Climate Change Research: Evaluation and Policy Implications
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ASSESSMENT REPORT ON NRP SUBTHEME

"GREENHOUSE GASES"

SOURCES AND SINKS OF CO_2 CH_4 AND N_2O, DATABASES AND SOCIO-ECONOMIC CAUSES

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ABSTRACT

The aim of the subtheme Greenhouse gases of the Dutch National Research programme on (NRP) is to quantify the sources and sinks of the major greenhouse gases to enable estimates of the future atmospheric concentration. The major part of the projects in this theme is focused on the Dutch situation, but the results can be extrapolated countries or regions. The information gained will be used for Dutch policy decisions regarding abatement of greenhouse gases.

Section 1 deals with the aim and organization of Causes of climate change, and relates the scope to increased awareness of uncertainties in sources and sinks of greenhouse gases: at the start of the National Research Programme the general consensus of the scientific community was that these uncertainties were not extreme large, it is nowadays accepted that these uncertainties are larger than assumed before.

The aim the Cluster CO_2 **(Section 2)** was devoted to study the exchange between terrestrial ecosystems and the atmosphere to gain more knowledge of the "fertilization" flux. The research was mainly focused on the development of a CO_2 exchange model for grassland describing diurnal and seasonal fluxes, and on the validation of this local scale model on a regional and national scale.

In both the clusters CH_4 and N_2O (respectively **Section 3** and **Section 4**) anthropogenic and biogenic sources were studied. Major criteria to study sources were the source strength, but also the uncertainty in the source estimate and the potential emission reduction, all projected on the Dutch situation. Exception were the projects on CH_4 emission from rice fields, and the sea/air exchange of N_2O in oceans; expertise was available in The Netherlands to carry out these studies. As in the sub-theme CO_2 the study of processes in grasslands was given a high priority in the sub themes CH_4 and N_2O in order to quantify emission the mentioned greenhouse gases. Moreover, in the CH_4-sub theme projects were performed to evaluate and validate the strength of various sources.

The two remaining clusters (limited in extend) were aimed at the development of emission databases and geographic quantification of soil processes controlling greenhouse gas fluxes (cluster Database Development, **Section** 5), and on national inventories (cluster Socio-economic Causes, **Section 6)**. In the framework of the first cluster two databases were developed, one was the World Inventory of Soil Emission potentials (WISE), a global gridded database of the primary soil factors controlling soil greenhouse gas emissions, and the other was Emission Database for Global Atmospheric Research (EDGAR) aimed to describe the processes as land use, energy consumption etc, which control the emissions of greenhouse gases and other air pollutants. The goal of the other sub theme was to develop and apply methodologies to compile national inventories of greenhouse gas emissions in The Netherlands, focused on the compounds CH_4 and N_2O.

1. INTRODUCTION

1.1 Aim and organization

The scope of this theme was the assessment of the causes of Dutch National Research Programme Global Air Pollution and Climate Change (NRP). This obviously includes the cycle of the greenhouse gases but also the description of anthropogenic activities resulting in changes in atmospheric concentrations. The aim of the causes of climate change was to provide information needed to quantify the sources and sinks of the major greenhouse gases in order to enable more accurate estimates of future atmospheric concentrations. While most of the programmes within National Research Programme on were defined in a bottom-up process, it was decided to organise the research on cycles of greenhouse gases in a different way.

First, a first order analysis of the uncertainty in the cycles of CO_2 (carbon dioxide), CH_4 (methane), and N_2O (nitrous oxide) was made by a small group of Dutch scientists, active in this field. The conclusions of this analysis were that the main uncertainty in the cycle of CO_2 was caused by insufficient information on important sinks such as uptake by oceans and terrestrial ecosystems. More knowledge on the level of mechanistic descriptions was clearly needed, but the process of integrating locally derived information regarding important sinks of CO_2 up to the level of relevant descriptions of CO_2 exchange on regional, continental, or global scale also introduce large uncertainties. The same situation was observed regarding the emissions of methane. Also in this case local measurements are extrapolated to regional and global scale, introducing large errors. For both CO_2 and CH_4 the scaling problem, this is generalisation of local measurements to regional and (sub)continental scale, was seen as a serious problem. In the case of N_2O the state of knowledge was worse, information on the mechanisms of e.g. emission of N_2O during nitrification or denitrification was not really available.

Second, it decided that coherent programs would be formulated for each of these greenhouse gases, which should address the mentioned main problems. These programs should be formulated in such a way that on one hand typical Dutch aspects of the cycle of greenhouse gases would be emphasised and that on the other hand the information gained in the research would constitute a worthwhile contribution to international programs, primarily within the scope of IGAC .

Based on these considerations the following clusters of projects were developed:

- study of the CO_2 exchange between grasslands and the atmosphere, and development of methodology to validate CO_2 exchange models for larger areas;
- investigation of CH_4 emissions of selected sources, thought to contribute significantly and with a large margin of error, and development of methodology to validate CH_4 exchange models for larger areas;
- quantification of N_2O emissions from fossil sources and important biogenic systems, in particular from grasslands, sewage treatment systems and from freshwater and marine systems.

Table 1.1
List of projects in the NRP subtheme "Greenhouse gases" by clusters

Title	Project leader	Number
Carbon dioxide (CO_2) ecosystem studies, model development and validation		
The seasonal cycle of the CO_2 exchange between atmosphere and vegetated surfaces	J. Goudriaan	852062
Quantification of carbon fluxes in grassland	P.J. Kuikman	852063
A feasibility study for aircraft based flux measurements of CO_2	W. Ruijgrok	852065
The development of a geographically explicit dynamic carbon cycle model	R Leemans	852067
Quantification of carbon fluxes in Dutch forests (Part 1: Desk study)	G.M.J. Mohren	852071
Determining relative importance of sources and sinks of carbon dioxide using carbon isotope measurements	W.M. Kieskamp	852076
Measurement of the exchange of CO_2 between the atmosphere and a grassland	W. Ruijgrok	853116
Methane (CH_4) biogenic sources, fossil sources		
Quantification of methane emissions due to natural gas losses and petroleum production	R.J. Nielen	850008
The influence of soil parameters on the production and emission of methane in/by wet rice paddies	N. van Breemen	850009
Greenhouse gases from landfills in the Netherlands	C. Verschut	850023
Methane formation by anaerobic consortia in organic grassland soils	A.J.M. Stams	853120
Programming study for methane research	J.J.M. Berdowski	852068
Validation of source strengths of atmospheric methane using carbon isotope ratios.	W.M. Kieskamp	852097
Quantification of methane emissions in the exploration and production of natural gas and petroleum The Netherlands	J. Oonk	853104

Measurement study landfill gas production emission and recovery	J. Oonk	853105
Effects of grassland management on the emission of CH_4 from grassland on peat soils	O. Oenema	853121
The methane consumption by indigenous grassland micro flora	J.A.M. de Bont	853122
From methane formation and oxidation to methane fluxes in organic grassland soils: modelling	P.A. Leffelaar	853123
Evaluation and validation of the CH_4 emissions in the Netherlands and contributions from various sources	J.C.Th. Hollander	853124
Determination of emissions of methane in rural areas	P. Hofschreuder	853125
Nitrous Oxide (N_2O) biogenic sources, fossil sources		
N_2O emission from fossil fuel combustion in power plants	H. Spoelstra	850006
Preliminary study on N_2O flux measurements	H.S.M. Diederen	850012
Investigation of the contribution of traffic to N_2O emissions both now and in the future	J. Baas	850030
Effects of nitrogen fertilization and grazing on the N_2O emission from grassland.	O. Oenema	852073
Factors influencing the ratio N_2/N_2O as nitrate is removed from the soil by denitrification	P.A. Leffelaar	852074
The emission of N_2O from grassland	H.G. v. Faassen	852078
Modelling of soil emissions of nitrous oxide for global studies	A.F. Bouwman	852079
Measurement of atmospheric emissions of N_2O from biogenous surface sources in general and grassland-ecosystems in particular	J.H. Duyzer	852096
Database development emission database, geographic quantification of soil controlling gas fluxes		
Global emission database	J.J.M. Berdowski	850032
Geographic quantification of soil factors and soil processes that control fluxes of greenhouse gases (Currently used acronym: WISE, World Inventory of Soil factors and processes that control Emissions of greenhouse gases)	E.M. Bridges / N.H. Batjes	851039

Emission Database for Global Atmospheric Research (EDGAR); Phase 2: data collection and implementation	J.G.J. Olivier	851060
Socio-economic causes national inventory, policy analysis		
Social causes of the greenhouse effect and emissions inventories	R.J. Swart	850019

The information gained in these clustered projects should contribute to existing or future emission data bases. In the CO_2 cluster, a specific project was formulated to ensure that the results of this cluster was transferred to the EDGAR and IMAGE data base (See Annex 2 for acronyms). In the CH_4 and N_2O clusters this transfer was regulated in a less formal way, mainly because the estimate was at the time of the start of this work, that insufficient information would be available after 3 years to perform this task rigorously.
These clusters started fairly late in the NRP phase-I, so only some two and a halve years were available for these studies, in stead of five for most of the other NRP-I projects.

1.2 Assessment of the uncertainties in sources and sinks of greenhouse gases at the time of the start of NRP

At the time of the start of NRP-I (1989-1990) general consensus existed in the scientific community as expressed in the 1992 IPCC report, on the uncertainties in the predictions of climatic changes. It was widely accepted that it was possible to predict future concentrations of greenhouse gases, with a considerable margin of errors of course. An important problem was perceived in the translation of these changes in concentrations of the radiative active gases in changes in the radiative balance of the earth. But the main problem, as perceived in that period (as reflected in the discussions during the Chamrousse conference in 1989) were the effects of these shifts in the radiative budgets expressed in terms of possible climatic changes.

The Carbon cycle

That the above mentioned consensus existed regarding the Carbon cycle was remarkable, in view of the fact that already many observations were available which indicated that the uncertainty in sources and sinks of greenhouse gases including CO_2 were much larger than thus far assumed. This was made very clear during a number of scientific meetings, e.g. in the proceedings of the IUPAC workshop 'Assessment of uncertainties in the projected concentrations of carbon dioxide in the atmosphere' (Slanina et al., 1991). In these proceedings it is stated that the uncertainties in the most important sinks of CO_2 are very large. The estimates of uptake by the oceans vary from 1-2.5 Pg C per year, with a value of between 1-2 Pg as the most probable range. The increase of CO_2 in the atmosphere will induce enhanced growth of vegetation and part of the carbon, fixed in this manner, will be present in the form of enlarged root systems. A certain fraction of the extra carbon, present in roots, will remain in the soil after the decay of the vegetation and be stabilised for periods between 50 and 500 years. Estimates of the value of this so-called 'fertilization' flux range from 1 to 3 Pg C y^{-1}

(Table 1.2) (Goudriaan, 1989 and Tans et al., 1990). These uncertainties have a decisive influence on future environmental policy decisions (Slanina et al., 1991).

If a large fertilization effect exists, abatement measures are feasible. If uptake by the oceans and fertilization effect are in the range of the lower estimates, stabilisation of the atmospheric CO_2 concentration is very difficult. If we assume annual fluxes of 6 Pg C, 1.5 Pg C, 2 Pg C, and of 2.5 Pg C, for respectively fossil fuel, landuse changes, oceanic uptake, and terrestrial fertilization, the difference of 3 Pg C between sinks and sources accounts for the accumulation of CO_2 in the atmosphere. If we are able to stop the deforestation and induce increase of forests (corresponding to -0.2 Pg C y^{-1}), the difference between sinks and sources would be in the order of 1.3 Pg C, corresponding to approximately 25% reduction of the emissions by fossil fuels. Measures directed to stop deforestation and to optimise agricultural production are probably cheaper than reductions in the order of 60% or more of emissions by fossil fuel. The relatively low agricultural productivity per unit land area of the countries containing the large tropical forests (0.1 to 0.2 of the potential production per unit of land area, to be compared with 0.6 to 1.1 for European countries) leave room for such a policy. A combination of different measures, reduction of the use of fossil fuel, reforestation, and optimisation of agricultural activities could be effective in this case and will leave room for extension of emissions by the developing countries.In the case that no fertilization effect exists and the uptake by oceans is only moderate, a very different picture emerges. A reduction of at least 60% of the emissions of fossil fuel could be necessary to stabilise the present CO_2 concentrations in the atmosphere.
Any increase in the emissions of the third world countries would ask an even more stringent emission abatement in order to prevent a further increase of the atmospheric CO_2 concentration. In the worst case even the most stringent emission reductions in the industrial countries could be insufficient to counteract increasing emissions in the developing countries. One would be tempted to invest in adaptation strategies rather than in abatement of emissions if this latter situation proves to be true.

It is clear that the uncertainties in the sinks of CO_2 will enormously influence future political developments and that the reduction of these uncertainties is of prime importance.

The conclusions of the IUPAC workshop made very clear that predictions of future CO_2 concentrations had far larger uncertainties as was assumed until then, and that additional research on the role of terrestrial systems on the CO_2 budget was urgently needed.

Table 1.2
Sources and sinks of CO_2 (IPCC, 1990)

	Flux (Tg y^{-1})
Source	
Fossil fuel combustion	5.4 ± 0.5
Deforestation/landuse	1.6 ± 1.0
Sink	
Uptake by oceans	2.0 ± 0.8
"Terrestrial fertilization"	1.6 ± 1.4
Atmospheric increase	3.4 ± 0.2

Sources and sinks of CH_4

The same situation as for CO_2 existed, in essence, regarding the sources and sinks of CH_4. In Table 1.3 the estimates are given of the source strength for the most important methane emissions, as presented by IPCC in 1990 (IPCC, 1990). It was felt that these estimates were, of course with a degree of uncertainty, fairly well established. The fact that the emissions of methane were nearly equal to the sum of methane oxidized in photo-chemical reactions and the amount tied in with increasing atmospheric concentrations, was regarded as an objective proof for this assessment.

