

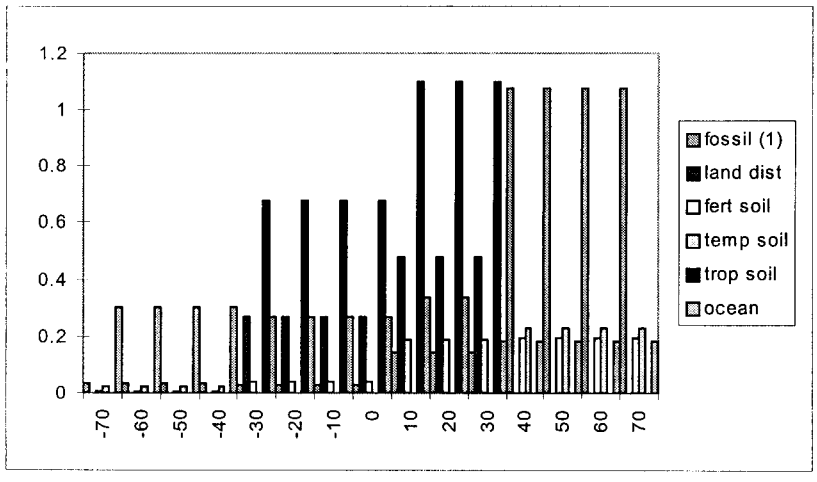


Figure 4.4 A priori emission estimate based on inverse modelling by Prinn *et al.* (1990)

Latitudinal distribution of most important sources of N_2O, modified by Janssen (1997) from the a priori emission estimate by Prinn *et al.* (1990). Fossil (1) includes coal combustion. Other sources are land use change (land dist), agriculture (fert soil) and natural sources (temperate soils, tropical soils and oceans).

Prinn reduces (assuming a large stratospheric fluxe in the NH) the *a priori* contribution of (NH) fossil fuel (1) emission and increased the (tropical) land sources (2)

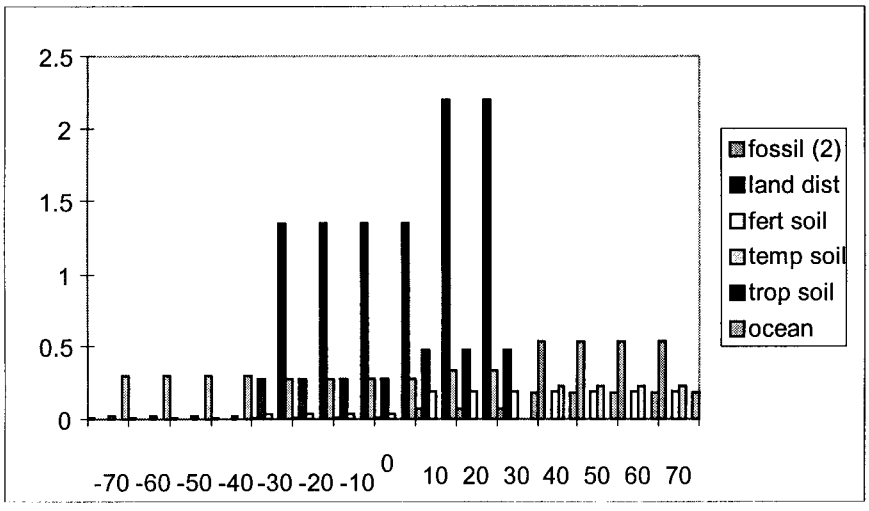


Figure 4.5 Latitudinal distribution of most important sources of N_2O. Adjusted *a priori* emission estimate based on inverse modelling by Prinn *et al.* (1990).

Latitudinal distribution of most important sources of N_2O, modified by Janssen (1997) from the adjusted a priori emission estimate by Prinn *et al.* (1990). Fossil (2) includes coal combustion in Prinn *et al.*'s study but is presently considered to be from traffic and industry mainly. Other sources are land use change (land dist), agriculture (fert soil) and natural sources (temperate soils, tropical soils and oceans).

4.3.4 Conclusions on the budget analysis for N_2O

Inverse modelling results indicate that global anthropogenic emissions of N_2O amount to 4-5 Tg N per year(Prinn *et al.* 1990). The uncertainty in bottom-up inventories is 3-8 Tg N according to IPCC (1995) which exceeds the range deduced from atmospheric measurements. Thus for global inventories the atmospheric data can be used to validate bottom-up emission estimates.

Inverse modelling of N_2O at a zonal scale is hampered by the small number of monitoring stations (only 11) and the relative small latitudinal gradient (0.3%). Nevertheless, Prinn *et al.* (1990) derived budgets for four zonal regions which were used by others to validate their global inventories. These analyses confirm that emissions of N_2O are largely of biogenic origin, with agriculture as the main contributor to anthropogenic emissions.

The observed interrannual variation in N_2O concentrations is most probably due to yearly variations in the (notably biogenic) sources.

4.3.5 Trajectory analysis

In this first phase of the study no trajectory analysis was found for N_2O. In the second phase this will be further looked into.

4.4. Evaluation of emission inventories

4.4.1 Introduction

In this section national emission inventories as reported to the Climate Convention Secretariat (National Communications) will be compared to the emissions as available in EDGAR. The GEIA inventory is identical to the EDGAR inventory. For this purpose, first a general description will be given of the methodologies used for emission estimates in the IPCC Guidelines (4.4.2) and EDGAR (4.4.3). Next, the country emissions will be compared (4.4.4).

4.4.2 IPCC Methodology for N_2O

The IPCC Guidelines provide methodologies for estimating national N_2O emissions from energy (stationary and mobile combustion), industry, agriculture, burning of savannas and agricultural wastes. Most countries used the 1995 version of the IPCC Guidelines for National Greenhouse Gas Inventories for their N_2O inventories (IPCC/OECD/IEA, 1995). The revised 1996 IPCC Guidelines, however, include a completely different method for agricultural emissions and sewage treatment (IPCC/OECD/IEA, 1997). Since agriculture is considered to be the most important anthropogenic source on a global scale, this may have important implications for the national inventories.

Since most countries used the 1995 IPCC Guidelines, we will only analyze this version of the IPCC Guidelines. In the following a brief summary is given. For details is refered to IPCC/OECD/IEA (1995) and Kroeze (1995).

Energy-related emissions are estimated in the IPCC Guidelines as a function of fuel input. Emission factors are available for stationary combustion of coal, oil and gas, which were adopted in the EDGAR method (Table 4.5), and for various vehicle types. The IPCC Guidelines moreover provide emission factors for industrial emissions due to production of adipic acid and nitric acid, which differ only slightly from those used in the EDGAR inventory (Table 4.6). The emissions from burning of savannas and agricultural wastes are calculated as a ratio to the nitrogen emitted, which in turn is calculated from the CO_2 emissions and the C/N ratio. The agricultural emissions are in the 1995 IPCC Guidelines simply estimated as a fraction of the nitrogen input to agricultural soils, where the nitrogen input is from fertilizers and biological N_2 fixation.

Kroeze (1995) concluded that the 1995 IPCC Guidelines did not consider the following sources of N_2O: nitrogen leaching from soils, other nitrogen inputs to surface waters, wastewater treatment, atmospheric deposition of NOx and NH_3, manure in stables, atmospheric formation and use in anaesthesia. The 1996 revised IPCC Guidelines include the missing agricultural sources emissions and sewage treatment. The non-agricultural sources, however, are still not considered.

4.4.3 EDGAR Methodology for N_2O

Fossil fuel combustion

The activity data for fossil fuel combustion as used in EDGAR are described in section 2.4. In EDGAR, default and globally uniform emission factors were derived for combustion of coals, oil products and natural gas, respectively, from the existing data (Olivier, 1994); these are also recommended in the IPCC Guidelines. Emission factors for gasoline cars equipped with catalytic convertors are known to show substantially higher N_2O emissions than vehicles without catalytic converters. The use of these convertors is still limited to a few countries. Based on information on the penetration rate of gasoline cars with catalysts we have used a set of country specific factors to estimate these emissions from 1971 to 1992. Table 4.5 summarizes the EDGAR factors used for 1990. These factors were also used in the GEIA N_2O inventory.

Table 4.5 EDGAR emission factors for N_2O from fossil fuel combustion in 1990 (in g N_2O-N/GJ)

Sector/fuel	Emission factor	Remark (range)	Reference
SOLID FUELS	0.8909	1.4 g N_2O/GJ x 28/44 = 0.890909 g N_2O-N/GJ (range for 1.4 is: 0-10 g/GJ)	IPCC, 1994 [5]
LIQUID FUELS	0.3818	0.6 g N_2O/GJ x 28/44 = 0.381818 g N_2O-N/GJ (range for 0.6 is: 0-2.8 g/GJ).	IPCC, 1994 [5]
GASEOUS FUELS	0.06364	0.1 g N_2O/GJ x 28/44 = 0.063636 g N_2O-N/GJ (range for 0.1 is: 0-1.1 g/GJ)	IPCC, 1994 [5]
Exceptions:			
Road transport-gasoline [3]	2.673	USA, Canada, Japan [1]	Hawker, 1990
	1.145	Australia [2]	Nat. GHG Ctee, 1994
	1.145	Germany (Fed. Rep.) [2]	UBA, 1994
	0.8095	Former DDR [2]	UBA, 1994
	0.7255	Netherlands [2]	CBS, 1995
Domestic air-jet fuel [4]	2.14	0.15 g N_2O/kg = 3.4 g/GJ = 2.14 g N_2O-N/GJ	Wiesen *et al.*, 1994
International air-jet fuel [4]	2.14	0.15 g N_2O/kg = 3.4 g/GJ = 2.14 g N_2O-N/GJ	Wiesen *et al.*, 1994

[1] In 1980 a global default value of 0.3818 for USA, CAN, JPN, increased linearly from 1980 to the 1990 value of 2.673 and keeping constant from 1990.