The conclusions of the IUPAC workshop on uncertainties of in the projected concentrations of methane in the atmosphere (Slanina et al., 1994) among other scientific meetings, were very different:

- the latest reports (Slanina et al., 1994) about the emissions of Northern Wetlands, indicated that these emissions could be substantially lower as formerly assumed. The emissions of Northern wetlands were extrapolated to be in the order of 20 Tg y^{-1} instead of 80 Tg y^{-1} ;
- an unexpected conclusion of the workshop was that the possibility exists that India, one of the main rice growing countries, contributes no more than 7% of global emissions from rice crops, because most Indian paddy crop is taken from irrigated field and only a small portion from water-logged fields (Slanina et al., 1994);
- the uncertainty in atmospheric oxidation is so large (in the order of 40% according the estimates of Calvert, 1994) that this mechanism cannot be used to check our emission inventories;
- the emissions of landfills could be much larger as previously assumed, based on the first results of measurements in Canada, China and other countries (Slanina et al., 1994).

In these proceedings it is very clearly concluded that the extrapolations of very local measurements to emission fluxes on regional, continental and global scale is probably one of the main sources of errors and uncertainties. The specific recommendations is made to develop methods and measurement strategies for specific validation measurements to evaluate emissions on regional and

(sub)continental scale. These validation measurements are required to check whether the measurements of methane emissions carried out on very small scale, have been extrapolated correctly in the past to global dimensions.

This assessment of the sources and sinks of methane makes very clear that the state of knowledge at that time did not provide a suitable scientific fundament for predictions of future development and hence for abatement policies.

This assessment already mentions that the trends are changing (Khalil et al., 1994). For an extended period a exponential growth, in the order of about 1% per year has been observed. This trend has abruptly changed and is much less than formerly observed (Steele et al., 1992). Rigorous explanations were not offered, a clear proof of the lack of knowledge in this area.

Table 1.3
Sources and sinks of CH_4 (IPCC, 1990)

	Flux (Tg y^{-1})	Range (Tg y^{-1})
Source		
Natural Wetlands	115	(100-200)
Rice paddies	110	(25-170)
Enteric fermentation	80	(65-100)
Gas drilling, venting, transmission	45	(25-50)
Biomass burning	40	(20-80)
Termites	40	(10-100)
Landfills	40	(20-70)
Coal mining	35	(19-50)
Ocean	10	(5-20)
Freshwaters	5	(1-25)
CH_4 hydrates destablilization	5	(0-100)
Sink		
Removal by soils	30	(14-45)
Reaction with OH in the atmosphere	500	(400-600)
Atmospheric increase	44	(40-48)

Sources and sinks of N_2O

The assessment of the sources of nitrous oxide was at the time different from the other gases. It was assumed that all estimates of sources were quite uncertain. Tropical forest soils were regarded as the single most important source of nitrous oxide to the atmosphere. N_2O is also emitted by a large number of smaller sources, such as biomass burning, agricultural activities leading to nitrification and denitrification processes and specialised industrial processes (Table 1.4).
The conclusion was that most of these sources were very difficult to evaluate. As a consequence the uncertainties in the emission estimates were estimated to be large. In Europe, the production of N_2O by agricultural systems with high loads of

nitrogen, the emissions of electricity generation plants and the exhausts of cars equipped with catalysts were seen as major sources. The high estimates of emissions of electricity generation plants were caused by artifacts in sampling and analysis of flue gases, as was proven in the first stages of NRP (Spoelstra, 1992).

Table 1.4
Sources and sinks of N_2O (IPCC, 1990)

	Flux (Tg y^{-1})
Source	
Ocean, estuaries	1.4 - 2.6
Fertilizer (including ground water)	0.01 - 2.2
Soils (tropical forest)	2.2 - 3.7
(temperate forest)	0.7 - 1.5
Combustion	0.1 - 0.3
Biomass burning	0.02 - 0.2
Sink	
Removal by soil	?
Stratospheric loss	7 - 13
Atmospheric increase	3 - 4.5

1.3 Assessment of the developments in the knowledge on sources and sinks of greenhouses gases during the course of NRP phase I

During the last 5 years, the period of NRP phase I, the scientific community has accepted that the uncertainties in sources and sinks of greenhouse gases are much larger as assumed in 1989. The warnings, exemplified by the proceedings of the IUPAC workshops on the uncertainties of in the projected concentrations of carbon dioxide and methane in the atmosphere (Slanina et al., 1991 and 1994) that these uncertainties are a major in future predictions, have now widely be accepted.

That a large uncertainties exist has been made clear by the fact that the trends in the concentration of greenhouse gases in the atmosphere have changed drastically and that no reasonable explanation can be provided for these changes in the trends. So the present state of affairs can be summed up as follows:

- The fair amount of research on sources and sinks of greenhouse gases, carried out internationally during this period of 5 years, has led to the conclusion that the uncertainty in these sources and sinks is much larger than formerly assumed. This may seem a rather negative conclusion, but it must be born in mind that a proper evaluation of the state of affairs is essential to formulate effective research in the future to remedy this problem.
- The long-term average growth rate of atmospheric CO_2 concentration has increased since the start of the measurements at Mauna Loa. This rate was about 0.8 ppmv, 1.3 ppmv, and 1.6 ppmv for respectively the 1960s, the 1970s, and the 1980s. Systematic higher CO_2 concentration growth rates have been observed during the years 1988-90, which exceeded the level of 2.0

ppmv y^{-1}, while in the subsequent years (1991, 1992, 1993) very low growth rates have been observed, in the order of 0.6 ppmv y^{-1}. Indications exist, based on the most recent data, that the trend is returning towards long-term growth rates. It must be kept in mind that the abrupt decrease in atmospheric CO_2 growth rate in the period 1991-1993 exceeds any previous variation in the existing time series of atmospheric CO_2 concentration.

CO_2

The research in this area has expanded, leading to an increase in knowledge of the oceanic and terrestrial sinks of the carbon dioxide. New insights have been obtained on the problem of "unidentified" terrestrial sink (indicated by Tans et al., 1990 and the IUPAC report Slanina et al., 1994, as "fertilization effect") to specific processes. The uptake of CO_2 by terrestrial systems is governed, most likely by two important processes:

1) Changes in land use. The present estimate is that the net emissions by changes in land use total 1.1 Pg with an uncertainty of 1.2 Pg. This net emission flux consists of the sum of emissions by tropical sources (1.6 Pg) minus a mid-latitude uptake due to forestation, etc. of about 0.5 Pg, according to recent estimates.
2) The existence of the CO_2 fertilization is increasingly accepted as an important sink. But there is increasing evidence that different interactions play a role. The direct CO_2-enhanced plant growth could provide a sink with a strength of 0.5-2.0 Pg C y^{-1}. Enhanced supply of nutrients, e.g. by transport and deposition of sulphur and nitrogen compounds leads to the so-called nitrogen fertilization, which could contribute to an uptake of 0.2-1.0 Pg C y^{-1}. As a result, the future effects of CO_2 are difficult to predict. Climatic change could have, on a global scale lead to a net uptake, equivalent to 0-1.0 Pg C y^{-1}.

The resulting picture is that indeed the knowledge of these processes has been increased. In 1989 sinks such as the "fertilization effect" were still hotly debated. But the uncertainty is still very large and remains in boundaries as indicated by the IUPAC report. This is the most important reason, why no consistent explanations can be offered for the variations in the yearly trends, observed in the last decade. The debate between scientists, who contribute most of the terrestrial sink to changes in land use, and those who claim that fertilization effects are the main cause, is still raging, see articles of Tans et al., (1990) and Smith et al., (1992). It is very clear that no reliable predictions of future CO_2 concentrations can be made and that the development of optimal strategies for abatement is severely hindered, until these questions are resolved.

CH_4

Essentially the same situation exists for CH_4 as described for CO_2. The changes in the trends for CH_4 have in fact been more pronounced as observed for CO_2. Results from different networks indicate that the globally averaged growth rates for methane have declined from approximately 20 ppbv y^{-1} in the period 1979-1980 to 13 ppbv y^{-1} in 1983, to 10 ppbv y^{-1} in 1990 and to about 5 ppbv y^{-1} in 1992 (Steele et al., 1992 and Khalil et al., 1993d en 1993e). The trend in southern hemisphere has halved and the increase in 1991-1992 in the northern hemisphere was close to zero (Dlugokencky et al., 1992). The cause of this change

in methane growth rates is unknown and still a matter of speculation. (Khalil et al., 1992c and Steele et al., 1992), (Dlugokencky et al., 1992 and (Dlugokencky et al.) A wide range of explanations are given:

- decreasing CH_4 emissions from the former Soviet Union;
- lowering of lowered the temperature of the northern wetlands and thereby decreasing methane emissions; indirectly caused by emission of the Pinatubo Volcano;
- lowering of the water table increases the thickness of the layer over which methane oxidation can take place, so northern wetlands appear to be more sensitive to changes in moisture than temperature;
- termination of the one-to-one correlation between methane emissions and growth of the global population, as result of lack of suitable areas for rice cultivation or cattle raising;
- increasing OH-radical concentrations, caused by increasing UV-B radiation, could have shortened the life time of CH_4.

This wide range of hypotheses demonstrates quite clearly the lack of information on the strength and the variability of sources of methane and this situation is acknowledged widely within the scientific community.

N_2O

The development in views about the emissions of N_2O have been slightly different compared to the other two gases mentioned. Uncertainty exists about the atmospheric concentration of N_2O in the pre-industrial period. Estimates range from 260 to about 290 ppbv, compared with a concentration of 310 ppbv in 1993. So, the estimates of the yearly trend show a considerable uncertainty (Khalil et al., 1992C) Recent results indicate that the trend of N_2O has been smaller in the last years than the average of the last two decades and decreased from about 0.8 ppbv to 0.6 ppbv.

The general opinion in the scientific community is that N_2O plays only a minor role in the changes of the radiative balance of the earth and that the increase in concentration has been much less spectacular compared to the other greenhouse gases.

Questions are raised however on the impact of new industrial processes (large scale introduction of catalytic devices for cars and catalytic NO_x reduction in industry), of changes in agricultural practice (large scale application of nitrogen fertilizers) or changes in the water tables of wetlands and agricultural areas on future N_2O emissions.

The developments of the last five years can be summed up in the following statements:

- the considerable amount of new information has made clear that the uncertainties in sources and sinks of greenhouse gases were much larger than assumed in the recent past;
- major causes for the uncertainties in sinks and sources of greenhouse gases have been identified. This increased knowledge will provide the necessary fundament for effective research in the future.

This summary of the recent results of could give the impression that less risk for climatic change is present, compared with a few years ago, as the concentrations of greenhouse gases are increasing slower as expected. This would be a very dangerous assumption. As the changes to lower trends in the atmospheric concentrations of greenhouse gases are not well understood it is impossible to indicate whether this situation will last. To the contrary, it is very well possible that the trends could change in upward direction very fast by the impact of changes in industrial and agricultural practice.

A certain lack of knowledge regarding the contribution of different sources of greenhouse gases is not a problem in the first stages of abatement policies. A wide range of so-called no regret options, which will not only reduce the emission of greenhouse gases but also contribute to abatement of other environmental problems, is available and environmental policies can been adapted accordingly. This state of knowledge, however, is not a good basis for developing abatement policies over longer periods. For this reason a better understanding of the sources and sinks of greenhouse gases must have a high priority on the scientific agenda.

2. CARBON DIOXIDE (CO_2)

2.1 Overview of the CO_2 cluster

The considerations in chapter 1 led to the decision that additional research on sources and sinks of CO_2 should be directed to the role of terrestrial ecosystems in the CO_2 cycle, apart from the already on-going activities on the exchange of CO_2 between the oceans and the atmosphere.

Reports on the impact of the "fertilization flux" that were published at that time, had made clear that the exchange between terrestrial ecosystems and the atmosphere was very important and must be known better in order to be able to model the CO_2 cycle and to predict future CO_2 concentrations in the atmosphere. It was proposed to study the exchange of CO_2 between grasslands and the atmosphere for the following reasons:

- The Netherlands are to a large extend covered by grasslands. Pastures are a major component in European land use and the amount of grassland has been considerably extended on a global scale during recent decades;
- grasslands exhibit the same behaviour as forests as far as the fertilization effect is concerned: net primary production increases, allocation to roots as well as losses into soil, and potentially more C will be stored as organic matter with large residence time. Farmlands are amongst the most productive ecosystems (in terms of net photosynthesis). The soils of grasslands contain generally large amounts of carbon, and the carbon content of soils increases at higher concentration of CO_2 in the atmosphere.
- less knowledge was available on the exchange of CO_2 between grasslands and the atmosphere compared to forest ecosystems;
- expertise was available in The Netherlands.

It was decided to develop a coherent program, dedicated to formulate and validate a improved model simulating the exchange of CO_2 between grassland and the

atmosphere. This model encompass diurnal to seasonal fluxes, and the exchange of carbon between the soils of grasslands and the atmosphere. Better knowledge on the gross exchange of carbon between grasslands and the atmosphere is urgently needed to:

- assess the effects of changes in land use on the global carbon cycle;
- understand short time to yearly trends of CO_2 concentrations in the atmosphere. (Analysis of these trends are essential tools to understand the CO_2 cycle);
- assess the potential contribution of fertilization effect to sequestering of carbon in soils.

In order to reach this goal the following activities were incorporated in a number of projects:

1) Development of an improved model describing the exchange of CO_2 between grassland and the atmosphere (LUW-TPE, project no. 852062). This model will provide a good description of diurnal and seasonal fluxes, and will include the exchange of carbon between the soils of grasslands and the atmosphere.

2) Measurements of the exchange flux of CO_2 over the most important types of grasslands (soil) in The Netherlands (KEMA, project no. 853116; ECN, project no. 852076). It was conceived that the different kinds of soil of grasslands (clay and peat) would have a strong effect on these exchange fluxes. The results of these measurements would be used to parameterize better models and to validate them on a local scale (ECN, project no. 852076).

3) Investigation of the fertilization effect on grassland by means of pulse-labelling by $^{14}CO_2$ (IB-DLO -at present AB-DLO-, project no. 852063). Grass is exposed to $^{14}CO_2$ during short periods and the distribution of ^{14}C between different parts of the vegetation and the soil is determined and fluxes of carbon are calculated.

4) Measurements of the exchange flux of CO_2 over larger areas of grassland, using eddy-correlation, gradient measurements at higher elevation (ECN, project no. 852076), and eddy-correlation measurements from aircraft (KEMA, project no. 852065).

5) Measurement of changes in CO_2 concentration and isotopic composition ($^{13}C/^{12}C$ and $^{14}C/^{12}C$) at an altitude of 200 meters on a tower, with the objective to obtain regional validation of sources and sinks of CO_2 with emphasis on the role of terrestrial systems (ECN, project no. 8520786). The isotope ratios are dependent on sources and exchange processes.