[2] In 1985 a global default value of 0.3818 for AUS, DEU, DDR and NLD, increased linearly from 1985 to their 1990 values as listed here. From 1990 to 1992 these values increase to 1.527, 1.527, 1.023 and 0.9545, respectively.

[3] Process code: E.TP1.TRA.MOG.ROA

[4] Process codes: E.TP1.TRA.JET.AIR and E.TP1.TRA.JET.INT, respectively.

[5] IPCC 1994 default emission factors.

Biofuels

The EDGAR activity data for biofuels are described in section 2.4. The emission factors in EDGAR for N_2O from biofuels are from Veldt and Berdowski (1995), except that Smith *et al.* (1993) was used for N_2O from fuelwood. Note that the N_2O factor for fuelwood in IPCC (1994) is about 50% higher than ours.

Industrial processes: adipic acid and nitric acid

Adipic Acid (AA) production data are primarily based on the production capacity of plants given by Castellan *et al.* (1991). For manufacturing of nitric acid (HNO_3 or NA), which is mainly used as feedstock in fertilizer production, global production estimates from UN statistics (UN, 1993) and by the industry (McCulloch, 1993, pers. comm.) are inconsistent. Therefore, we adopted statistics of N-fertilizer production as a correlate for NA production (IFA, 1992). This may cause differences compared to what countries use in National Inventories. EDGAR emission factors of N_2O for adipic acid and nitric acid production are based on Reimer *et al.* (1992).

Table 4.6 EDGAR emission factors for N_2O from industrial processes in 1990

Sector/product	Emission factor	Unit	Region	Reference
Adipic acid	147.1	kg N_2O-N/ton	World [1)]	Reimer *et al.*, 1992
Nitric acid	16.0	kg N_2O-N/ton N	World	Reimer *et al.*, 1992
[1)] Exceptions:				
Adipic acid	184.9	kg N_2O-N/ton	Canada	Reimer *et al.*, 1992
Adipic acid	110.0	kg N_2O-N/ton	USA	Reimer *et al.*, 1992

Landuse and waste treatment

Landuse and waste treatment sources in EDGAR include fertilizer use (N_2O), animals (CH_4 and N_2O), biomass burning (all gases) and agricultural waste burning. Waste water and sewage treatment, which are considered to be sources of nitrous oxide, are not included because to date no representative spatial emission estimates exist. Sources of activity data are described in Section 3.4.

For animals, the EDGAR emission factors for N_2O are described in Bouwman *et al.* (1995). For biomass burning, including agricultural waste burning, the EDGAR emission factor for N_2O was taken from Crutzen and Andreae (1990) are described in detail in Bouwman *et al.* (1995).

4.4.4 Country comparison

In table 4.7 an overview is given of differences between National Communications (NCs) and EDGAR data. Table 4.8 and Figure 4.6 show the same information for different world regions, while also showing estimates for six different sources, following the IPCC sector categories.

The present analyses focuses on differences between EDGAR and National Communications for different groups of countries: Eastern Europe (Figure 4.7), European Union (Figure 4.8), rest of OECD (Figure 4.9) and rest of world (see for instance Table 4.8 and Figure 4.10). The most important sources of nitrous oxide are agriculture and industry (Table 4.8). In industry nitric and adipic acid production are the main sources. According to IPCC (1995) worldwide anthropogenic emissions amount to 5.7 Tg N per year, or 8960 Gg N_2O per year (Table 4.8). The totals for the EDGAR database (5700 Gg N_2O) and the National Communications (1700 Gg

N_2O) are much lower than the IPCC total. Table 4.8 shows that the differences are most pronounced for agricultural emissions.

For EU-15 and Eastern Europe, EDGAR and the National Communications are in general agreement with respect to total emissions (Table 4.8; Figure 4.6). However, there are considereble differences for emissions from different sectors for Eastern Europe. For OECD, the EDGAR total estimate is about twice the NC estimate. And the differences are even larger for the total OECD and the rest of the world (RoW1, RoW2).

Table 4.7 Total nitrous oxide emissions in 1990 in OECD and Eastern European countries.

Comparison of EDGAR and National Communications.

	Total NC Gg N2O/y	Total EDGAR Gg N2O/y	Difference Gg/yr	Difference %
Australia	60,1	69,0	8,9	13%
Austria	4,1	7,0	2,9	41%
Belgium		15,0	15,0	
Bulgaria	22,5	15,0	-7,5	-50%
Canada	95,5	369,0	273,5	74%
Czech Republic	24,0		-24,0	
Sloval Republic	16,0		-16,0	
Czechoslovakia	40,0	22,0	-18,0	-82%
Denmark	10,3	13,0	2,7	21%
Estonia	2,4		-2,4	
Finland	22,0	143,0	121,0	85%
France	176,7	125,0	-51,7	-41%
Former FRG			0,0	
Former DDR			0,0	
Germany	211,0	126,0	-85,0	-67%
Greece	13,7	14,0	0,3	2%
Hungary	11,4	12,0	0,6	5%
Iceland	0,6	1,0	0,4	40%
Ireland	42,3	14,0	-28,3	-202%
Italy	120,3	44,0	-76,3	-173%
Japan	55,2	51,0	-4,2	-8%
Latvia	2,4		-2,4	
Liechtenstein	0,1		-0,1	
Luxembourg	0,6	0,0	-0,6	
Monaco			0,0	
Netherlands	51,5	19,0	-32,5	-171%
New Zealand	17,1	41,0	23,9	58%
Norway	15,0	21,0	6,0	29%
Poland	156,0	81,0	-75,0	-93%
Portugal	10,5	15,0	4,5	30%
Romania	106,8	30,0	-76,8	-256%
Russian Federation	89,6	181,0	91,4	50%
Spain	93,9	39,0	-54,9	-141%
Sweden	15,2	171,0	155,8	91%
Switzerland	15,6	4,0	-11,6	-290%
United Kingdom	108,3	110,0	1,7	2%
United States	411,4	532,0	120,6	23%

Table 4.8 Nitrous oxide 1990 emissions per sector (Gg N_2O/y) for different world regions as estimated in EDGAR and National Communications (NC). For each sector the IPCC sector code is given between brackets [1A to 6]. Global IPCC emissions are from IPCC (1995); n.a. = not available.

		Energy [1A]	Industry [2]	Solvents [3]	Agriculture andland use [4]+[5]	Land use [5]	Waste [6]	Total
NC	EU-15	145	243	2	380	9	6	780
EDGAR	EU-15	168	200	0	337	0	0	705
NC	E.Eur.	69	13	0	51	1	0	134
EDGAR	E.Eur.	52	11	0	118	0	0	181
NC	rest of OECD	206	137	2	297	5	7	648
EDGAR	rest of OECD	160	623	0	503	0	0	1287
NC	RoW1	56	0	0	80	9	0	137
EDGAR	RoW1	64	51	0	658	16	0	773
NC	RoW2	0	0	0	0	0	0	0
EDGAR	RoW2	151	175	0	2450	55	0	2776
NC	World	476	393	4	808	24	13	1699
EDGAR	World	595	1060	0	4064	71	0	5722
IPCC	World	2000 [1)]	n.a.	n.a.	6885	785	n.a.	8960

[1)] including industrial emissions

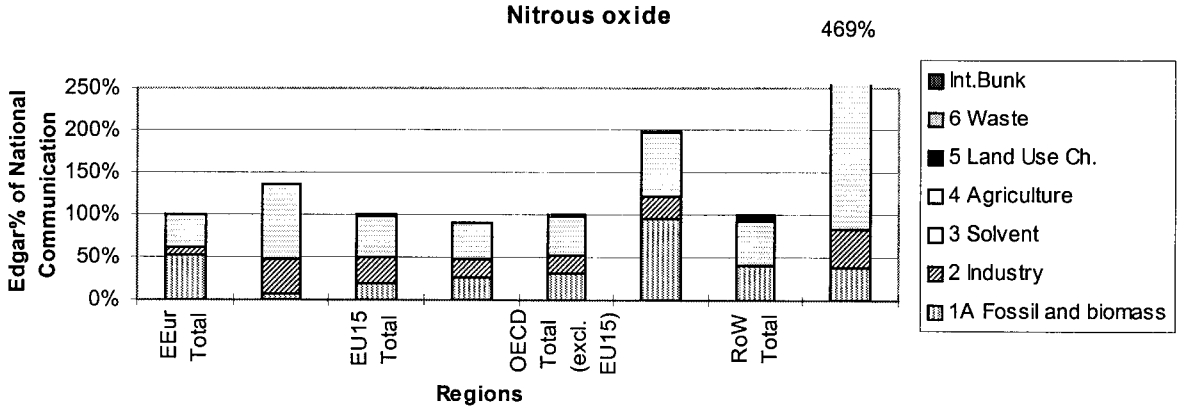


Figure 4.6 Comparison of N_2O emissions as estimated in National Communications (NatComm) and EDGAR for several world regions.