An co-ordination group monitored the progress of the different projects and facilitated that coherent results could be obtained.

The sub-theme CO_2 encompasses two additional projects. One was a desk study to assess the role of (Dutch) forests as apart of the carbon cycle (IBN-DLO, project no. 852071), while the other project was on the development of a geographically

explicit dynamic carbon cycle model that will be incorporated in more complex, integrated models as IMAGE 2.0 (RIVM, project no. 852067).

2.2 Methodology

Measuring CO_2 concentrations with sufficient precision and accuracy does not present severe problems in the present state of methodology development. Several monitors, based on IR absorption methods, are commercially available. They can measure atmospheric concentration with an accuracy of better than 1 ppmv. Accuracy is dependent on the quality of calibration and quality assurance standards, but generally an accuracy of 1 ppmv or better is attainable without major problems.
The situation is different when flux measurements must be applied in the field to study in detail the exchange fluxes of CO_2 between grasslands and the atmosphere. Three available methods to measure gas exchange between the atmosphere and biosphere have been used in the described projects, i) enclosure, ii) eddy-correlation, and iii) gradient.

Enclosure methods

A box is placed over vegetation, water, or soil. Air is pumped through the enclosure and the difference in concentrations measured at inlet and outlet is used to assess deposition or emission rates. Enclosure methods suffer from two problems: one, the enclosure can alter the behaviour of vegetation or soil, and two, the deposition or emission measurement is extremely local. The advantage of enclosure measurements is that the present state of instrumentation can be applied in nearly all studies. In many cases enclosure methods have to be used as no other alternative is available. In view of the extreme local effects and as alternatives are available for CO_2, it was decided to not to apply enclosure techniques for the standard CO_2 exchange measurements. Box measurements are applied in this cluster to investigate the exchange of CO_2 between atmosphere and grass with exclusion of soil respiration. In order to quantify the CO_2 flux by grass (in contrast to the integrated CO_2 flux: grass + soil) an enclosure system was developed which measures continuously the CO_2 flux under conditions of overpressure. This overpressure prohibits the exchange of CO_2 between the soil and the atmosphere. The measured CO_2 flux is, under these conditions, related to net CO_2 assimilation of a grass canopy under field conditions - one of the two components of the integrated CO_2 flux. Moisture content, temperature and CO_2 concentration of the circulating air are regulated to avoid the already mentioned artifacts caused by deviation in the box from local conditions.

Eddy-correlation

Eddy-correlation measurements are based on the covariance of fluctuations in ambient concentrations and vertical windspeed. Turbulent transport in the atmosphere takes place by eddies. In eddy-correlation measurements the difference in concentration of the investigated compound is measured with a time resolution of 1 to 10 hertz in air moving downward to the surface and moving upward from the surface. The upward moving air has been in contact with the surface and the concentration has altered due to exchange at the surface. As eddy correlation measures directly, it is very often the preferred method. The regarding speed and precision of the instrumentation for eddy correlation are very often so

extreme that the method cannot be used for many trace gases. However, the method can be applied for CO_2, and was used in the CO_2 cluster.

Gradient measurements
Depletion or emissions of pollutants at the surface results in a gradient in concentration. Air concentrations of compounds, temperature, and windspeed are measured at different heights over the surface. From these gradients the turbulence of the atmosphere is derived and the fluxes can be calculated. The problem with the gradient method is that a high precision is required of the measurement method, as the concentration gradients are often in the range of a few percent of the atmospheric concentration. The instrumentation for CO_2 measurements can fulfil these requirements and gradient measurements have been applied in two projects in the CO_2 cluster to measure fluxes.

Micrometeorological methods (eddy-correlation and gradient measurement) enables to estimate fluxes over a certain area as function of the height of the measurements. If local exchanges are studied, gradient measurements are carried out at heights between 1 and 5 m. The integrated exchange flux over an area of some hundreds of square meters is characterized this way. To study the exchange over an area of a few hectares, the gradient is measured between 1 and 20 m.
The precision of flux measurements are limited, typically a precision in the order of 20 % can be reached in most cases. The consequence is that direct measurements of the fertilization flux is not possible. This can be illustrated easily if the total exchange flux of terrestrial ecosystems with the atmosphere, in the order of 100 Pg C y^{-1} is compared with a high estimate of the fertilization flux of 2 Pg C.

The quality of flux measurements was validated in an intercomparison experiment, organized by ECN. The participants of the CO_2 cluster ECN and KEMA, and also KNMI and TNO, the latter two institutes are engaged in flux measurements at sea, took part in this one-week experiment in November 1993 at Cabauw, The Netherlands. Two methods were used: the eddy-correlation technique and the gradient technique. Unfortunately, the temperature was about 0 °C and reduced the flux of CO_2 considerably. Although the small magnitude of the fluxes makes comparison difficult, it seems that the gradient method tends to result in larger fluxes compared to the eddy-correlation method. Moreover, this experiment made clear that there was a considerable differences between the calibration standards that were used. This implies that direct comparison of absolute concentration values of different set-ups as at the Cabauw experiment can only be achieved by inter-calibration of the standards.

Validation of exchange fluxes over larger regional areas (The Netherlands and surroundings) were investigated by two different methods, a dynamic method (aircraft measurements) and a static method (tower measurements).
Eddy-correlation measurements in aircraft were applied as a method to obtain integrated exchange fluxes over larger areas. Variations in CO_2 concentration and isotopic composition were measured at an tower (200 m) at Cabauw, The Netherlands. Uptake and emission of CO_2 over large areas changes not only the CO_2 concentrations, but also the isotopic composition. Emissions of fossil fuel contains no ^{14}C and the ^{13}C-^{12}C ratio is dependent on the sources of CO_2. The

concentrations were measured with a non dispersive infrared spectrometer with an accuracy better than 0.1%. Working standards were calibrated against so called NOAA station standards. An wet annular rotating denuder, filled with a NaOH solution, was used to extract CO_2 from the air quantitatively. In the laboratory the formed carbonate was isolated (using barium chloride), stored, and before analysis re-converted into CO_2. The $^{13}C/^{12}C$ ratio and the $^{14}C/^{12}C$ ratio were determined at ECN and the University of Utrecht respectively. Meteorological data were provided by the Royal Netherlands Meteorological Institute (KNMI). An 2-dimensional 2-compartment mesoscale transport model was developed at ECN. Data on the spatial distribution as well as descriptions of CO_2 exchange (both biogenic and anthropogenic) will be used as input-parameters to model the observed CO_2 concentration and carbon isotopes.

To quantify the potential fertilization effect directly, gross annual carbon flows were estimated in grasslands with ^{14}C pulse labelling. $^{14}CO_2$ was supplied to grass plants as a single pulse (1-2 hours in a plastic bag covering the plants growing within a soil column) and subsequently the distribution within the plant and soil compartments was measured after a 21-days period in which carbon allocation was completed. This labelling was repeated on 13 representative moments during the growing season. Moreover, the decomposition of shoots and roots and the remaining carbon in soil organic matter was estimated by adding uniformly labelled dead shoots to planted soils in the field and by leaving pulse-labelled plants in the field and measuring the dynamics of the remaining carbon over 18 months following the addition and labelling of shoots and roots, respectively. The fate of carbon compounds that are exuded from living roots within 21 days after being assimilated was followed by adding 'model-rhizodeposits' and measuring the remaining carbon.

2.3 Results

Overview of the results

As the cluster of projects started late (some were stated at the end of 1992, others in the middle of 1993) only initial results are available. The most important results are summarized below:

1) A detailed model is developed to describe the exchange of CO_2. This new model provides information on exchange fluxes with a resolution of 30 minutes (Figure 2.1), and thus be used for mechanistic studies. The process to incorporate this model in IMAGE has been started.

2) Exchange fluxes with a time resolution of 1 hour or better have been measured over meadows on clay, and peat soils. The results have been transferred to the modellers. The flux measurements are of good quality and provide a good basis for the parameterization.

3) Gradient measurement of exchange fluxes on a scale of hectares have been carried out near Cabauw by measuring concentrations at altitudes between 1 and 10 m over clay/peat soil. A first comparison between model results and actual measurements is given in Figure 2.1. The difference between observed and calculated values are most probably caused by oxidation of peat in the soil

regulated by the water table. The combination of better models and good flux measurements enables the study of these important phenomena.

4) Eddy-correlation measurements of CO_2 exchange using an aircraft have been tested. The resolution of the method is insufficient to be useful under the prevailing conditions in The Netherlands.

5) The experiments with labelled $^{14}CO_2$ have been carried out and the distribution of carbon has been measured. The interpretation of the results is in progress.

6) Regional validation of models, describing the exchange of CO_2 between vegetation and atmosphere, by monitoring variations in CO_2 concentrations and isotopic composition at an altitude of 200 m is not possible yet. Distribution of sources and probably very large homogeneous processes like uptake in oceans can be studied by this method, but a more detailed assessment will be difficult in view of the limitation in the present models.

Detailed results

1) A dynamic simulation model was developed to calculate the CO_2 flux related to net CO_2 assimilation of a grass canopy. The existing carbon cycle model WCCM2 (Goudriaan, 1989) operates with annual time steps and does not consider the precise seasonal and diurnal pattern of CO_2 exchange. This study has incorporated these cycles while retaining the final result of the net annual exchange rate. To this end an existing simulation model for crop growth (SUCROS) has been utilized as a basis, in combination with other models for carbon dynamics in the soil (CENTURY). The model first generates the diurnal cycle to obtain the net diurnal assimilation rate, and diurnal soil respiration. These diurnal rates follow a seasonal cycle and are integrated to generate a net annual uptake. The net annual uptake of the above ground vegetation is called the Net Primary Productivity. Factors such as green soil cover, progress in the growing season on basis of accumulated temperature, soil wetness, partitioning of assimilates between plant organs, root dynamic are considered. Respiration rate of plant and soil have been modelled on basis of temperature, biomass and growth rate. The model has the potential to drive a 3-D model for atmospheric CO_2 content, first to generate a diurnal cycle in the vertical profile, second to obtain net CO_2 exchange rates of a region on a seasonal basis.

   A first comparison of the model calculations (grass component) with the integrated CO_2 flux measurements (grass and soil organic matter components) at Cabauw (The Netherlands), in combination with environmental conditions is presented in Figure 2.1, in which the measured and calculated (grass component - potential) CO_2 flux for Cabauw is given (Period March 18 to 20, 1993). LAI2 and LAI4 represent different leaf area index used in the model. The results indicate larger emission fluxes for CO_2 as calculated by the model. The difference between calculated and measured CO_2 fluxes may well be attributed to oxidation of soil organic matter, as discussed in the section dealing with the integrated flux measurements performed in

Cabauw. The combination of modelling activities and experimental approach provides a very useful tool to develop mechanistic descriptions of exchange processes between soil and vegetation and the atmosphere.
The development of a model for the calculation of the CO_2 flux related to oxidation of soil organic matter was initiated.

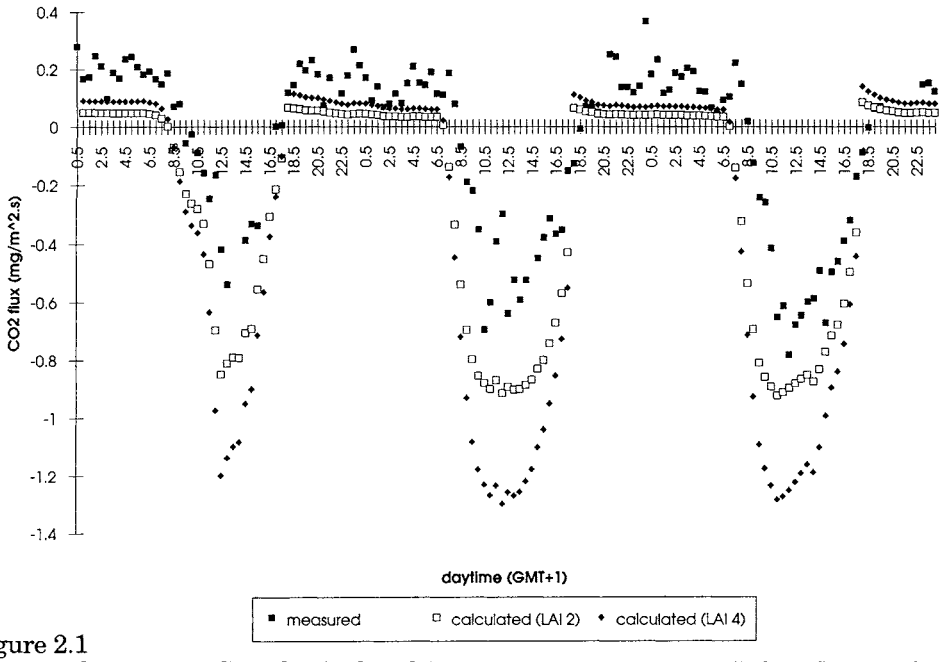

Figure 2.1
Measured (integrated) and calculated (grasscomponent-potential) CO_2 flux on the experimental site Cabauw (The Netherlands) form March 18 to March 20, 1993

2) The project on the development of a geographically explicit dynamic carbon cycle model has been fully integrated within the IMAGE 2 projects and data, experience and personnel and results were exchanged and model developments were immediately implemented in the IMAGE 2 framework.

A geographic explicit C cycle model has been developed based upon a series of global databases with topography, soil, climate and land cover characteristics. The model identifies globally different land cover types, each of which is divided into its appropriate compartments for C storage and dynamics. The model is driven by Net Primary Production, that is a function of local climate, soil and land use. Several feedback processes are implemented in a mechanistic manner. Sensitivity analysis was carried out and some specific applications to analyze the importance of different feedback processes and the influence of a transient dynamics vegetation response. Large regional differences were obtained for different feedback processes. For example, CO_2 fertilization was the dominant feedback in tropical regions, while the temperature response on growth and respiration became dominant in boreal regions. Although changes through feedbacks processes are important determinant of C cycle properties,

changes in land use will probably more dominant in the near future. The model is appropriate to also assess the impact of land use change on the C cycle.

Components of the project have now been reviewed twice during the international IMAGE review meetings and adjustments in the approach have been added after recommendations of the review committee.
Together with the Potsdam Institute for Climate Impact Research an improved version of the IIASA Climate database has been developed. This database (CLIMATE: Cramer, Leemans Interpolated Meteorology for Applications in Terrestrial Ecology) now forms the basis for several global modelling efforts (e.g. IGBP-GAIM).
The structure of the modelling approach and its main databases used within IMAGE 2.0 Terrestrial Environment Subsystem is accepted by IGBP-GCTE as a valid contribution to their core-project research.