In the above graph the first bar is the sum of national inventories of Eastern European countries. The second bar is the sum of the same countries from EDGAR data. The third bar is the sum of national inventories of the European Union 15 countries. The fourth bar is the same from EDGAR data. The fifth bar is the sum of national inventories from the rest of the OECD countries. The sixth bar is the same from EDGAR data. The seventh bar is the sum of national inventories from the US country studies programme. The eighth bar is the sum of all countries other than EU15, OECD, Eastern Europe from the EDGAR data. Sum of national inventories in each region was set to 100%. Estimates are for the following sectors: Nitrous oxide emissions from combustion of international marine and aviation bunker fuels; nitrous oxide emissions from waste treatment; nitrous oxide from land use change; nitrous oxide from agriculture (fertilizer and manure application); nitrous oxide from solvent use (anesthesia use); nitrous oxide from industrial processes (nitric and adipic acid production); nitrous oxide from combustion of fossil fuels and biofuels.

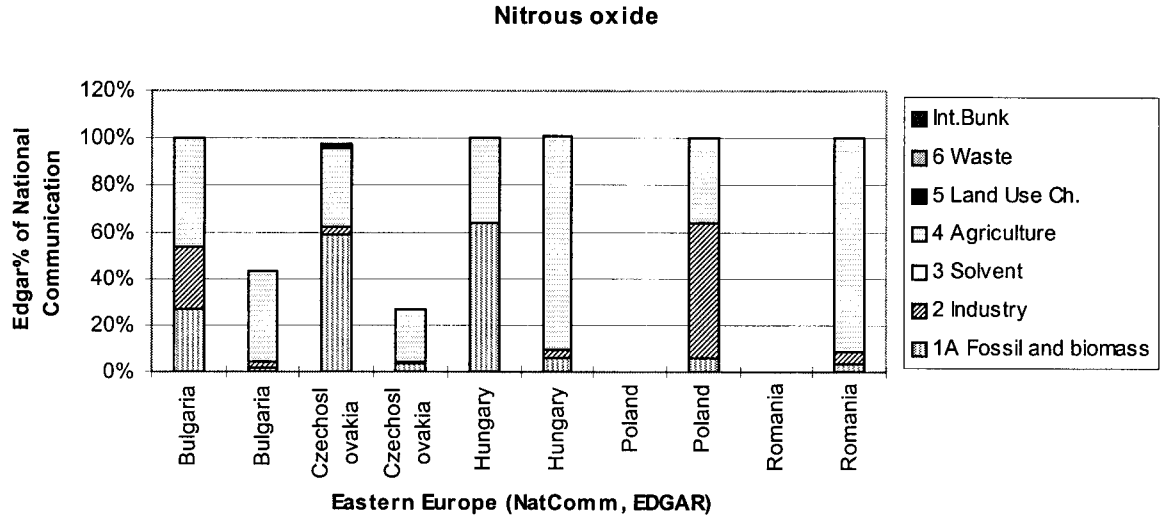


Figure 4.7 Comparison of N_2O emissions as estimated in National Communications (NatComm) and EDGAR for Eastern Europe.

In the above graph the first bar is the national inventory of Bulgaria. The second bar is the total of the same country from EDGAR data, etc.. The national inventory was set to 100%. Estimates are for the following sectors: Nitrous oxide emissions from combustion of international marine and aviation bunker fuels; nitrous oxide emissions from waste treatment; nitrous oxide from land use change; nitrous oxide from agriculture (fertilizer and manure application); nitrous oxide from solvent use (anesthesia use); nitrous oxide from industrial processes (nitric and adipic acid production); nitrous oxide from combustion of fossil fuels and biofuels.

In the following the EDGAR and National Communications estimates are compared for the IPCC emission categories 1A (fossil fuel combustion), 2 (Industry) and 4 (Agriculture). The countries for which the differences exceed 20% are listed. Explaining these differences needs further detailed analysis of the background documents which is outside the scope of this study. Nevertheless, some preliminary interpretation of the differences may follow from sections 4.4.2 and 4.4.3.

Fossil fuel combustion [IPCC sector 1A]

The differences between EDGAR and National Communication estimates were more than 20% for the following countries: Bulgaria, Czech and Slovak Republic, Hungary in Eastern Europe; Austria, Finland, Greece, Italy, Sweden, Portugal in Europe; Canada, Japan, New Zealand and Norway in the rest of OECD. In the rest of the world absolute amounts in this category are small. The differences may be due to different activity data used, or different emission factors, in particular for coal combustion and for road traffic.

Industrial processes [IPCC sector 2]

Differences between National Communications and EDGAR were more than 20% for the following countries: Bulgaria, Poland in Eastern Europe; France, the Netherlands, United Kingdom in Europe; Japan and Norway in the rest of OECD; and Algeria and Russia in the rest

of the world.

Reasons for differences may be partly caused by the fact that EDGAR neglects the efforts countries take in reducing their emissions from adipic acid production. This could explain the difference for the UK. Other reasons could be differences in statistics on nitric acid production.

Agriculture [IPCC sector 4]

Differences between the National Communications and EDGAR were more than 20% for the following countries: Hungary, Poland, Romania in Eastern Europe; Austria, Denmark, Greece, Ireland, Italy, Portugal, Spain and the United Kingdom in Europe; Canada, Japan, New Zealand, Switzerland and the United States in the rest of the OECD; and all considered countries from the US country studies programme. In the latter group of countries EDGAR was consistently higher than the NCs.

The most likely reason for the difference is the fact that most countries used the 1995 IPCC Guidelines for their estimate, while this method was not complete (see 4.4.2). EDGAR on the other hand, includes a much more complete method for agricultural emissions. Another reason may be that statistics about fertiliser application from the FAO are used in EDGAR, which may be different from what countries used.

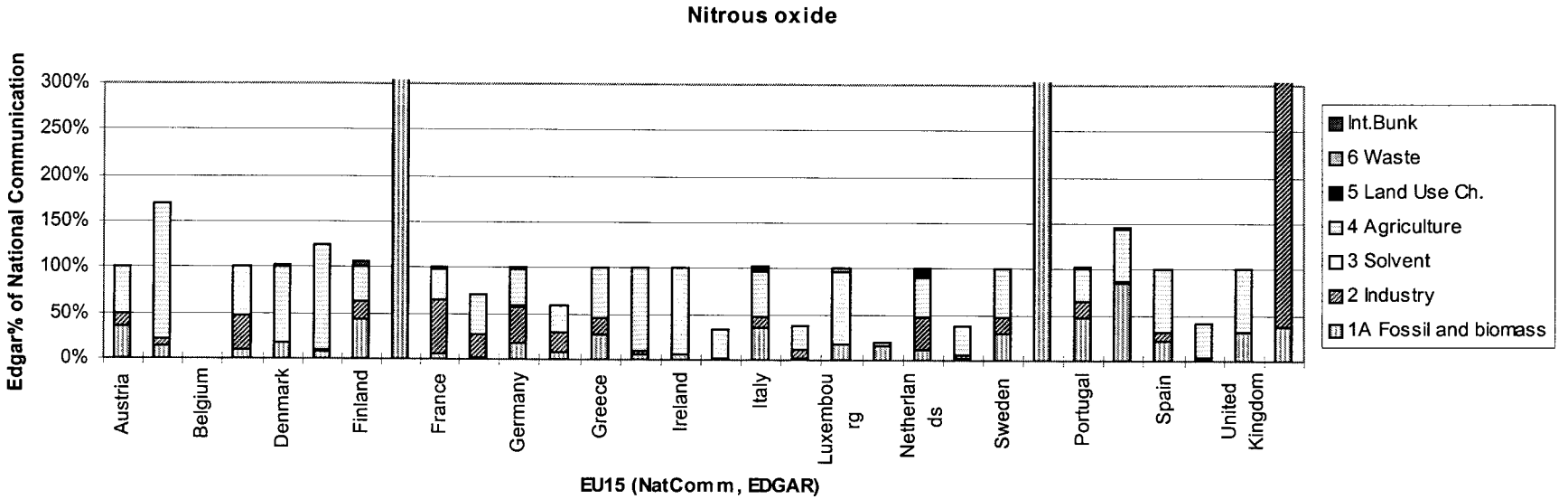


Figure 4.8 Comparison of N_2O emissions as estimated in National Communications (NatComm) and EDGAR for the 15 countries of the European Union.

The first bar is the national inventory of Austria. The second bar is the same country from EDGAR data, etc. Total of national inventory was set to 100%. Estimates are for the following sectors: Nitrous oxide emissions from combustion of international marine and aviation bunker fuels; nitrous oxide emissions from waste treatment; nitrous oxide from land use change; nitrous oxide from agriculture (fertilizer and manure application); nitrous oxide from solvent use (anesthesia use); nitrous oxide from industrial processes (nitric and adipic acid production); nitrous oxide from combustion of fossil fuels and biofuels.