3) Local scale exchange fluxes with a time resolution of 1 hour or better have been measured over grasslands on clay and peat soils using both the eddy correlation method and the gradient method. Eddy-correlation was performed at Zegveld (grassland over peat soil), gradient measurements at Lelystad (grass on clay) and at Cabauw. The latter measurements were carried out between 1 and 10 m, so integrated fluxes were measured over several km^2 of surface. The soil at Cabauw (peat covered with a layer of typically 20 cm of clay) is not as well characterized as is the case in Zegveld and Lelystad. The sites at Zegveld and Lelystad are part of experimental farms, so soil characteristics and all agricultural treatments are very well documented. In contrast, the meadows around Cabauw are commercially farmed, so little information is available about any application of fertilizer, grazing, pasture, etc. The basic idea was to use the data from Zegveld and Lelystad to parameterize the models and carry out validation by means of the flux measurements at Cabauw over an integrated area. As all the infrastructure at Cabauw was already in condition, it was possible to evaluate the measurements of fluxes, convert the results in a format which is suitable for the modellers and transfer them in a very short period. A nearly complete data set of fluxes for Cabauw with a time resolution of 30 minutes for the period March 1993 to March 1994 has been transferred to the modellers and is used for parametrisation and validation (Figure 2.2). Examination of the results indicate that the fluxes are measured with, for this kind of measurements, a very good precision, in the order of 10% relative.

Evaluation and validation of the raw data at Zegveld and Lelystad has not been completed yet, as time was needed to establish the necessary instrumentation, to carry out the actual measurements, and to perform intercomparison measurements in order to compare the results form both locations etc. Results, as far as available, have been transferred to the modellers. The evaluation of the total data set of both sites is nearly completed, so they will be transferred to the modellers in the near future.
The flux measurements performed at the Cabauw site are of such a quality that they indeed provide a good basis for the intended parameterisation and validation. The comparisons of models and experimental results, as reported later in this section, is therefor based on the results of Cabauw only.

Normally flux exchange measurements are carried out during relatively short periods. The only measurements performed over longer periods were measurements of dry deposition of NH_3 and SO_2, carried out by ECN and RIVM over forests and grassland. These measurements were based on gradient methods. The experience at Zegveld, where eddy-correlation was applied, has led to the conclusion that gradient methods are much more suitable for measuring over long periods than eddy-correlation based methodology.

4) CO_2 fluxes on km^2 scale have been estimated by measuring concentration gradients up till 10 m altitude at Cabauw, The Netherlands. The results are available to validate the exchange models. The measurement of the exchange of CO_2 between grass-on-peat and the atmosphere showed that fluxes caused by photosynthesis as well as soil respiration due to oxidation of peat can be assessed (Figure 2.2).

A net uptake only takes place in March, April and May, while during all other months a net emission occurred. The total net emission for the period March 1993 to March 1994 was calculated to be about 3000 kg C ha^{-1}.

Two sources contributing to the soil respiration are oxidation of peat layers and animal waste. The CO_2 emission due to animal waste was estimated at approximately 600 kg C ha^{-1} y^{-1} (about 20%) of the net estimated emission. This leaves 2400 kg C ha^{-1} y^{-1} for the oxidation of soil organic matter (e.g. peat) and is of the same order as the potential emission estimated for the oxidation of "shallow-drained" peat soil (about 2300 kg C ha^{-1} y^{-1}, Wolff). This is approximately 2.5% and 4% of the total anthropogenic CO_2 emission in the Netherlands respectively with and without emission from animal waste. Because the peat layer at Cabauw is covered with clay, it is expected that emissions for grass-on-peat will be larger due to the larger amount of peat that can be oxidized.
This type of measurement, in combination with models describing the exchange of CO_2 between the atmosphere and grass and soils, is in principle capable to provide the necessary information on uptake or loss of carbon as a function of changes in land use.

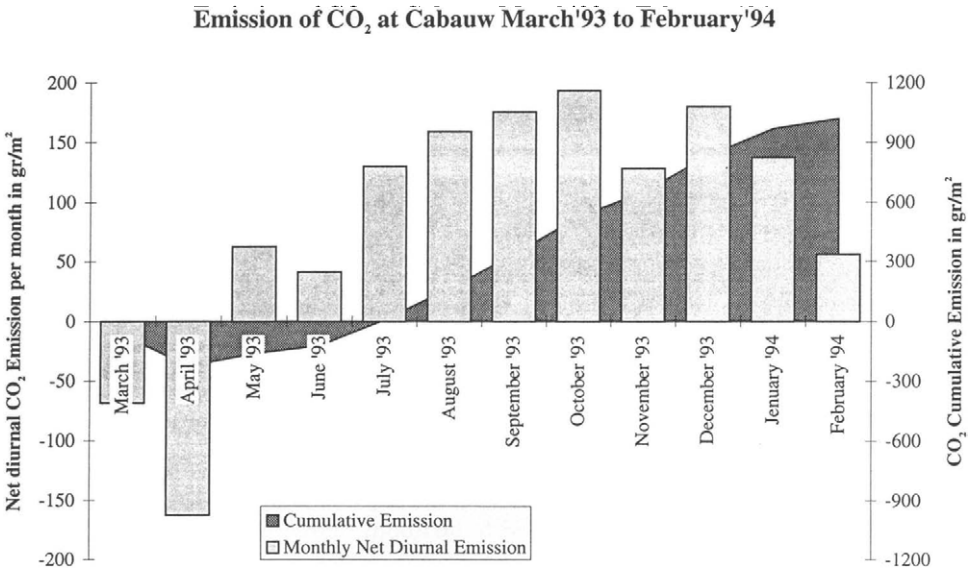

Figure 2.2
Emissions of CO_2 at Cabauw (The Netherlands) from March 1993 to February 1994

5) The instrumentation has been developed and tested to measure CO_2 exchange fluxes by eddy correlation measurements using aircraft. Airborne flux measurements provide information on fluxes over very large areas, depending on height and on large scale geographical distribution of these fluxes. Two measuring flights were performed to test an airborne eddy correlation system developed in the Netherlands. The results of these tests show that the accuracy of the system is insufficient to be useful to support model development under the prevailing conditions in the Netherlands. Fluxes are measured with a precision of 30 to 50% relative and parametrisation and validation of models require data of a better quality. The generally very inhomogeneous landscape of The Netherlands is a severe handicap for aircraft measurements, even if they are carried out at a minimum altitude to reduce the surface area which is observed. It was concluded that the airborne flux measurements were not feasible for application in the Netherlands taking a number of considerations into account: inaccuracy of the measurements, the heterogeneous character of the Dutch landscape and the relatively high costs of this type of measurements.

6) The experiments with labelled $^{14}CO_2$ have been carried out and the distribution of carbon has been measured. The interpretation of the results is in progress. Preliminary conclusion is that in peat soils decomposition (mg C added per mg soil C) is twice as fast as in sand and clay, and results in 5-10 times higher incorporation of carbon in biomass and microbial products. Structural organic carbon was retained more in clay (80%) than in sand (58%) whereas the extract in clay was decomposed as fast as in sand. The latter is

surprising given the generally higher retention capacity of clay soils than of sandy soils. Care should be taken since these data concern similar additions to all soils whereas the carbon input (total C, structural and soluble rhizodeposits) to soil might differ between the soils studied.

7) Regional validation of models, describing the exchange of CO_2 between vegetation and atmosphere, by monitoring variations in CO_2 concentrations and isotopic composition at an altitude of 200 m has not been possible yet, although a mesoscale transport model was developed. Also "mesoscale" measurement of CO_2 concentration were performed and carbon isotopes ratios were determined. Main problem in this project was the availability of detailed databases to be used as input for the model. Also good model descriptions of CO_2 exchange were lacking. Both problems will be (partly) solved in the near future.

Trajectory analysis show that in periods with enhanced CO_2 concentrations air was transported over the continent i.e. industrialized areas, especially over Germany (South East). These periods mostly occurred in winter. The concentrations of CO_2, and both the carbon isotopes were also strongly correlated during these periods indication that the main source for CO_2 was combustion of fossil fuel.

By use of the carbon isotopes the relative contribution of the anthropogenic sources can be estimated. An example of such an estimate is given for December 24, 1992. The concentration and ^{14}C changed from 380 to 425 ppmv, and from 113 to 107 pmC (percent modern carbon) respectively. The air was coming from the north of Germany during this period. The anthropogenic contribution of approximately 50 % was calculated using the increase in concentration and the decrease in ^{14}C. It is assumed here that the CO_2 exchange by vegetation in this winter period was negligible, and consequently, soil respiration and litter decomposition are the only biogenic sources for CO_2. These sources were assumed to contribute for the other 50% in the change in concentration. The ^{13}C of the emitted CO_2 (anthropogenic and biogenic) was calculated on -24 promilles. The absolute emission of anthropogenic CO_2 was estimated using a emission inventory and the trajectory of that particular day. An anthropogenic flux contributing to the change in atmospheric CO_2 was estimated to be 9 g CO_2 m^2 d^{-1}.

Using the calculated ratio between anthropogenic and biogenic sources (50%/50%) a biogenic flux of 9 gram CO_2 m^{-2} d^{-1} is calculated. As comparison the estimated flux of the grasslands in the surroundings of the site Cabauw is 2 g CO_2 m^{-2} d^{-1}. This value for the flux is calculated for the November 1992, when the assimilatory and respiratory processes of the vegetation were still active. (In the period November 1993 to December 1993, estimated emissions by grasslands range from 1 to 10 gram CO_2 m^{-2} d^{-1}).
A better estimate of the anthropogenic emission of CO_2 will be obtained in the near future with better description of spatial distributions of sources combined with the transport model.

8) The present stock of carbon in living biomass, litter and stable humus and the annual accumulation of carbon in stems for fifteen forest types has been quantified from inventory data on growth and standing volume, and forest soil information in combination with literature data on forest biomass. The forrest area in The Netherlands is about 330000 ha, mainly young plantations of conifers. The present standing volume is 170 m^3 ha^{-1} and the average volume increment was 9.0 m^3 ha^{-1} y^{-1} over the period 1984 to 1989. At present approximately 63.7 Tg C is stored in the entire forest, including dead organic matter in the forrest soil. About 60% of the carbon is stored in the humus of the soil compartment. The average carbon stock in the stable humus is approximately 110 Mg C ha^{-1}, whereas only 60 Mg C ha^{-1} and 20 Mg C ha^{-1} is contained in respectively the living biomass and the litter layer. About 0.66 Tg C of atmospheric carbon is stored annually (by means of stem volume increment). About 50% of the annual storage is harvest each year. This implies that the Dutch forests act as a sink with a strength of approximately 0.33 Tg C y^{-1}. The nett accumulation for the whole forest area amounts at present about 1 Mg C ha^{-1} y^{-1}. The current sink acting of the Dutch forest can most likely be explained by the fact that the forests are young and still in building phase. However, this sink is not always as strong as reported here. The latest forest inventory reported an average annual volume increment of 7.8 m^3 ha^{-1} y^{-1}. The net storage rate as reported here, decreases correspondingly. The presented results therefore, depend very much on year tot year variation in growth of forest caused by climatic variability. The net annual sequestration probably varies in between 0.2 and 0.4 Tg C y^{-1}. According to the investigators long rotation with species as oak, beech, and Douglas-fir are most suitable for long-term storage.

2.4 Future research

The uncertainties in the estimates of the so-called fertilization flux is still very high. Another problem is the uptake and loss of carbon by changes in land use. The measurements of NRP I have indicated that a substantial amount of CO_2 (equivalent to 3000 kg C ha^{-1} y^{-1}) is emitted by peaty soils induced by lowering of the water table. It is to be expected that the reverse process, uptake of carbon by peat formation will take place if the water level in peaty meadows is at lower depth than present.

The plans to restore the former conditions in many areas in The Netherlands of very high water tables have clear consequences regarding CO_2 emissions. Emissions due to peat oxidation will be stopped and an enhanced uptake of CO_2 will take place not only due to the fertilization effect but also due to peat formation. This situation indicates that is very important to improve our knowledge regarding the exchange of CO_2 between the atmosphere and ecosystems which are able to sequester large amount of carbon in their soils. Intensified research on the exchange of CO_2 between grass lands and the atmosphere, including fertilization effect, peat formation and peat destruction has a high priority in this respect.

3. METHANE (CH_4)

3.1 Preparation studies and organization

The programming of the CH_4 cluster has been based on three preparatory studies. One study presented estimates of the CH_4 emissions and their uncertainty ranges for The Netherlands based on literature (Born et al., 1991). The other two studies (Leffelaar et al., 1991 and Diederen, 1992) discuss priorities and criteria for the CH_4 research in The Netherlands.

Inventory of Dutch CH_4 emissions

A first attempt to quantify Dutch CH_4 emissions and to estimate ranges of uncertainties was made by Van den Born et al. (1991). The major conclusions of this work are presented in Table 3.1. This inventory indicated that enteric fermentation, landfills, the oil and gas industry, and organic soils cover about 90% of the total national methane emissions. On a global scale these sources are relatively less important, covering about 47% of the global methane emissions. Sources like rice paddies, biomass burning and coal mining are important on a global scale, but are absent or minor sources in the Netherlands. The large uncertainties in emission estimates are large, both on a national and global scale.

Criteria and priorities

Starting point of the research program on methane within the NRP was to achieve a significant reduction of uncertainties in knowledge on important emission sources (Leffelaar et al., 1991 and Diederen, 1992). Important criteria developed for the planning of the programme were the relative importance of the sources, the ranges of uncertainty and the availability of specific expertise on emission sources. The source strength of methane from enteric fermentation in The Netherlands is relatively well known. It was therefore decided not to plan any research activities on this item, although it is the largest source on a national scale. The uncertainty of this source in developing countries, however, is large, but the specific expertise on the differing diet situation and the impact on physiology was too poor to plan a research project. Rice paddies are of no importance on a national scale, but contribute significantly on a global scale. As the appropriate expertise on this subject was available, it was decided to formulate a research project on this topic. Furthermore, it was decided to plan research on the emissions from landfills, the oil and gas industry, and organic soils. Also, a number of research projects were formulated to validate local emission measurements, and to extrapolate the information to a larger spatial scale.