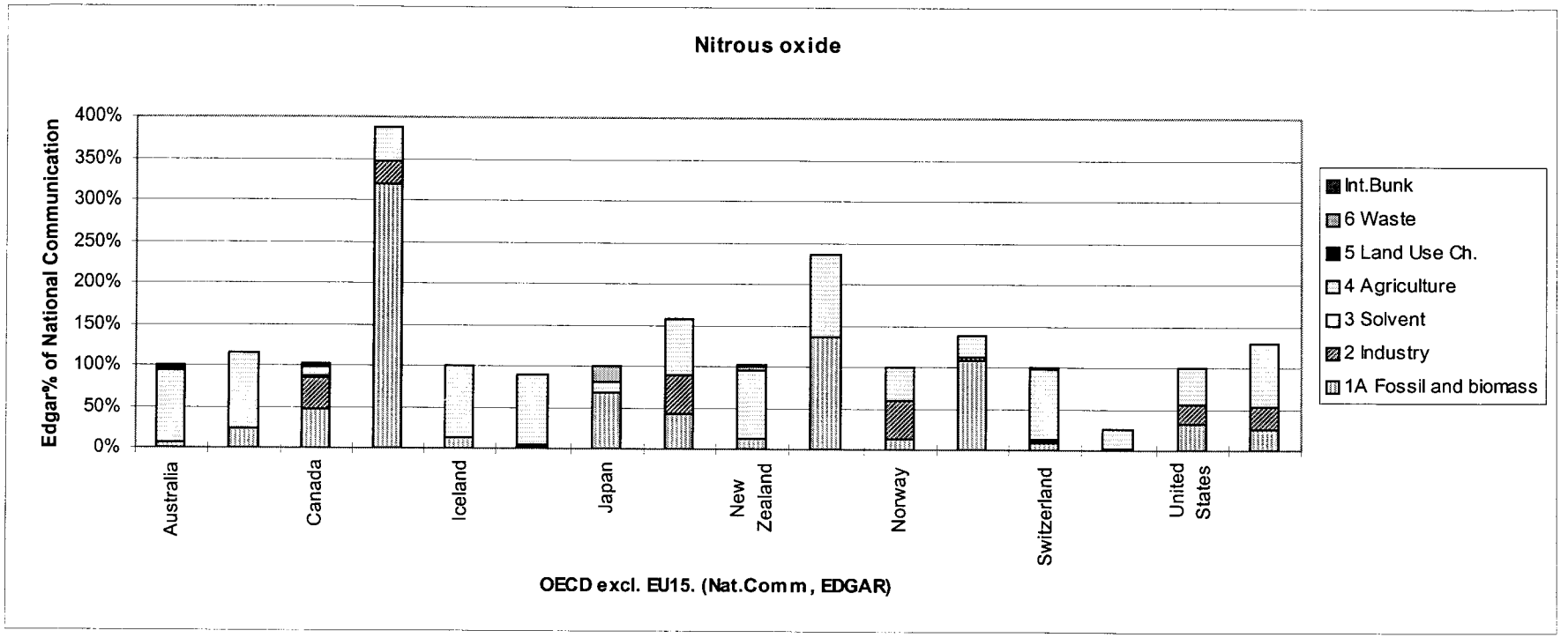


Figure 4.9 Comparison of N_2O emissions as estimated in National Communications (NatComm) and EDGAR for the rest of the OECD.

The first bar is the national inventory of Australia. The second bar the same country from EDGAR data, etc. Total of national inventory was set to 100%. Estimates are for the following sectors: Nitrous oxide emissions from combustion of international marine and aviation bunker fuels; nitrous oxide emissions from waste treatment; nitrous oxide from land use change; nitrous oxide from agriculture (fertilizer and manure application); nitrous oxide from solvent use (anesthesia use); nitrous oxide from industrial processes (nitric and adipic acid production); nitrous oxide from combustion of fossil fuels and biofuels.

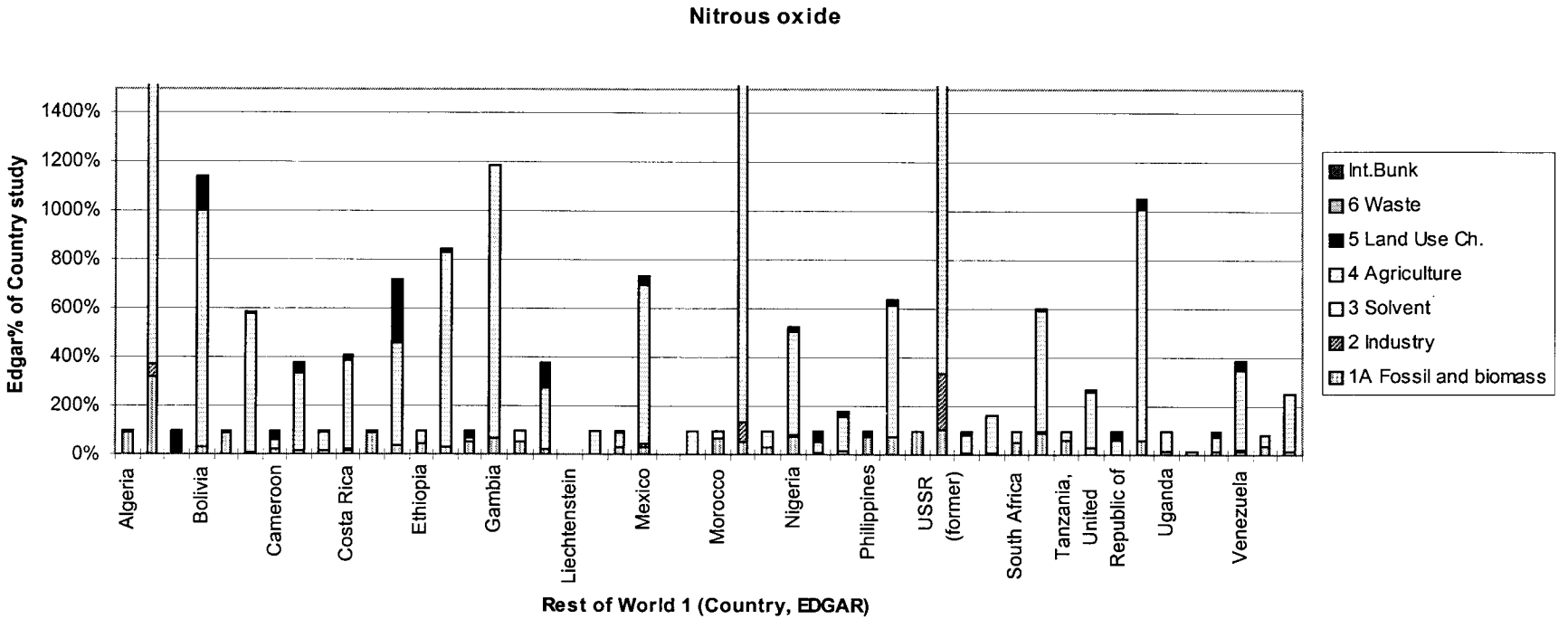


Figure 4.10 Comparison of N_2O emissions as estimated in US Country Studies Programme and EDGAR for Rest of the World 1.

The first bar is the US country study of Algeria. The second bar the same country from EDGAR data, etc. Total of country studywas set to 100%. Estimates are for the following sectors: Nitrous oxide emissions from combustion of international marine and aviation bunker fuels; nitrous oxide emissions from waste treatment; nitrous oxide from land use change; nitrous oxide from agriculture (fertilizer and manure application); nitrous oxide from solvent use (anesthesia use); nitrous oxide from industrial processes (nitric and adipic acid production); nitrous oxide from combustion of fossil fuels and biofuels.