Table 3.1
Relative contribution (%) of national and global sources tot CH_4 emissions in The Netherlands in 1989/1990 and the world (Van den Bom et al., 1991)

Source	Netherlands	Globe
Animals - enteric fermentation	40	13
Landfills	27	7
Oil & gas industry/distribution	16	8
Wetlands/organic soils	7	19
Ocean/coastal waters	4	2
Freshwater	2	1
Animal waste	2	6
Waste Water treatment	0.3	4
Rice paddies	-	18
Termites	-	7
Biomass burning	NE	9
Coal mining	NE	6
Other	2	1
Emission range (weight units)	710-1230 Gg CH_4 y^{-1}	310-990 Tg CH_4 y^{-1}

NE: Not Estimated, not zero; -: Not applicable

Organization of the CH_4 cluster
The overview of the coherence between the research projects within the cluster is presented in Figure 3.1. Three relative important national sources were studied: the oil and gas industry, the landfills and the organic grassland soils. The organic grassland-soil related projects aimed at understanding the processes of methane formation and consumption in the organic soils. The results of experiments and measurements are integrated in a model of the methane flux from the soil to the atmosphere.

The following projects are part of the CH_4 cluster:

B. Biogenic sources

BRP. Soil parameters controlling methane production and emission from rice paddies (LUW; project no. 850009)
BMF. Methane formation by anaerobic consortia in organic grassland soil (LUW; project no. 853120)
BMC. Methane consumption by indigenous grassland microflora. (LUW; project no. 853122)
BGM. Effects of grassland management on the emission of methane from grassland on peat soils (LUW; project no. 853121)
BMMF. Modelling methane fluxes from and to grass covered peat soils (LUW; project no. 853123)

A. Anthropogenic sources

AL1. Greenhouse gases from landfills in The Netherlands. (TNO-ME; project no. 850023)

AL2. Landfill gas formation, emission and recovery in The Netherlands (TNO-ME; project no. 853105)

AOG1. Quantification of CH_4 emissions due to natural gas losses and petroleum production (TNO-ME; project no. 850008)

AOG2. Quantification of methane emissions in the exploration and production of natural gas and petroleum in The Netherlands. (TNO-ME; project no. 853104)

V. Evaluation and validation

VCI. Validation of source strengths of atmospheric CH_4 using carbon isotope ratios (ECN; project no. 852097))

VEV. Evaluation and validation of the CH_4 emissions in The Netherlands and contributions from various sources (TNO-MW; project no. 853124)

VUA. Methane emission of the Amsterdam urban area (LUW; project no. 853125)

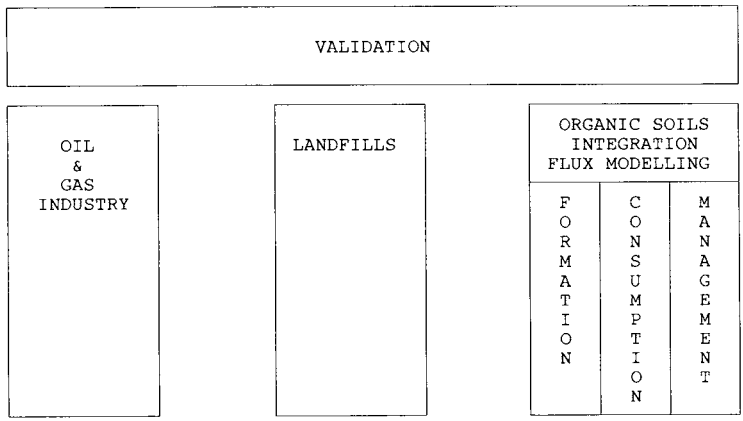

Figure 3.1
Schematic overview of the research of the projects within the methane cluster of the NRP in The Netherlands

3.2 Methods

Biogenic sources

Rice paddies (BRP). In this project the impact of various soil related parameters on the CH_4 emission from wetland rice fields was studied. Methane fluxes from wetland rice fields in the Philippines were monitored with a closed chamber

technique as described by Schütz et al. (1989) during two wet seasons (1991 and 1992) and one dry season (1992). The effects of soil-sulphate, soil-salinity, and organic manure on CH_4 emission were studied in experiments where gypsum, salt and green manure were added respectively. The effect of a calcareous soil was studied by a comparison with a non-calcareous soil. Methane oxidation in the rhizosphere was studied using a specific inhibitor of methane oxidising bacteria. The research was done in close co-operation with a project of the International Rice Research Institute (IRRI) in the Philippines which aims at collecting base-line CH_4 emission data from Asian rice fields.

The integrated CH_4 grassland projects. The main aims of the CH_4 research projects on grassland are the understanding and quantification of methane formation and consumption in grassland on peat soils, and of the net fluxes of methane between soil and atmosphere by experiments and simulation modelling.

Four different scales are distinguished, i.e.

(i) micro organisms in pure culture studies;
(ii) batch experiments with homogenised soils;
(iii) intact soil columns;
(iv) field scale.

The modelling aims to inter-relate the data obtained from the different scales.

Grasslands cover more than 35% of the total surface area in the Netherlands, of which 32% is on peat soils. The study sites are located in the major peat area of the western part of the Netherlands, around Zegveld (52°07'N 4°52'E). The predominantly eutrophic peat originates from sedges, reeds and wood, and generally have a clayey top-layer. Maximum peat depth is about 6 m. The organic matter content ranges from about 40% in the top 10 cm to about 90% below a depth of 60 cm, generally. Soil pH ranges between 3.5 and 5.0. A lowered mean ground water level, fertilizer application and removal of the grass crop via grazing and mowing are the major measures that take place on intensively managed grassland. On extensively managed grassland the vegetation is cut once a year in summer. The studied sites include both intensively managed, drained grassland and extensively managed natural grasslands. On intensively managed grassland, located at Zegveld, two typical sites have been chosen, i.e. site '8B' with a mean ground water level of 30 cm and site 'Bos 6' with a mean ground water level of 60 cm. Next to the effect of ground water level, effects of fertilizer application and grazing versus mowing on net exchanges of CH_4 between peat soil and atmosphere are investigated. On extensively managed grassland, three typical sites have been chosen in the Nieuwkoopse Plassen area with mean ground water levels of 5, 10 and 15 cm. Data on ground water level, soil and air temperatures, soil water filled pore spaces, soil nitrate contents and net CH_4 fluxes have been monitored on a weekly basis from September 1993 onwards. Measurements will continue till about August 1995.

Methane formation grassland soils (BMF). Soil profiles were taken from the two Zegveld grassland sites, with water tables of 30 cm and 60 cm below surface. The soil profiles were sectioned, taken to the laboratory in sealed plastic bags, and stored at 4°C. Inside an anaerobic glove box soil samples (20 g wet weight) from each section were transferred to 300 ml serum bottles, containing 40 ml of anoxic

distilled water. The stoppered bottles were incubated under a N_2 atmosphere (50 kPa overpressure) at 15°C, in the dark. The initial pH of the suspended soils ranged from approximately 4.8 to 5.5. At certain time intervals samples were taken from the head space as well as the liquid phase and analyzed for gases (CH_4, CO_2, H_2) and fatty acids or alcohols, respectively.

Methane consumption grassland soils (BMC). Soil samples from different depths (0-5, 5-10, 10-20, 30-40 cm) were taken from Zegveld. To investigate the kinetics of methane oxidation of these different depths the soil was placed in bath cultures in 300 ml flasks with gas tight septa and incubated with 1, 10, 100 and 10,000 ppmv methane, respectively, in artificial air with 1% (v/v) CO_2 (Bender et al., 1992). For the enrichment of methanotrophic bacteria with different affinities for methane, soil (100 g) was incubated in a system receiving a continuous gas-flow of 4 ml/min containing methane at 4 different concentrations (1, 10, 100 and 10,000 ppmv).

Grassland management (BGM). Net CH_4 emissions from grassland on peat soils in The Netherlands have been monitored with vented closed flux chamber (Hutchinson et al., 1981) from September 1993 onwards. Monitoring will continue in 1994 and 1995. At Zegveld, intensively managed grassland on peat soil with a mean ground water level of 30 cm and intensively managed grassland on peat soil with a mean ground water level of 60 cm have been investigated. Also, on both Zegveld sites the effects of nitrogen fertilization and grazing versus mowing on net CH_4 emissions have been investigated. Finally, CH_4 fluxes from three extensively managed grasslands at Nieuwkoop have been measured as well.

Modelling methane fluxes grassland soils (BMMF). This project started in September 1993 and will last for 4 years. It aims at developing a process model for methane fluxes to and from organic grassland soils. Water dynamics at Zegveld will be obtained to be used as input for a gas transport model for Zegveld. With this gas transport model oxygen dynamics and methane transport will be described. The oxygen dynamics will be used as input for the methane production model. Results of the integrated model of methane production (results from project BMF), consumption (results from project BMC) and transport will be compared with field fluxes measured in project BGM. This comparison in combination with a sensitivity analysis of the model will show which aspects need most attention in further research. The model for methane production has already been developed. It calculates the dynamics of biomass, acetate, and methane formation. Moreover, it describes experimental data from the BMF project quite well. In this model, besides oxygen, a time lag is incorporated before methane production can start. This lag period could, in a later stage, be specified in terms of the presence of electron acceptors like nitrate and sulphate. If we succeed in incorporating all major processes in a realistic way, it will be possible to test two hypothesis: in-situ methane emission is low, because production is limited by the short duration of the anaerobic periods during wet periods; during dry periods, methane uptake by the soil is controlled by methane transport from the atmosphere to the methanotrophs.

Anthropogenic sources

Landfills - emission measurements (AL1). This preparatory study compared two measurement methods at three landfills from June until September 1991 (Verschut et al., 1991) a dynamic closed-chamber method Balfour et al., 1987, Reinhart et al., 1992) measuring concentration differences between air entering and leaving a closed chamber
system (10 m^2; 8-10 replicates per landfill; during ≥24 h) and a micrometeorological method (Fowler et al., 1989) measuring concentration gradients and wind velocity along a pole of 10 m height situated for about two weeks at the centre of a landfill, combined with vertical and horizontal flux calculations and wind speed profiles.

Landfills - gas formation, emission and recovery (AL2). To improve the reliability of the emission quantification from landfills the methane material balance (Emission = Formation - Oxidation - Recovery; no accumulation assumed) is investigated. Landfill gas formation is determined in two ways: by emission measurements at various landfill sites (micrometeorological method Oonk et al., 1995) and adding the amount of landfill gas recovered and oxidized to the amount emitted; by evaluating recovery efficiency from the results of recovery projects in relation with landfill geometry, composition of the top liner system and the lay-out of the recovery system - landfill gas formation is subsequently obtained as the product of the amount of landfill gas recovered and the recovery efficiency. Oxidation data used are from literature (UK-DoE, 1993, US-EPA, 1990, Oonk, 1993). The formation of landfill gas is subsequently modelled, by correlating the formation to waste composition, age and amount of waste landfilled.

Natural gas losses and petroleum production (AOG1). A first estimate of CH_4 emissions due to the production and treatment, the high pressure transport, the distribution and consumption of natural gas and the production and transportation of petroleum in The Netherlands was made by an engineering approach (Nielen, 1991). This estimate was based on available information of measurements, emission factors and production and consumption data for 1989.

Exploration and production of natural gas and petroleum (AOG2). Three types of methane emissions related to oil and gas production were examined. The continuous emissions due to leakages of systems used and from off-gases of various gas treatment installations have been quantified by an engineering approach using: emission factors, material and energy balances; knowledge on maintenance and testing procedures; information from NOGEPA on amounts of associated gas produced; information on process equipment, compressors, turbines etc. The irregular emissions due to periodic tests and maintenance of installations were quantified partly as a result of an engineering approach as described above, partly by a combination of measurements and dispersion modelling. The third type of methane emissions, the incidental emissions due to failures of devices also are estimated by the combination of measurements and dispersion modelling. The ambient CH_4 concentration at Kollumerwaard, located North-west of the Groningen gas field, which has been registered permanently since July 1991, was screened on indications of events elevating the background concentration. This was done in combination with information on meteorology, potential source location and distribution calculations by the Plume model.

Valuation and validation

Validation by carbon isotope ratios (VCI). At two sites atmospheric CH_4 concentration has been monitored continuously (Eisma et al., 1995 and Kieskamp et al., 1995): the 200 m meteorological tower at Cabauw and the 'Vuurtoren' island near Amsterdam. The latter to examine the emissions of the Amsterdam area (see VUA). Carbon isotopic analysis in atmospheric CH_4 taken at Cabauw has been performed as well. However, due to interference from near-by $^{14}CH_4$ emissions of Pressurised Water Reactors (PWR) ^{14}C could not be used as a tracer for fossil and biogenic CH_4 in Europe. Instead, from the $^{14}CH_4$ record at Cabauw, emission factors from PWRs have been determined.
A laboratory intercomparison was organized for ambient CH4 measurements exercise, with ECN, TNO, KEMA and LUW as participants.
Results obtained for Cabauw are interpreted by analysis of air mass trajectories, meteorological data and the application of a Lagrangian transport model in combination with CH_4 emission inventories and comparison of isotopic measurements with characteristic isotopic values of CH_4 sources. Also, the CH_4 concentration records measured simultaneously at Cabauw and Durgerdam are compared. As a first approximation a GIS application has been developed in which the excess CH_4 concentration at Cabauw (> 1.75 ppmv) is related to the area over which the air mass was transported (using 36 h backwards air trajectories at Cabauw and assuming a constant, 1000 m mixing layer height). The total area covered by the air trajectory was calculated as a reverse plume. The CH_4 emission flux required for explanation of the observed excess CH_4 was subsequently assigned to all points in a 0.1° x 0.1° grid over Europe. Methane emission from water surfaces was assumed to be zero with exception of the oil-and gas-production sites on the North Sea.

Evaluation and validation from various sources (VEV). This project validates present knowledge of CH_4 emissions by comparing measured concentrations with those calculated by atmospheric dispersion models from emission and meteorological data. Continuous measuring is performed by using gas chromatography on two monitoring sites, namely, Arnhem and Kollumerwaard from 1990 and 1991 onwards, respectively. Dispersion modelling has not started yet.