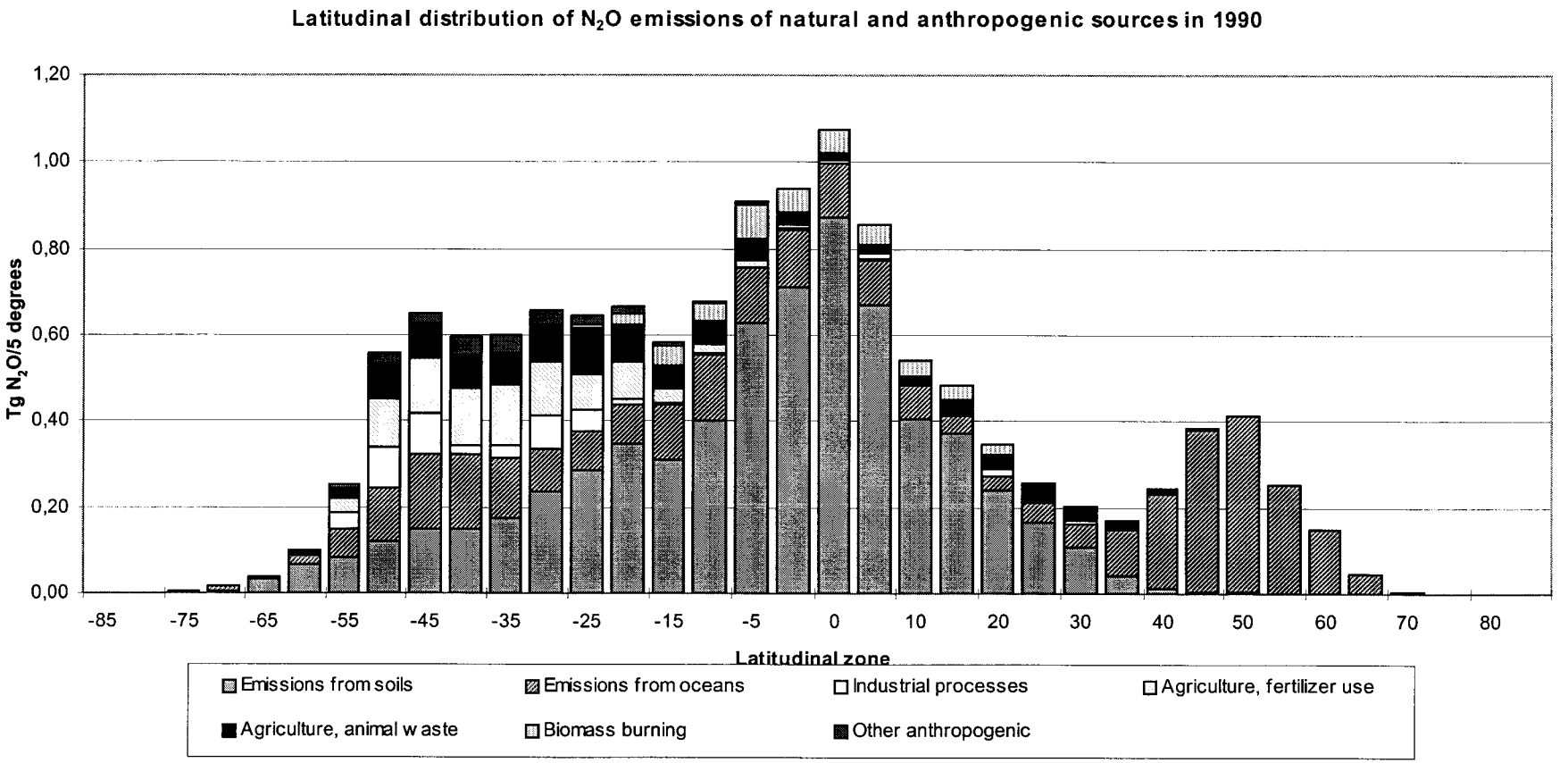


Figure 4.11 Zonal distribution of anthropogenic N_2O emissions in 1990 according to EDGAR. (-90 = North, 90 = South)

4.5 Conclusions for nitrous oxide

The research questions as formulated in section 1.2 will be answered for N_2O in the following.

Available inventories

For N_2O several inventories are available on a global, zonal, gridded an national scale. Inverse modelling was used to derive global and zonal budgets. In addition, 3-D atmospheric transport models were used to simulate atmospheric concencatration fields using gridded emission inventories as input. There are two databases including national inventories for a large number of countries: National Communications (mostly derived by using the 1995 IPCC Guidelines) and EDGAR/GEIA.

Presentation of emission data

The emission data presented in g N_2O or in g N which can be confusing when inventories are compared. The National Communications present their emissions according to the IPCC emission categories. EDGAR data are presented for different source categories per country or on a grid of 1° by 1° longitude and latitude.

A method for comparison of emission inventories

From this chapter can be concluded that for N_2O inverse modelling is only useful on a global or zonal scale. For zonal comparisons the following latitudinal belts seem appropriate: 30-90° N, 0-30° N, 0-30° S and 30-90° S.

On a lower aggregation level (country of regional scale) only comparisons of bottom-up inventories are useful. A comparison seems most useful on a national as well as regional level. In this preliminary study the following regions were identified: EU-15, Eastern Europe, OECD, Rest of the World (1 and 2). Furthermore, it seemed useful to report emissions for the sectors as distinguished in the IPCC Guidelines (energy, industry, solvents, agriculture, land use and waste) (see Appendix I for a short description of each of these sectors).

Uncertainties

The uncertainties in inventories have not been studied systematically. The data presented here indicate that the least uncertain emission estimates are probably global totals for anthropogenic emissions derived from trends in atmospheric N_2O (4-5 Tg N/year). Using the trend in the atmospheric increase (4-5 Tg N/year) and the best estimate of the stratospheric sink (12-13 Tg N/year), it could be shown from top-down analysis that a large part (50%) of the global anthropogenic N_2O emission derived from bottom-up emission estimates was missing. Based on this information, a careful bottom-up analysis of all possible sources led to identification of a new source of N_2O emissions: cattle and feedlots. This is a clear example of how top-down estimates can improve the bottom-up emission estimates.

Other estimates are surrounded with relatively large uncertainties. This is a result of the fact that

the stratospheric removal cannot be accurately quantified. In addition, most of the atmospheric N_2O is from biogenic origin and biogenic source show large variability, both spatial and temporal. However, when reasonably described, the latter can help to distinguish between man-made and natural sources.

Prior to 1988 coal combustion was thought the most important anthropogenic source of N_2O. However, nowadays there is wide consensus that N_2O formation in electricity generation or fossil fuel combustion in general, is relatively small and that the majority of atmospheric N_2O is of biogenic origin. Nevertheless, Bouwman *et al.* (1996) concluded that the new source candidates identified in recent years did not reduce the uncertainty in the various source estimates. In the 1990 and 1992 IPCC Scientific Assessments it was concluded that the observed concentration increase could not be easily explained by the known sources (IPCC 1990, 1992). Recently, however, Mosier *et al.* (in preparation) developed a new method for estimating N_2O emissions from agriculture for the IPCC Guidelines for National Greenhouse Gas Inventories, using which the atmospheric increase may be explained to a reasonable extent (IPCC, 1995).

Preliminary comparison

Global scale: Atmospheric concentration trends indicate that the anthropogenic emissions are within the range of 4-5 Tg N per year (6 - 8 Tg N_2O per year). The total for EDGAR is at the lower end of this range. The total for the National Communications and other country studies (1.7 Tg N_2O per year) is much lower, indicating that more than 70% of the anthropogenic emissions have not been reported in National Communications. This is in agreement with the EDGAR estimate for countries that have not reported yet, those countries show the largest emissions in agriculture.

Zonal scale: In addition, inverse modelling by Prinn *et al.* (1990) indicates how N_2O emissions are distributed over four zonal bands. These results are in agreement with the zonal distribution of the gridded inventory by Bouwman *et al.* (1995), which is almost similar to the EDGAR database.

Regional scale: Comparison for world regions showed that for EU-15 and Eastern Europe the total estimates by EDGAR and NCs are in general agreement, while for the other world regions the totals differed by at least a factor of two.

National scale: Comparing National Communications to EDGAR reveals that for many countries the differences exceed 20%. The largest differences were found for agricultural emissions and are most probably caused by differences in methodology (emission factors and sources included).

Recommendations for methodology development

In this preliminary study we did not analyse the observed difference in detail. Clearly, further in-depth analysis is needed to explain the difference between EDGAR and National Communications and other inventories. Nevertheless, the preliminary comparison indicates that

differences between EDGAR and NCs were large for most sources of N_2O. Therefore, it seems useful and needed to evaluate the methodologies used. Agriculture is considered the most important anthropogenic source on a global scale and them differences were particularly large for this source. Although the revised 1996 IPCC Guidelines may reduce the difference between EDGAR and NCs it is still a source worth evaluating. It is furthermore recommended to focus on industrial processes, biomass burning and waste.

5. Discussion and conclusions

5.1 Conclusions for each gas

Conclusions for carbon dioxide

The research questions as formulated in chapter 1.2 will be answered for CO_2 in the following:

Available inventories
For CO_2 the following measurements and models were used: Tans (1989, 1990), Keeling (1993), Conway *et al.* (1994), Kaminski *et al.* (1997) and Ciais *et al.* (1995, 1997a, 1997b). The inventories were used from the Climate Secretariat database, completed with the US Country Study results as summarised by Braatz *et al.* (1996). They were compared with the EDGAR data from Olivier *et al.* (1996).

Presentation of inventory data
National inventory data are presented per sector per year according to the IPCC recommended summary table format. EDGAR data are presented per sector per year according to own sector definitions. These could however be translated easily to IPCC recommended format.

A method for comparison of emission inventories
Concerning the top-down bottom-up comparison of emission inventories for carbon dioxide it can be concluded that global comparisons are possible, but by doing this the uncertainty can not be reduced while the model results have an uncertainty of 25-30% and the national inventories of the single largest source, namely fossil fuel combustion have an uncertainty of about 10%.

Comparisons of bottom-up inventories like the comparison of national inventories with EDGAR are feasible and useful. The recommended format for the comparison is the summary tables 7A and 7B of the IPCC Guidelines. Comparisons can be done by country and by groups of countries. Here the following groups were used: EU15, other OECD, Eastern Europe and Russia, Rest of the World 1 (US country studies), and Rest of the World 2 (only EDGAR data available).

Uncertainties
The uncertainties are seldom quantified in the inventories. It is 10% in the single largest source: carbon dioxide from fossil fuel combustion. The uncertainties in the other sectors are not quantified but much larger. The uncertainties in the national inventories can not be reduced by validation with model studies, while these are larger. Uncertainties can be reduced by deterministic studies and model development to get a grip on the processes of carbon dioxide exchange between atmosphere and biosphere and oceans.