Urban area (VUA). The urban methane emissions have been quantified by modelling methane air concentrations and comparison of calculated and measured imission concentrations. Measured concentrations were obtained from ECN (project VCI, Eisma et al., 1995) from the methane monitoring station at Vuurtoreneiland, about 2 km east of Amsterdam. Methane emissions estimates from the urban area were based on literature data Amstel et al., 1993 Veldt et al., 1993) road traffic, the natural gas distribution network, and industrial sources. The Danish OML model was selected for calculation of immission concentrations (Lofstom et al., 1992). This Gaussian plume model has a preprocessor to calculate dispersion height, atmospheric stability and turbulent mixing from synoptic and balloon meteorological measurements. The synoptic measurements were obtained from Schiphol Airport, the balloon data from De Bilt. Emission data and dispersion data were fed to the OML model to calculate immission concentrations for the

continuous methane monitoring station. The calculated emissions were compared with data collected at the monitoring site. To obtain an estimate of the increase in ambient methane concentrations due to the urban plume, methane concentrations of Cabauw (some 40 km south of Amsterdam) (data obtained from ECN) were subtracted from the Vuurtoreneiland data.

3.3 Results

Biogenic sources

Rice paddies (BRP)

Soil-sulphate. The methane emission from plots amended with 6.66×10^3 kg ha^{-1} gypsum ($CaSO_4$) was reduced by 55-70% compared to non-amended plots (Figure 3.2). The reduced CH_4 emission upon gypsum application was most likely due to inhibition of methanogenesis by sulphate-reducing bacteria. Observed SO_4^{2-} concentrations in the soil solution of gypsum-amended plots were well above minimum concentrations reported in the literature for successful competition of sulphate-reducing bacteria with methanogens. The data indicate that CH_4 emissions from rice grown on high-sulphate containing soils or gypsum-amended soils is low compared to non or low-sulphate containing soils. However, fertilization of rice fields with $(NH_4)_2SO_4$ will not necessarily result in lower CH_4 emissions because the amounts of sulphate added are relatively low.

Soil-salinity. Rice is often grown on saline soils. To investigate whether the presence of salinity results in lower CH_4 emissions NaCl salt was added to a rice field. Pore water EC increased to about 4 dS m^{-1}, which caused a reduction by 25% only in CH_4 emission. It was shown that the CH_4 production in the salt-amended field was strongly reduced compared to the control field (Table 3.2).
However, CH_4 oxidation in the salt-amended plot was even more inhibited than CH_4 production. This resulted in about equal net CH_4 fluxes from both salt-amended plots and non-amended plots. The data illustrate the importance of knowledge of both CH_4 production and CH_4 oxidation when estimating CH_4 emission and show that a reduction in CH_4 production does not necessarily lead to reduced CH_4 emissions.

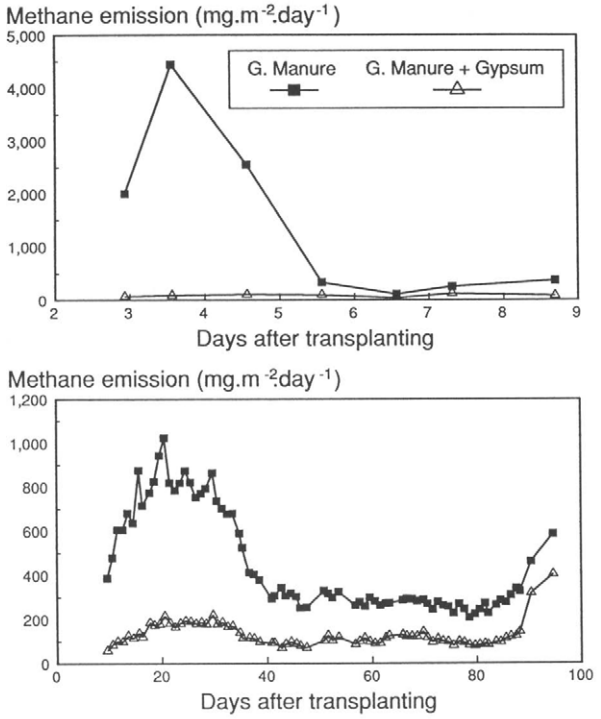

Figure 3.2
The impact of Gypsum application on methane emission from wet rice fields during the wet season in 1992 at The Philippines

Calcareous soils. CH_4 emissions from rice grown on a calcareous soil were higher than from a non-calcareous soil. The seasonal pattern of CH_4 emission differed, with a more pronounced emission peak early in the season, probably due to the favourable pH for CH_4 production in the calcareous soil. The difference in emission between the two soil types was no longer observed when the fields were fertilized with green manure, indicating that the "soil"-factor may be overruled by the input of organic matter.

Application of organic manure. Application of green manure stimulated CH_4 emissions. CH_4 emission was highest during the first half of the growing season in plots that received more than $11x10^3$ kg ha^{-1} of green manure. Ebullition contributes significantly to total CH_4 transport, if rice fields receive high inputs of organic matter. The impact of organic manure on CH_4 emissions, at different locations of the world, can be described by a dose-response curve if CH_4 emission from the organically amended plots is expressed relative to CH_4 emission from mineral fertilizer treatments. Such an approach may prove to be useful when estimating CH_4 emissions from larger regions if information on the amounts of organic manure used in the region becomes available.

Plant-mediated gas transport. Plant mediated CH_4 transport was shown to be described by diffusion only. The results combined with data from the literature suggest that the rate limiting step in plant-mediated methane transport is diffusion of CH_4 across the root/shoot junction.

CH_4 oxidation in the rice rhizosphere. CH_4 oxidation in the rice rhizosphere was studied using methyl fluoride, a specific inhibitor of methane oxidising bacteria. CH_4 oxidation in the rice rhizosphere depended on the growth stage of the rice plant and becomes much less important when the rice plant reaches the ripening stage. Therefore seasonal patterns of CH_4 emission in rice fields do not only depend on changes in CH_4 production but also on changes in CH_4 oxidation. These findings indicate that methanotrophs do not oxidise a constant percentage of the CH_4 produced throughout the growing season.

Table 3.2
Average methane flux from triplicate soil cores of rice fields with and without salt amendment, during anaerobic and aerobic incubation, and percentage CH_4 oxidized

Sampling date a) Flux)	CH_4 flux (nmol cm^{-2} h^{-1})				CH_4 oxidized (nmol cm^{-2} h^{-1})		CH_4 oxidized (% of anaerobic	
	anaerobic		aerobic					
	no salt	salt	no salt	salt	no salt	salt	no salt	salt
76 DAT	11.02	2,75	1.31	1.32	9.71	1.43	88	52
96 DAT	24.53	7.04	3.29	2.17	21.24	4.87	88	70
110 DAT[b)]	20.82	7.42	2.75	2.99	18.07	4.43	87	60

a) DAT = days after transplanting
b) 110 DAT =1 week after harvest

Methane formation grassland soils (BMF)
Formation of CH_4. Methane formation was observed almost exclusively in the upper 10 cm of the soil, with the upper 5 cm of the soil being most active. Below 10 cm, methane formation decreased drastically. The compact peat layers, below 30 cm did not show any methane formation (Figure 3.3). In the upper two soil layers (0-5 cm, 5-10 cm) formation of CH_4 was exponential indicating that there was no substrate limitation. The difference observed for both layers therefore probably results from a difference in the number of methanogens originally present. A doubling time of approximately 2-3 days could be determined. In the top soil methane formation started immediately. The highest rate of CH_4 formation was reached after approximately 40 days for the low water table soil type (0.41 mmol l^{-1}; 0.042 mg CH_4 g^{-1} dry soil d^{-1}). Ultimately, CH_4 formation more or less paralleled CO_2 formation.

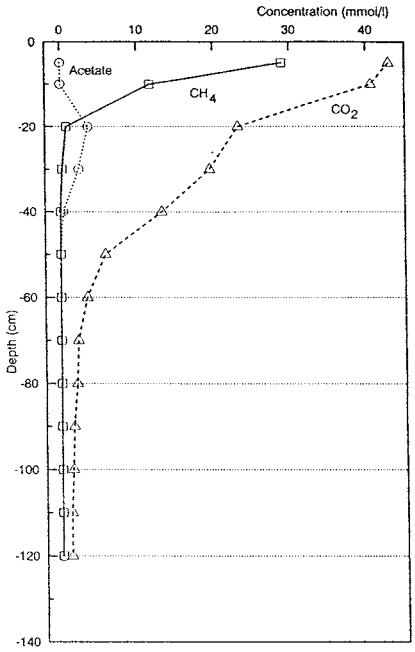

Figure 3.3
The relationship between formation rates of CH_4, CO_2, acetate and soil depth of various soil slurries after incubation under a N_2 atmosphere. All concentrations expressed in mmol l^{-1}. Data obtained for the low water table series (60 cm)

Formation of CO_2. Also, the formation of CO was highest in the top soil (0 - 10 cm). The initial rate of CO_2 formation amounted up to 1.3 mmol.l^{-1} d^{-1} (0.36 mg CO_2 g^{-1} dry soil d^{-1}). CO_2 formation showed no lag-phase; the rate of CO_2 formation decreased in time. After 40 days a small increase was observed again, due to an increased activity of aceticlastic methanogens that produce CO_2 and CH_4 from acetate.

Formation of acetate. Analysis of the soil suspensions for fatty acids showed that the two upper layers (0-5 cm, 5-10 cm) produce considerable amounts of acetate. The top layer produced up to 4.8 mM acetate, which was rapidly degraded by methanogens after 35 days. Small amounts of propionate and butyrate were formed as well.

Methane consumption grassland soils (BMC)
Methane oxidation. All 4 applied concentrations were biologically degraded by this type of grassland soil. The highest oxidative activities, especially for lower concentrations (1-100 ppmv), were observed between 5 and 20 cm. One reason for the lower activity in the highest depth (0-5 cm) may be that this region was very wet at the time of sampling, resulting in hampered gas diffusion. The time course of methane degradation is plotted in Figure 3.4 for the initial concentrations of both

10,000 (A) and 1 (B) ppmv methane. A correlation between CH_4-concentration and degradation rates was observed (0.19 nmol and 1.9 µmol g^{-1} dry soil d^{-1} for 1 and 10,000 ppmv, respectively) and, most important, it is demonstrated that this soil acts as a sink for methane even at concentrations well below 1 ppmv.

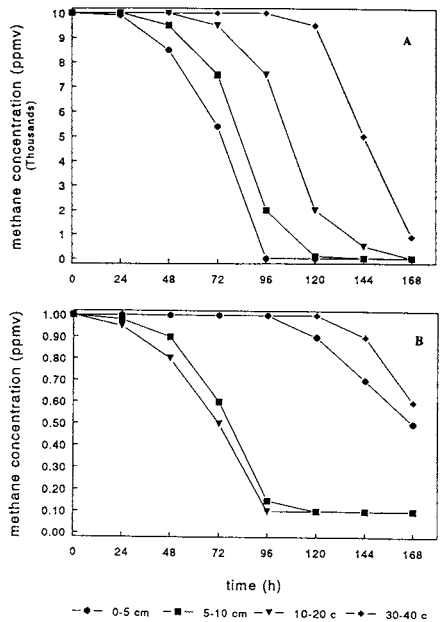

Figure 3.4
Degradation rate of methane in samples from organic grassland soil from different depths in batch cultures at initial concentrations of 10,000 ppmv (A) an 1 ppmv (B) CH_4

Identification of methanotrophic bacteria. A decrease of the efflux concentration was observed after 14 days of incubation in the columns incubated with 10,000 ppmv. Here the degradation rate (caused by microbial growth) increased to 19 µmol g^{-1} dry soil d^{-1} within 7 days. In the column incubated with 100 ppmv the increase was observed after 35 days of incubation. At lower concentrations (1, 10 ppmv) the efflux concentrations remained constant for 40 days, but then these methane concentrations were also degraded. The main goal of this part of work will be to isolate and identify the strains of methanotrophic bacteria which are responsible for the degradation of atmospheric methane concentrations.

Grassland management (BGM)
The effect of ground water table. The results presented here are based on observations until July 1994. Grassland with a high ground water level and a relatively thin aerobic layer is expected to show more CH_4 emission and/or less immission than grassland with a low ground water level and a relatively thick aerobic layer. However, the site with the relatively high ground water level showed equal or only slightly higher net CH_4 emissions than the site with the relatively low

ground water level during the measuring period. Net CH_4 fluxes were low in the period October 1993 - July 1994, in general less than 0.1 mg CH_4 m^{-2} d^{-1}.

The effect of N-fertilization and mowing/grazing. Nitrogen fertilization could decrease the consumption of CH_4. Mowing or grazing could effect CH_4 emissions by influencing the amount of organic material that is added to the soil annually. However, there were no clear differences between the treatments during the measuring period.

The effect of management intensity. Differences between the different sites were quite large (Figure. 3.5) as were the spatial variations at each of the sites. The site with the lowest CH_4 emission had a somewhat lower ground water level than the other sites. In the period January - June 1994, CH_4 emission ranged from 0 to 185 mg CH_4 m^{-2} d^{-1}. CH_4 fluxes were much higher at the Nieuwkoop area than at Zegveld.

Modelling methane fluxes grassland soils (BMMF) (See before)

Anthropogenic sources

Landfills - emission measurements (AL1)
Dynamic closed-chamber measurements. The spatial variation in methane emissions proved to be large on each landfill: differences over a factor of 1000 were measured (e.g. 0.09 g m^{-2} d^{-1} to 225 g m^{-2} d^{-1} for one landfill and 7.2 g m^{-2} d^{-1} to 3150 g m^{-2} d^{-1} for another). It is clear that emissions are spatially very inhomogeneous and that local coincidental factors determine the emission rate through the top-layer. So, the measurement data are not representative for the total landfill area.

Micrometeorological measurements. Depending on the type and age of the landfill, the amount of waste available, the height of the waste tip, the efficiency of landfill gas recovery and the composition of the top layer emissions were measured ranging from 2 to 150 g m^{-2} d^{-1}. The method as such yielded quite reliable and uniform emission data.

Decay rate. A waste decay rate constant (k) of $k = 0.1$ could be calculated on the basis of these emission measurements, combined with data on amounts of waste, landfill composition and age. This implicates a half-life of 7 years.