Preliminary comparison
The country comparison with EDGAR showed that differences are large in all important sectors even in Europe, although the totals match more closely. In a second phase the reasons for these differences have to be investigated. The differences were large especially in the land use change sector, the agriculture sector and the biofuels combustion sector.

Recommendations for methodology development
In this preliminary study we did not investigate the reasons for observed differences in detail. Clearly, a further in-depth analysis is needed. A database of emission factors is needed to carry out this in-depth study. The recommended format for the comparison is the IPCC summary tables 7A and 7B. Target areas for potential IPCC methodology improvement are in the following sectors: land use change, agriculture and biofuel combustion.

Other conclusions
Because global carbon cycle models ignore short term perturbations due to fluctuations in the climate system they are not able to reproduce these shorter-term atmospheric CO_2 variations. It is therefore not possible to estimate fossil fuel emissions derived from atmospheric measurements and global CO_2 (Top-down) budget calculations for a year with more accuracy then based on Bottom-up emission inventories.

The four main sources of uncertainty for CO_2 budgets are:

a) The measuring dataset and representativeness of the measuring data, because of poor world coverage;
b) Whether the ocean or the biosphere is the main sink;
c) Yearly variations in sources and sinks;
d) Different budget assumptions between various authors.

Due to large year to year variations in net fluxes a zonal comparison of Top-down model results with Bottom-up inventories for CO_2 for one year can not lead to validation of these Bottom-up inventories.

Yearly variations in the CO_2 budget are much larger than the yearly variations in the fossil fuel emissions.

Long term trends of the CO_2 concentration in the atmosphere are representative of the long term disturbance of the carbon balance due to anthropogenic CO_2 emissions.

The uncertainty in the estimated CO_2 emission due anthropogenic activities derived from atmospheric measurements is 25-30%.

The comparison of national inventories with EDGAR data has pointed to some areas for potential improvement in the IPCC methodology for CO_2, namely parts of the fuel use sectors, the land use change sector, the agriculture sector and the biofuels combustion sector.

The comparison of national inventories with EDGAR data will be completed in the second phase of the project, when overviews of activity data and emission factor data are made.

Conclusions for methane

The research questions as formulated in chapter 1.2 will be answered for CH_4 in the following:

Available inventories
For methane many different model studies are available. Here we have used the global model synthesis by Fung *et al.* from 1991, Lelieveld and Crutzen 1993, Muller 1993, Hein *et al.* 1994 and 1997, The and Beck 1995, and Saeki 1997. For local validation studies for NW Europe we have used various studies in the Netherlands based on four measurement sites

(Hollander and Vosbeek, 1996; Vermeulen *et al.* 1996; Janssen *et al.* 1997). Two databases of national inventories have been used: the Climate Secretariat database of national inventories and the EDGAR database. The summary of US country studies by Braatz *et al.* 1996 has been incorporated.

Presentation of inventory data
National inventory data are presented per sector per year according to the IPCC recommended summary table format. EDGAR data are presented per sector per year according to own sector definitions. These could however be translated easily to IPCC recommended format.

A method for comparison of emission inventories
Concerning the top-down bottom-up comparison of emission inventories for methane it can be concluded that global, regional and gridded comparisons are possible, but by doing this the uncertainty can not be reduced while both model results and national inventories have the same overall uncertainty of about 20-35%. Gridded comparisons using data assimilation with a Kalman filter, are only feasible in Europe and North America at this moment, while long term high frequency measurements are still lacking in the other regions.

Comparisons of bottom-up inventories like the comparison of national inventories with EDGAR are feasible and useful. The recommended format for the comparison is the summary tables 7A and 7B of the IPCC Guidelines. Comparisons can be done by country and by groups of countries. Here the following groups were used: EU15, other OECD Europe, Eastern Europe and Russia, OECD North America, OECD Pacific, Latin America, Africa, Middle East, India region, China region, East Asia, Rest of the World.

Uncertainties
The uncertainties are seldom quantified in the inventories. From some experimental studies we know that uncertainties in the range of 20-35% for all sources. The uncertainties in the national inventories can not be reduced by validation with model studies, while these are of the same magnitude. Uncertainties can be reduced by deterministic studies and model development to get a grip on the processes of methane formation in soils. To reduce uncertainties more measurements of the type of the Cabauw site (at 200 m height) are needed in regions other than Europe and North America.

Preliminary comparison
The country comparison with EDGAR showed that differences are large in all important sectors even in Europe, although the totals match more closely. In a second phase the reasons for these differences have to be investigated.

Recommendations for methodology development
In this preliminary study we did not investigate the reasons for observed differences in detail. Clearly, a further in-depth analysis is needed. A database of emission factors is needed to carry out this in-depth study. Target areas for IPCC methodology improvement are in the following sectors: agriculture, land use, biofuel combustion, landfills and waste water treatment.

Other conclusions
The short term variations in the atmospheric increase of the CH_4 concentration are in the order of magnitude of the yearly increase.

The long term atmospheric increase of CH_4 concentrations is representative of CH_4 emissions from human activities.

The uncertainty of total CH_4 emissions due to human activities derived from atmospheric measurements is in the order of 35% (derived from the uncertainty of emissions estimates of 40-50 Tg CH_4 and uncertainty of the OH sinks which is about the same).

For specific sectors the "a priori" emission estimates can be reduced by 30%. This is the case in rice, biomass burning and landfills. Reduction is reached by inverse modelling using measurement results.

The comparison of top-down with bottom-up emission inventories is possible on a local scale for NW-Europe because long term measurement results are available.

Trajectory analysis with one measuring station like Cabauw is capable of assessing the national inventories for surrounding countries up to a distance of 500 km. The sectoral detail is less than the inventories but the totals can at least be validated.

The comparison of inventories with models for zonal bands is possible but the uncertainty in the models is the same or higher as national emission inventories, around 35%.

The EDGAR database could be used for a zonal comparison with various country estimates. EDGAR can load country totals, distribute them on the grid and calculate zonal totals.

The comparison of national inventories with EDGAR data indicates some areas for possible improvement of the IPCC methodology. Especially methodology for estimating methane from agriculture, methane from fossil fuel production and transmission, methane from biofuel combustion, methane from land use change, methane from landfills and methane from waste water treatment could be improved.

For a first comparison of national and global inventories: Use IPCC standard data tables.

For comparison of national inventories with global top down model results three levels of detail are possible for methane: global totals, zonal averages and local comparisons with geographical detail (gridded).

For Africa, Asia, and Latin America long term measurements in polluted areas are lacking for this kind of local comparisons.

For gridded comparisons the national inventories could be disaggregated to a grid and to monthly totals for groups of sources to compare with model results. The distribution functions used in the EDGAR database could be used for this.

Conclusions for nitrous oxide

The research questions as formulated in section 1.2 will be answered for N_2O in the following.

Available inventories

For N_2O several inventories are available on a global, zonal, gridded an national scale. Inverse modelling was used to derive global and zonal budgets. In addition, 3-D atmospheric transport models were used to simulate atmospheric concentration fields using gridded emission

inventories as input. There are two databases including national inventories for a large number of countries: National Communications (mostly derived by using the 1995 IPCC Guidelines) and EDGAR/GEIA.

Presentation of emission data

The emission data presented in g N_2O or in g N which can be confusing when inventories are compared. The National Communications present their emissions according to the IPCC emission categories. EDGAR data are presented for different source categories per country or on a grid of 1° by 1° longitude and latitude.

A method for comparison of emission inventories

From chapter 4 can be concluded that for N_2O inverse modelling is only useful on a global or zonal scale. For zonal comparisons the following latitudinal belts seem appropriate: 30-90° N, 0-30° N, 0-30° S and 30-90° S.

On a lower aggregation level (country of regional scale) only comparisons of bottom-up inventories are useful. A comparison seems most useful on a national as well as regional level. In this preliminary study the following regions were identified: EU-15, Eastern Europe, OECD, Rest of the World (1 and 2) (see Appendix X for a list of the countries included). Furthermore, it seemed useful to report emissions for the sectors as distinguished in the IPCC Guidelines (energy, industry, solvents, agriculture, land use and waste) (see Appendix X for a short description of each of these sectors).

Uncertainties

The uncertainties in inventories have not been studied systematically. The data presented here indicate that the least uncertain emission estimates are probably global totals for anthropogenic emissions derived from trends in atmospheric N_2O (4-5 Tg N/year). Using the trend in the atmospheric increase (4-5 Tg N/year) and the best estimate of the stratospheric sink (12-13 Tg N/year), it could be shown from top-down analysis that a large part (50%) of the global anthropogenic N_2O emission derived from bottom-up emission estimates was missing. Based on this information, a careful bottom-up analysis of all possible sources led to identification of a new source of N_2O emissions: cattle and feedlots. This is a clear example of how top-down estimates can improve the bottom-up emission estimates.

Other estimates are surrounded with relatively large uncertainties. This is a result of the fact that the stratospheric removal cannot be accurately quantified. In addition, most of the atmospheric N_2O is from biogenic origin and biogenic source show large variability, both spatial and temporal. However, when reasonably described, the latter can help to distinguish between man-made and natural sources.

Prior to 1988 coal combustion was thought the most important anthropogenic source of N_2O. However, nowadays there is wide consensus that N_2O formation in electricity generation or fossil fuel combustion in general, is relatively small and that the majority of atmospheric N_2O is of biogenic origin. Nevertheless, Bouwman *et al.* (1996) concluded that the new source candidates identified in recent years did not reduce the uncertainty in the various source estimates. In the 1990 and 1992 IPCC Scientific Assessments it was concluded that the observed concentration increase could not be easily explained by the known sources (IPCC 1990, 1992). Recently, however, Mosier *et al.* (in preparation) developed a new method for estimating N_2O

emissions from agriculture for the IPCC Guidelines for National Greenhouse Gas Inventories, using which the atmospheric increase may be explained to a reasonable extent (IPCC, 1995).

Preliminary comparison

Global scale: Atmospheric concentration trends indicate that the anthropogenic emissions are within the range of 4-5 Tg N per year (6 - 8 Tg N_2O per year). The total for EDGAR is at the lower end of this range. The total for the National Communications and other country studies (1.7 Tg N_2O per year) is much lower, indicating that more than 70% of the anthropogenic emissions have not been reported in National Communications. This is in agreement with the EDGAR estimate for countries that have not reported yet, those countries show the largest emissions in agriculture.

Zonal scale: In addition, inverse modelling by Prinn *et al.* (1990) indicates how N_2O emissions are distributed over four zonal bands. These results are in agreement with the zonal distribution of the gridded inventory by Bouwman *et al.* (1995), which is almost similar to the EDGAR database.

Regional scale: Comparison for world regions showed that for EU-15 and Eastern Europe the total estimates by EDGAR and NCs are in general agreement, while for the other world regions the totals differed by at least a factor of two.

National scale: Comparing National Communications to EDGAR reveals that for many countries the differences exceed 20%. The largest differences were found for agricultural emissions and are most probably caused by differences in methodology (emission factors and sources included).

Recommendations for methodology development

In this preliminary study we did not analyse the observed difference in detail. Clearly, further in-depth analysis is needed to explain the difference between EDGAR and National Communications and other inventories. Nevertheless, the preliminary comparison indicates that differences between EDGAR and NCs were large for most sources of N_2O. Therefore, it seems useful and needed to evaluate the methodologies used. Agriculture is considered the most important anthropogenic source on a global scale and them differences were particularly large for this source. Although the revised 1996 IPCC Guidelines may reduce the difference between EDGAR and NCs it is still a source worth evaluating. It is furthermore recommended to focus on industrial processes, biomass burning and waste.

Main conclusions

An international programme to review and evaluate national inventories of greenhouse gases is useful for several reasons:

- The *bottom-up* comparison of greenhouse gas emission inventories (in-depth studies of emission inventories) is a powerful method for improvement of plausibility, consistency, accuracy, and appropriateness of IPCC and other national methodologies. It provides a discussion platform and identifies areas for future improvement of the IPCC methodologies.

- Exchange, review and comparison of data promotes dialogue, data sharing and consensus on the data among scientists and policy-makers.
- Using both *bottom-up* and *top-down* types of emission data improves the scientific understanding of the global total budgets, increases the quality of emission data and refines methodologies to compile national emissions inventories, thereby increasing confidence and credibility in the emissions inventory process.
- Evaluation of different *bottom-up* and *top-down* emission data sets increases the credibility of emission inventories, which may facilitate development of climate policy and measures.
- The use of atmospheric measurements of greenhouse gases together with models appears to be an objective tool for monitoring progress in attaining national and global emission reduction goals and may develop into a verification mechanism.

Top-down

It is possible to evaluate greenhouse gas emissions on a lesser than global scale e.g. regional or national, using direct measurements and atmospheric models if the required accurate measurements are available. However, at the moment measurement data of greenhouse gas concentrations come from a limited number of remote background stations. Measurements on small spatial and temporal scales, notably in emission source regions, are lacking for most world regions. For CO_2 ,only the global budget can be evaluated. No accurate estimation of anthropogenic sources at zonal or regional level can be made on the basis of the existing atmospheric measurements due to the overriding influence of the large fluxes of CO_2 between the different reservoirs (atmosphere, ocean and biosphere) which also vary in time. For CH_4 global, zonally averaged and several regional emissions can be estimated on the basis of measurements and model calculations. At the moment the regional detail is only feasible for Europe. For N_2O only global and (limited) zonally averaged emissions can be estimated or evaluated at the moment.

Bottom-up

National emission inventories, to be prepared by all countries signing the Framework Convention of Climate Change (FCCC) as part of their National Communications, are now available for many industrialised countries. However, a substantial number of National Communications from developing countries are still lacking. Global *bottom-up* emission data are collected by using the EDGAR database. About 65% of the global CO_2 emission, 55% of the global CH_4 emission and less than 30% of the global N_2O emissions are estimated to have been reported in the first National Communications submitted in 1994 and other country studies. Source categories of IPCC Guidelines and EDGAR have been made compatible. Results of a *bottom-up* versus *bottom-up* comparison of sectoral emissions data from National Communications and EDGAR data have shown occasional substantial differences for specific greenhouse gases between sectors and countries. Analysis has revealed *bottom-up* comparison of greenhouse gas emission inventories (scoping and in-depth studies of emissions inventories) to be a powerful method for improving plausibility, consistency, accuracy, and suitability of IPCC Guidelines and other national methods to estimate emissions. As it provides insight into the transparency of the inventory calculations, it supports scientific dialogue and provides a discussion platform for future updates of the IPCC methodology.

Comparison of Top-down versus Bottom-up emission estimates

Various models and datasets are available for a *top-down* versus *bottom-up* comparison of emission inventories. Models used in the Netherlands are the EUROS, LOTOS, and COMET models for Europe; the IMAGE, DIALOOG, TM2 and MOGUNTIA model for the world. In

this report we used results from the literature of these models and of various other global climate models. Datasets which contain global measurement data are the CDIAC and WMO-WCDCGG databases. The NOP-CH_4 dataset at RIVM contains CH_4 concentration data of Europe with high spatial and temporal resolution. The most important datasets on national emissions carrying sectoral detail and information on emission factors and activity data are the National Communications database of the UNFCCC Secretariat, the IEA database on energy use, the FAO database on agriculture and forestry, and the EDGAR database of RIVM/TNO on emissions, per country and per gridcell on 1^0x1^0 longitude and latitude. In order to be able to compare *bottom-up* and *top-down* results it is necessary to make these datasets compatible. In the following table the initial differences are given.

Table 51. Differences between Bottom-up and Top-down methods

	Bottom-up inventories	Top-down models
Gas	CO_2 CH_4 N_2O	CO_2 CH_4 N_2O
Emission	per sector	per source category
Spatial scale	country	global/zonal/regional/grid
Time scale	base year	any period from hour to year

Evaluation of the greenhouse gas budget using information from measurements and atmospheric transport models, can be employed to test the plausibility of emission inventories. Measurements of radioactive and stable isotopes can be used to provide information on specific source categories, even on a regional scale. However, the capability of the *top-down* method to derive detailed information on emissions on a small spatial and temporal scale is limited at the moment (Table 5.2.)

Table 5.2 Maximum attainable temporal and spatial resolution for inverse modelling

	CO_2	CH_4	N_2O
Spatial resolution	global (zonal)	global, zonal or 1^0x1^0	global and zonal
Temporal resolution	5-10 years average	year (1990)	year (1990)
Period	5 years	year	year
Source categories	fossil fuel	anthropogenic	anthropogenic
	land-use change	natural	natural
	vegetation		

National inventories (National Communications) are available for most countries for 1990 and 1995. Since EDGAR 2.0 emission data have 1990 as a base year a comparison with model results is recommended for those years. For CO_2 ,however, deforestation and land cover and land use data are available only for 5-10 year averages and the exchange between atmosphere, biosphere and ocean fluctuates substantially from year to year. Therefore is recommended to use only 5-10 year periods to compare *bottom-up* and *top-down* data.

In developing a standard methodology for *top-down/bottom-up* comparisons national totals per sector of any dataset, e.g. official UNFCCC submissions, need to be distributed over a global grid, thereby providing means to compare results of *bottom-up* estimates with those of a *top-down* analysis using global dispersion models. In this report the unique facility of the

EDGAR software was used to convert country totals per sector into global 2-D distributions and 1-D latitudinal (zonal) distributions, which can then be compared with zonal emission estimates resulting from inverse modelling.

Carbon dioxide

It was concluded for carbon dioxide that global carbon cycle models ignore short-term perturbations due to fluctuations in the climate system. It is therefore not possible to estimate yearly fossil fuel emissions derived from atmospheric measurements and global carbon dioxide budget calculations with more accuracy than the emissions based on *bottom-up* emission inventories. The main sources of uncertainty in establishing the CO_2 budget are the poor world coverage offered by the measuring stations, the yearly variations in sources and sinks, the still unsolved problem of magnitude of oceanic and biospheric sink leading to different assumptions on the budget by modellers. Yearly variations in the CO_2 budget are much larger than the yearly variations in the fossil fuel emissions. Therefore no accurate validation of national emission inventories is possible with budget studies. The uncertainty in the estimated CO_2 emission due to anthropogenic activities derived from atmospheric measurements is 25-35% and 10% or less in the *bottom-up* emission estimates. EDGAR (*bottom-up*) global CO_2 emissions due to use of fossil fuel are well within the range of *top-down* global estimates (difference < 13% between central estimates). Differences of CO_2 emissions due to land-use changes are larger (up to a factor of 3). Besides a global comparison of CO_2 emissions some additional spatial detail may be retained taking the indications of an additional biospheric sink in the Northern Hemisphere into account. The comparison of national inventories with EDGAR data has identified areas for future improvement in the IPCC Guidelines for CO_2, namely land use, agriculture, and biofuel combustion. Differences between national inventories and EDGAR data will be analysed further in the second phase of the project.