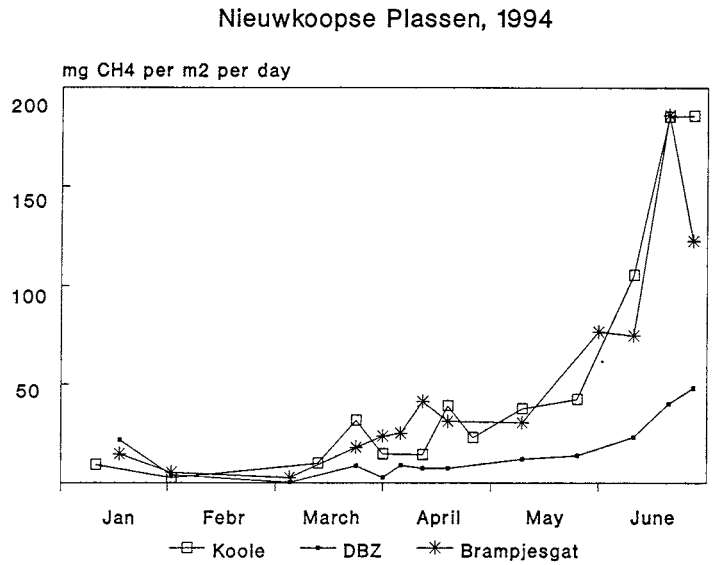

Figure 3.5
Time course of mean CH_4 fluxes (mg CH_4 m^{-2} d^{-1}) at three different sites in the Nieuwkoop area

Landfills - gas formation, emission and recovery (AL2)

Landfill gas formation. A first - very preliminary - result of the modelling of landfill gas formation by correlating the formation to waste composition, age and amount landfilled is:

$$\alpha_t = \xi \; 1{,}87 \; Q \; C_0 \; k_1 \; e^{-k_1 t}$$

where, C_0 = amount of degradable organic carbon in the waste in kg ton^{-1} (the Dutch mean value for C_0 is 112 kg ton^{-1}; k_1 = rate constant of biodegradation (0.094 y^{-1}); Q = amount of waste landfilled in ton; t = time after dumping of the waste; α_t = formation of landfill gas in m^3 y^{-1} (with a mean methane content of 57 vol%); ξ = 0.58 (formation factor).

Landfill gas recovery. In 1993 about 124 million m^3 of landfill gas was extracted; 85 million m^3 was utilized. In 1992 methane emission reduction was 57 Gg (Adviescentrum Stortgas, 1994). During the exploitation period of the landfill, landfill gas formation increases with increasing amounts of waste in place. After closure of the waste tip, landfill gas formation gradually declines. Landfill gas recovery normally starts when the landfill is closed. The effectiveness of recovery is increased when a top-liner system is applied. Normally this is done 5 years after closure of the landfill. The environmental impact of landfill gas recovery is closely connected to its integral recovery efficiency, being the ratio of formation and recovery throughout the years. High integral efficiencies can only be obtained,

when landfill gas recovery starts during the exploitation period of the landfill. The technology for doing this is available, and proves to be very cheap, when landfill gas recovery is reckoned in the design phase of the landfill (J. Oonk 1993 and 1994).

Emission estimates. Landfill gas emissions have been estimated using the material balance as described in 3.2.2. The CH_4 emission estimates from landfills in The Netherlands range from 400-500 Gg y^{-1}. Uncertainties in these estimates are due to uncertainties in the amounts of waste landfilled, amounts of methane formed per ton of waste and amounts of methane oxidized in the top-soil of the landfill.

Natural gas losses and petroleum production (AOG1)

The results of this study are summarized in Table 3.3. Total CH_4 emissions have been estimated between 127 and 220 Gg y^{-1}. At this point it must be emphasised that none of these estimates have been based on actual measurements, which are required for more accurate quantification.

Exploration and production of natural gas and petroleum (AOG2)

The project has not been finalized yet. Preliminary results of the engineering study are partly in a qualitative form still. Table 3.4 presents the sources and source strengths of methane in the oil and natural gas exploration and production.

Table 3.3
Methane emissions from natural gas losses and petroleum production in The Netherlands in 1989 (Gg y^{-1})

Sector	Methane emission
Natural gas	
Production/treatment	40-70
High pressure transport	6.5
Distribution	65-79
Consumption	15-30
Petroleum	
Production	1-35
Total	127-220

Table 3.4
Sources of methane emission in the oil and natural gas exploration and production

Sources	Strenght[1]
Emission during exploration	
- drilling	minor
- well tests	moderate/major
Emission during exploitation of natural gas	
continuous	
- vents	major
- flares	moderate
- exhaust gases of turbines	minor
- exhaust gases of reciproking engines	moderate
- exhaust gases of furnaces	minor
- chronic leaks in production	major
- chronic leaks in gathering and transport	minor
- glycol dehydration	major
- treatment of formation water	moderate
- use of pneumatic devices	moderate/major
- condensate treatment	moderate
- condensate storage	minor
- purge gas drom venting systems	moderate
non-continuous	
- maintenance in production	minor
- maintenance of gathering and transport pipelines	minor
- incidents and accidents in production	moderate
- incidents and accidents pipelines	minor
Emission due to exploration of oil	
continuous	
- flaring of associated gas	minor
- exhaust gases of reciproking engines	minor
- treatment of production water	minor
non-continuous	
- non-exhaust engine emissions	minor
Abandoned phase	
- chronic leaks from abandoned wells	none

[1]minor: <2 Gg y^{-1}; moderate: 2-10 Gg y^{-1}; major: 10-100 Gg y^{-1};

The emission estimates are presented for the three phases exploration (drilling and testing), exploitation (production) and abandonment (closed wells) which can be distinguished within the oil and gas industry. Venting, chronical leaks in production and glycol dehydration are major sources during natural gas exploitation. Well-tests and the use of pneumatic devices can be major sources as well. The

stationary measurements show several times a year extremely high methane concentrations (e.g. Figure 3.6). The peak of 10 July 1991 was probably caused by an emission of $150x10^3$ kg of methane at a distance of 25 km from Kollumerwaard. Three mobile measurement campaigns were carried out, each covering one day. The first campaign was around the Groningen gas field and consisted of several gas production locations. The second campaign consisted of two exploration activities in the very near neighbourhood of measuring location Kollumerwaard. The third campaign consisted of some of the production locations of the first campaign and two exploration locations of the second campaign. On each of these three occasions no elevation of background concentration was measured.

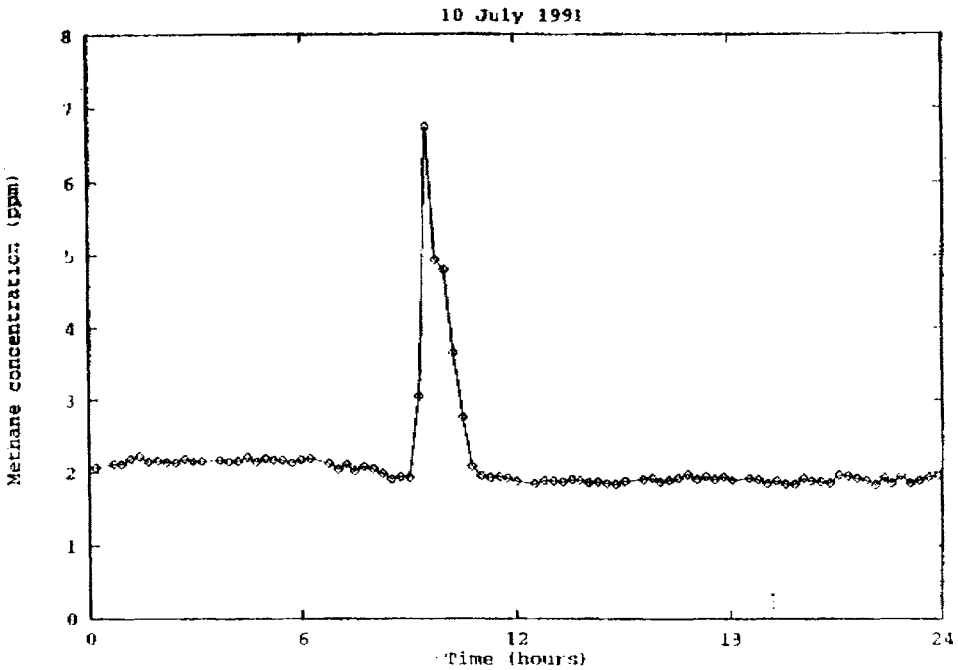

Figure 3.6
Daily course of air methane concentration at Kollumerwaard, The Netherlands, at July 10, 1991

Evaluation and validation

Validation (VCI)
Laboratory intercomparison exercise. A CH_4 laboratory intercomparison exercise was organized, with ECN, TNO, KEMA and LUW as participants. From the exercise, concluded that interlaboratory variation of ambient CH_4 analysis (2.0 ppmv) is better than 2% (R.S.D.). The within-laboratory variation varied from between 0.1 - 0.9% (R.S.D.).

Trajectory modelling. Figure 3.7 shows CH_4 emissions calculated from the CH_4 concentration record at Cabauw in combination with air trajectory analysis. A strong preference of air trajectories coming from the south-west direction is observed. Accordingly, the calculated CH_4 emissions from that direction will be obtained with satisfactory accuracy whereas for other wind directions, prolonged CH_4 monitoring is still required. 'White spots' can be observed in Figure 3.7 for which no emissions have been calculated because since the start of Cabauw CH_4 monitoring only 1 or 2 air trajectories have crossed these areas.

Comparison of monitoring sites. By comparing the concentration records of Durgerdam and Cabauw, CH_4 sources around Amsterdam have been analyzed. In particular, elevated CH_4 emissions from the areas south-east and north of Amsterdam have been observed. These sources have not yet been positively identified due to lack of sufficient overlapping data but may result from cattle and wetland activity in these areas.

Isotopes. The applicability of $^{13}C/^{12}C$ analysis in atmospheric methane is demonstrated in Figure 3.8 which shows a CH_4 concentration record at Cabauw on July 22 (1994) and its corresponding change in $^{13}C/^{12}C$ isotopic ratio. The isotopic ratio of the source of the CH_4 peak is calculated from the excess CH_4 (i.e., above 1.70 ppmv background) and the measured ^{13}C. In this case, the average of the CH_4 sources in the peak amounts of 55 promilles, which indicates a 'normal' composition of fossil and biogenic methane in the peak.
From the $^{14}CH_4$ record at Cabauw, emission factors from Pressurised Water Reactors (PWR) have been determined. The $^{14}CH_4$-PWR-emissions are higher by a factor 1.5 ± 0.3 as compared to the present knowledge (Eisma et al.). In effect, a larger fossil methane contribution is needed in order to obtain the average global $^{14}CH_4$ concentration.

Evaluation and validation from various sources (VEV)
Levels and phenomenology of methane concentrations. The measurements at Arnhem and Kollumerwaard have revealed the characteristic levels and main variables determining variation in atmospheric ground level concentrations. Average concentration levels on both monitoring sites were 2.0 ppmv (compared to 1.5 ppmv in 1979 at Terschelling Hollander, 1979) and range from 1.8 to generally 3.0 ppmv and incidentally higher values. These incidental short-lasting high concentrations are most probably due to local sources close to the monitoring site and their occurrence differed therefore at both monitoring sites (Vosbeek, 1993a and 1994b.

Ground level atmospheric concentrations appeared to relate mainly to wind direction and diurnal variation. The variation with wind direction (Figure 3.9) mainly reflected the presence of up wind source areas, modified to a small extent by systematics in meteorological variables like wind speed and mixing height.

The patterns for both monitoring sites are striking similar. When conditions with higher wind speed only were selected from the data set, the wind direction profile remained largely unchanged, only resulting in a slightly lower concentration level. These lower concentrations were most pronounced in the maximum of the profile.

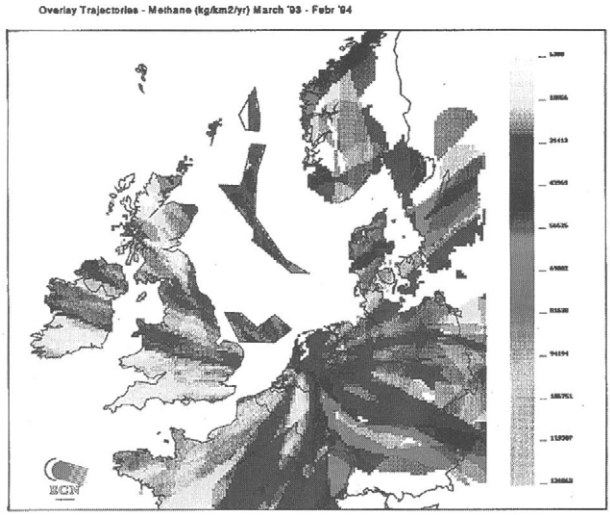

Figure 3.7
Methane emissions (kg CH_4 km^{-2} y^{-1}) calculated from CH_4 concentration records from March 1993 until March 1994 at the 200 m meteorological tower at Cabauw, The Netherlands, in combination with air trajectory analyses

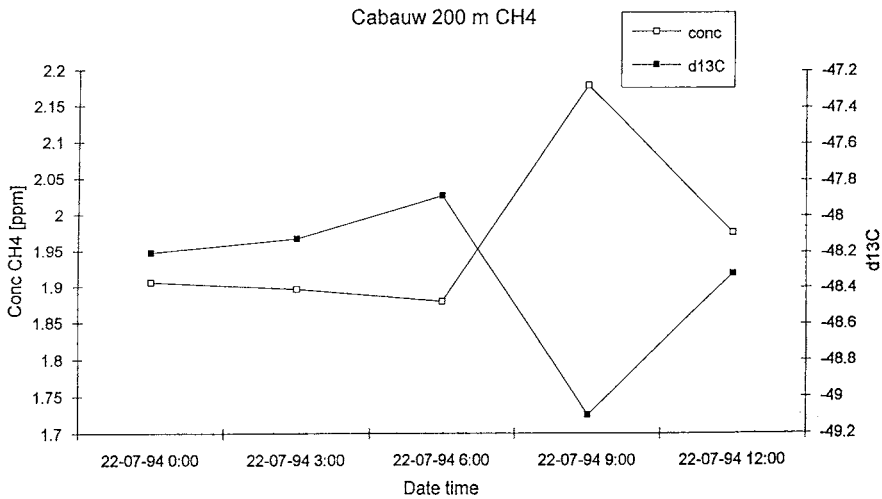

Figure 3.8
Methane concentrations (ppmv, left axis) and ^{13}C of peak source the ^{13}C values have been calculated from the measured ^{13}C and the excess of CH_4 in the peak (I.E. > 1.70 ppmv). ^{13}C are expressed relative to PDB international carbon isotopic standard

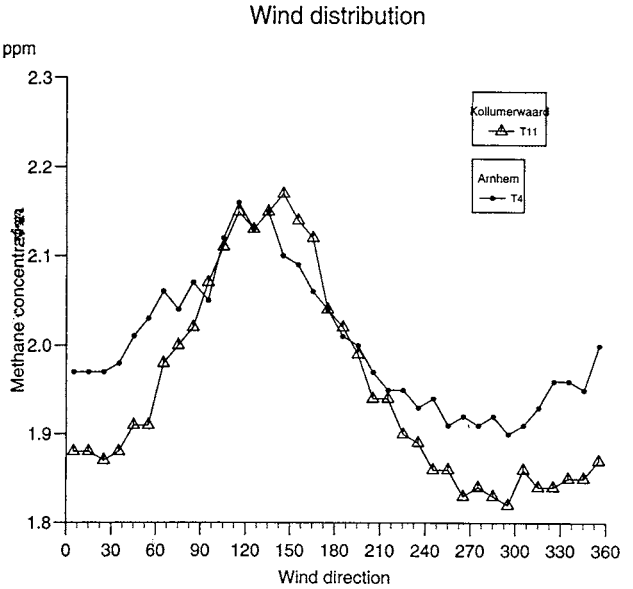

Figure 3.9
The effect of wind direction on the methane concentration in air at Arnhem and Kollumerwaard, The Netherlands

Diurnal variation mainly reflected the effect of mixing height on the concentrations resulting essentially from ground level sources and probably to a lesser extent from diurnal variations in source strength. The shift in the maximum at Arnhem compared to Kollumerwaard is related to traffic emissions. The maximum is delayed to rush hour time and coincides with a maximum in CO concentrations.