Methane

The short-term variations in the atmospheric increase of the methane concentration are in the order of magnitude of the yearly increase. The uncertainty of global methane emissions due to human activities derived from atmospheric measurements is in the order of 30%. The uncertainty in the sources estimated with *bottom-up* methods is about the same, some being higher and others lower (25-35%). Therefore a global or even a zonal comparison of *bottom-up* emission inventories with *top-down* results from transport models is possible and a reduction in uncertainty for specific sources may be achieved. EDGAR *bottom-up* estimates of global CH_4 emissions due to anthropogenic activities are well within the range of 35% from a central *top-down* estimate. A comparison of *top-down* with *bottom-up* emission inventories is possible on a much smaller - regional - scale for north-west Europe thanks to long-term high resolution measuring data being available for this area. Using data of one measuring station, Cabauw in the Netherlands, at a height of 200 m above sea level, it is possible to evaluate the national inventories of surrounding countries up to a distance of 500 km. Sectoral detail is less developed than in the inventories, but it is possible to evaluate the totals. The comparison of national inventories with EDGAR data has identified areas for possible future improvement of the IPCC Guidelines, especially the methodology for estimating methane from agriculture, biofuel combustion, land use, landfills and waste-water treatment.

Nitrous Oxide

Top-down inverse modelling exercises used to derive global and zonal budgets have been carried out. In addition, global 3-dimensional model calculations were used to simulate atmospheric concentration fields where gridded N_2O emissions served as input. Uncertainties

are high, both in the inventories and in the emission estimates based on results of global models. One of the reasons is that the stratospheric removal cannot be accurately quantified as yet. In addition, most of the atmospheric N_2O is of biogenic origin and from biogenic sources and shows especially in soils, large variability, both spatial and temporal. The least uncertain estimate is the trend in the atmospheric increase of nitrous oxide (4-5 Tg N/year). Using this figure and a central estimate for the magnitude of the stratospheric sink (12-13 Tg N/year) it could be shown by a *top-down* analysis that a large part (50%) of the global anthropogenic N_2O emission, derived from *bottom-up* emission estimates, was missing. Based on this information, a careful *bottom-up* analysis of all possible sources led to the identification of a new source of N_2O emissions: cattle and feedlots. This is a clear example of how a *top-down* estimate can improve *bottom-up* emission estimates. Nevertheless uncertainties are still large up to a factor of 2, so large in fact that we can expect both *top-down* and *bottom-up* emission estimates to benefit from results of a careful comparison of emission estimates. On the basis of present models and datasets we expect a comparison of national inventories with global model results to only be useful on a global or zonal scale. The global totals from the EDGAR database for N_2O emissions due to anthropogenic activities are much higher than the National Communications total (factor of 3), partly because a large number of countries did not submit a National Communication. However, totals are still much lower than the IPCC *bottom-up* total (factor of 2) based on the *Revised IPCC Guidelines* and *top-down* global estimates. A first analysis showed differences between EDGAR and National Communications to be large for most sources. The second phase of the study will further analyse these differences. For future improvement of the IPCC Guidelines it is recommended to focus on industrial processes, biomass burning, land use change and waste treatment.

Recommendations

The effectiveness and accuracy of the **Top-down** monitoring and modelling techniques can be increased through the following research activities:

1. Expansion of the network of concentration measuring sites, both background locations and source regions.
2. Measuring meteorological parameters and mixing-layer height simultaneous with concentration measurements.
3. Greater frequency of measurements at sites with seasonally varying source strengths.
4. Greater spatial coverage and accuracy of isotope measurements.
5. Establishment of international primary standards, and improved maintenance and calibration of standards, for individual laboratories and internationally.
6. Standardised collection and cataloguing of measurement data at international and regional centres.
7. Effective availability of measuring data through web sites and FTP sites is very important. The databases at CDIAC (Oak Ridge) and WMO-WCDCGG (Tokyo) are therefore important initiatives which should be supported.
8. Support the development of inverse modelling techniques such as Kalmanfiltering and the application of adjoint models.
9. Increase the temporal resolution of models by making them suitable for actual meteorological information.
10. Improvement of time profiles (seasonality) of emission sources in modelling.

The completeness and consistency of **Bottom-up** national inventories can be increased by the following activities:

1. Comparison of aggregated emission factors between national inventories or between different years in one inventory may identify deviating emission factors; likewise, comparison of sectoral emission strength indicators may indicate apparent typing or calculation errors.
2. Comparison of national inventories with global sectoral emission inventories constructed from international statistics and consistent sets of emission factors may reveal missing sources and identify areas with large deviating emission factors.
3. In-depth review of deviating emission factors should be done to check that they are country specific and not biased compared to similar countries.
4. Global sectoral inventories can be used to estimate the main sources in non-reporting countries as well as their share in the global total.
5. Compilation of global emission databases such as EDGAR and GEIA are important as scientific reference databases to perform quick checks on completeness and strength class of sources and for more detailed investigation of large deviations when identified in the comparisons mentioned above.
6. To be useful for comparison with national inventories, these global inventories should include emissions per country and distinguish key source sectors.
7. Maintenance and update, in particular of emission factors, in scientific databases are important for improvement of the consistency between countries.
8. Carry out measurements near sources to accurately determine specific emission factors

For **comparison of Top-down and Bottom-up** emission estimates the following aspects need to be considered to increase the accuracy of comparison results:

1. For an independent check of national inventories with *top-down* estimates from inverse modelling, often additional information is required on the temporal variation within a year (e.g. seasonality) and on the spatial distribution in a standardised way (e.g. on grid [1D]or latitudinal bands [1D]).
2. Inverse models require these 'fingerprints in space and time' of sectoral emissions as *a priori* input and also often generate their results in this spatial format.
3. For a proper evaluation by *top-down* models it is also important to have a fair description of the natural sources in the target area.
4. The uncertainty in the estimate of annual sectoral emissions of a country is important additional *a priori* information for inverse model calculations.
5. Facilities like the EDGAR system are required to consistently convert national emissions into 1D or 2D distributions as a bridge between national, annual inventories and atmospheric chemistry models.
6. In the future a Top-down and Bottom-up comparison of emission inventories should also include other greenhouse gases.

5.3 Second phase of the project

In the second phase of the project the analysis of differences between national inventories and EDGAR will be further elaborated. Further development of the top-down bottom-up analysis will be based on the conclusions of the IPCC expert meeting at 5 and 6 November 1997 at RIVM, the Netherlands..

Acknowledgements

This preliminary study by WIMEK was funded by the Ministry of Housing, Physical Planning and the Environment, Directorate of Air and Energy, Department of Climate Change (project number 9714003).

Constructing and maintaining of EDGAR is part of Dutch National Research Programme on Climate Change (project number 954222) and of GEIA, a component of the IGAC Core Project of IGBP, which is carried out by RIVM and TNO and also funded by the Dutch Ministry of the Environment (VROM), (project number 728001).

We thank Ing. J.A.H.W. Peters at RIVM for his assistance in extracting and processing of EDGAR data in the IPCC source formats and Mrs. R.E. de Wijs-Christensen for editorial advice on the Executive Summary.

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Appendix

Table A.1 Summary table sectors from IPCC

Source identification	Source
1A1	Energy and transformation industries
1A2	Industry
1A3	Transport
1A4	Commercial/Institutional
1A5	Residential
1A6	Agriculture/Forestry
1A7	Other (fuel combustion)
1A8	Biomass burned for energy
1AT	TOTAL Fuel combustion
1B1	Oil and natural gas systems
1B2	Coal mining
1BT	TOTAL Fugitive fuel emissions
2A0	Iron and Steel
2B0	Non-ferrous metals
2C0	Inorganic chemicals
2D0	Organic chemicals
2E0	Non-metallic mineral products
2F0	Other (industrial processes)
2TT	TOTAL Industrial processes
3A0	Paint application
3B0	Degreasing and dry cleaning
3C0	Chemical products manufacture/processing
3D0	Other (Solvent use)
3TT	TOTAL Solvent use
4A0	Enteric fermentation
4B0	Animal wastes
4C0	Rice cultivation
4D0	Agricultural soils
4E0	Agricultural waste burning
4F0	Savannah burning
4TT	TOTAL Agriculture
5A0	Forest clearing and on site burning cleared forests
5B0	Grassland conversion
5C0	Abandonment of managed lands
5D0	Managed forests
5TR	TOTAL Removals - Land use and forestry
5TT	TOTAL Land use and forestry
6A0	Landfills
6B0	Wastewater
6C0	Other (waste)
6TT	TOTAL Waste
7A0	International shipping (bunkers0
7B0	International air transport
7TT	TOTAL Bunkers
8TT	NATIONAL TOTAL