Source contributions. This part of the project cannot be reported in this stage.

Urban area (VUA)
Urban methane emission estimate. The total CH_4 emission for the Amsterdam area, based on the use of recent emission factors, was estimated to be about 2 Gg-CH_4 y^{-1} (Veenhuysen et al., 1994). The overall methane emission from an urbane area in the Netherlands is low when compared to other sources of methane.

Comparison of measured and calculated concentrations. Both calculations and measurements showed a low increase in methane concentrations due to urban emissions (some tenths of a ppmv). This was supported by some incidental windward and leeward measurements of concentration levels of methane around Amsterdam. The measurements indicated incidental elevated concentrations of some ppmv, that could be attributed to nearby highways and special meteorological conditions. The calculated concentrations were in general higher than the measured concentration. This should be seen in the perspective that for Gaussian dispersion modelling a margin of error of about ± 100% is usually being accepted. Even if this margin is considered and uncertainty in roughness length is

taken into account, the calculated averages were high with respect to the observed ones. This points to the use of too high (although generally accepted) emission factors for road traffic.

3.4 Integration of results

Biogenic sources

Rice paddies. Up to now, global estimates of CH_4 emission from rice agriculture do not take differences in soil types into account. Bachelet and Neue (Bachelet et al., 1993) proposed to include soil characteristics to revise these global estimates. Using three published techniques to estimate global CH_4 emissions Bachelet and Neue found that emission estimates were reduced by 25% if a distinction according to soil type was made. This indicates that including soil characteristics in the global estimates could significantly alter the global estimate. The results from project BRP provide a legitimate basis for the use of 'soil type' correction factors to estimate CH_4 emissions from flooded rice fields.

On the other hand, the results of BRP showed that in the pre-flooding period and the period after harvest considerable amounts of methane may be released from flooded rice fields. In previous monitoring studies these periods were not included, which may cause an underestimation of total seasonal emission by 10-15%.
Furthermore, the BRP results show that a reduction in CH_4 production does not necessarily cause a proportional reduction in CH_4 emission. The data illustrate the importance of both CH_4 production and CH_4 oxidation when estimating CH_4 emission, which emphasises the relevance of the lay-out chosen for the grassland research projects (BMF, BMC, BGM and BMMF). The data also show that the ratio between production and oxidation may depend on environmental conditions. This is important when looking for mitigation options to reduce methane emission from rice fields.

Organic grasslands
Formation. By choosing an experimental set-up with certain physical parameters a microbial community is selected, which might represent the natural community only partly. Since this project aimed at the microbial groups that lead to CH_4 formation, anaerobic conditions were applied (N_2-atmosphere). In all incubations immediate formation of CO_2 was observed, indicating that an immediate anaerobic mineralization of organic matter took place. Mineralization was highest in the top soil. Comparable CO_2 formation rates have been described for other organic soil types Magnusson, 1993). In this initial stage no H_2 and almost no CH_4 was formed, suggesting that the reducing equivalents produced during mineralization may be transferred to alternative electron acceptors, like NO_3^- or SO_4^{2-}. However, the contribution of reduction of NO_3^- or SO_4^{2-} cannot be deduced from the experiments, yet. In the absence of alternative electron acceptors, H_2 is used to form methane or acetate by methanogens or homoacetogens respectively. Addition of H_2/CO_2 resulted in the top soil in the formation of high amounts of acetate, indicating that active homoacetogens are present. It has been shown before that homoacetogens play a significant role in H_2 turnover (Conrad et al., 1989) and CO_2 conversion to acetate (Thebrath et al., 1992). It cannot be

concluded from this projects data, however, to what extent homoacetogens and hydrogenotrophic methanogens contribute to the conversion of H_2 and CO_2. The accumulation of acetate in the top soil indicates that the activity of aceticlastic methanogenesis is limiting during the first 30 days. However, between 30 and 50 days the degradation of acetate started to exceed the rate of acetate formation in this layer. From that moment on methanogenesis is limited by the supply of acetate and the rate of CH_4 formation parallels the rate of CO_2 formation. This 1:1 ratio is expected when organic matter (CH_2O) is fully degraded to CO_2 and CH_4 under methanogenic conditions.

The accumulation of acetate observed here, without immediate conversion to CH_4, has also been described for a Beech forest soil (Küsel et al., 1994) and for a waste water disposal pond (Kotsyurbenko et al., 1993). This phenomenon appears to occur especially at low temperatures, in anoxic environments, where homoacetogenic bacteria contribute to the turnover of H_2 (vide supra). At the start of the experiment the soil samples did not contain detectable amounts of acetate. This indicates that the conditions that were applied in the experiments, differ from the conditions in situ. Apparently, in the field the top soil is aerobic enough to prevent the accumulation of acetate.
Consumption. The degradation of atmospheric concentrations of methane has often been observed (King, 1993 and Whalen et al., 1990). The found depth profile with a very high methanotrophic activity in depths between 5 and 15 cm has been found by Jones and Nedwell as well (1993). The evidence for different types of methanotrophic bacteria, a low affinity type for the degradation of high concentrations of methane and a high affinity type for the degradation of low (atmospheric) concentrations of methane could be described in kinetic studies by Bender and Conrad (1992) But until now, the high affinity strains could not be isolated and described.

Management. Measured net CH_4 fluxes were low in the period October 1993 - July 1994, in general less than 0.1 mg CH_4 m^{-2} d^{-1}. Literature data for comparable sites range from 0.1 for a poorly drained grassland soil in winter (Jarvis et al., 1994) to 0.8 mg CH_4 m^{-2} d^{-1} for an unfertilized pasture (Mosier et al., 1991). Soil analyses in Zegveld showed relatively high sulphate and nitrate concentrations in the soil, especially in the top soil. Both nitrate and sulphate, will have blocked CH_4 production. Low soil temperatures in winter will also have contributed to low microbial activities in the soil. It is suggested that flux measurements in summer will provide better insight in possible differences in CH_4 fluxes due to grassland management. The measured emission rates from extensively managed grasslands ranged from 0-185 CH_4 m^{-2} d^{-1} over the period January-June, 1994. In a review and assessment report of methane emissions from wetlands, Bartlett & Harris (1993) arrived at an estimate of 90 mg CH_4 m^{-2} d^{-1}. The results presented here indicate that, in the period September 1993 - July 1994, the impact per hectare natural grassland on cycling of CH_4 was much higher than the impact per hectare intensively managed grassland. It has to be emphasised that methane production by cattle is not included in these estimates.

Anthropogenic sources

Landfills

Formation. There are numerous factors, that influence either the amount of degradable organic carbon in the waste, the emission factor, or the gas formation rate. In general these factors are all connected to: waste composition, waste treatment and site management. The waste composition determines the amount of degradable organic carbon in the waste, which is the raw material for landfill gas. Besides that waste composition determines numerous other factors, such as the presence of nutrients or inhibitors and the humidity of the waste. Mechanical pre-treatment, homogenisation, particle size reduction and baling, the extent of composition, the dumping method and the addition of water have significant effects on landfill gas formation. Finally, site geometry, landfill gas recovery, percolate water management and top-liner system determine gas formation. Waste composition is rapidly changing in the Netherlands, due to separate collection of the vegetable, fruit and garden (VFG) waste. The remaining waste, after separation, still contains large amounts of degradable organic carbon. This waste will be less humid, and the organic carbon left is relative slow degradable. So as a result of this separate collection, landfill gas formation will decrease, without any doubt, but to what extent is not yet clear.

Recovery. Due to efforts of energy companies, waste treaters, NOVEM, and the 'Adviescentrum Stortgas' [the landfill advisory board] the number of landfill gas projects is increasing rapidly. About 30 landfill gas projects will be operational by the end of 1995. Developments after 1995 are uncertain. Due to the separate collection of VFG, landfill gas formation becomes unpredictable and landfill gas projects are economically less attractive. This is counteracted by increased legislation. Landfill gas projects are more and more requested in allowances, and in future high (integral) efficiency recovery projects will be prescribed. This means that in future landfills are obliged to start gas recovery gas during the exploitation of the landfill.

Oil and natural gas. The results of the AOG projects allow for an engineering study in which first order estimates of methane emissions can be made. The main result of such an inventory will not be the emission estimate as such, but it facilitates the choice of future research needs of specific sources. It is not a simple matter to compare emissions from the Netherlands with those reported from abroad. Partly, this is because the quantifications are not equally detailed and partly because regional circumstances make any comparison difficult. With this proviso, one can conclude from Table 3.5 that the relative methane emissions from oil and gas production in the Netherlands are comparable with emissions in other countries of north-western Europe. The emissions per cubic meter of gas produced are somewhat higher in the United States, while the emission situation in the former Soviet Union is far worse.

Table 3.5
Emission factors for natural gas production (in percentage of production)

Netherlands	
- on shore	0.03-0.05
- off shore	0.6-1.0
Total	0.15-0.25
Western Europe	0.15-0.3
United States	0.2-0.3
Former USSR	±2

Validation

Source strength. In general, the CH_4 emissions which have been estimated by trajectory modelling in this project are in reasonable agreement with results from national emission inventories, except for the North Sea area. For this region, the CH_4 emission as calculated from the Cabauw record, is higher as compared to the emission inventory for this region. Interestingly, the CH_4 emission calculated for the Netherlands from the Cabauw record (Figure 3.7) is about 1100 Gg y^{-1}, which corresponds quite well with the national data of the Dutch Emission Inventory (Berdowski, 1994). Detailed interpretation of Cabauw data using a mesoscale transport model has not yet been performed since this requires emission data on a European scale which are not yet available.

Isotopic ratios . The applicability of the $^{13}C/^{12}C$ analysis of atmospheric CH_4 is demonstrated in Figure 3.8. For this example it can be concluded from the $\delta^{13}C$ data that the excess CH_4 is of mainly fossil origin. Detailed stable isotopic analysis of atmospheric CH_4 peaks (time series) can be a promising tool in source apportionment studies. By contrast, isotopic analysis in air samples taken at Cabauw at a fixed time of the day was found to be of little use. Due to interference of $^{14}CH_4$ emissions of Pressurised Water Reactors (PWR), ^{14}C could not be used as a tracer as discussed before.

'Local' monitoring sites. Average concentration levels at the monitoring sites Arnhem and Kollumerwaard were 2.0 ppmv and ranged from 1.8 to generally 3.0 ppmv and incidentally higher values. These incidental short lasting high concentrations are most probably due to local sources close to the monitoring site. The lower limit generally is measured at higher wind speeds and western wind direction and it agrees with the northern hemisphere background concentrations as reported recently (Khalil et al., 1993D, 26: 59-62]).
Although a Gaussian type of model is not well suited to predict immision concentrations for exceptional situations, the model was used in the Amsterdam Urban Area study for more detailed analysis. It has been investigated whether the elevated concentrations, which were observed incidentally, resulted merely from extreme meteorological conditions and/or source configuration, or were the results of unknown sources of methane. Analysis indicated a possible influence of nearby

motorways on the monitoring site although calculated concentrations were higher than measured concentrations. With the large uncertainty of Gaussian dispersion modelling kept in mind the following conclusions can be made:

- the sources in the urban area still remain only roughly constrained;
- the emission of methane from the Amsterdam area is only a minor contributor to the national methane emission;
- no major unknown urban sources of methane are present in the Amsterdam area;
- traffic is the dominant source of urban methane;
- the gas distribution system is of such a high quality, that losses are an unimportant source of methane in the Amsterdam urban environment.

Finally, there are indications that, given the uncertainty and variation in roughness length, the source strengths were even lower than estimated.

3.5 Future research needs

Integration and validation

Most research during NRP phase-I involved reduction of uncertainty of source estimates. Extrapolation, integration and validation have been formulated, but could have been performed to only limited extent so far. Extrapolation to national and western European scale have to be carried out using the results achieved during the NRP phase-I, combined with international data. This requires a GIS approach and the development of emission models of the methane sources. Such an approach will lead to a database, including the relevant information required for flux extrapolation. The validation of source strengths has to be performed by a combination of several modelling approaches (e.g. empirical and trajectory modelling) on a national and regional scale, concentration monitoring at a number of permanent sites and the results of emission maps, produced by extrapolation.

Specific research needed for CH_4

Biogenic sources

Global sources. For the estimate of global emissions from rice paddies, already several techniques have been applied in which differences in soil types were accounted for. These techniques have to be developed further, also based on the results achieved in project BRP. Combining soil-type specific CH_4 production potentials with the WISE and EDGAR databases (Chapter 5) will allow identification of areas where high CH_4 emission can be expected and mitigation options may be especially successful.

No attention has been paid to methane emissions from ruminants, as the level of knowledge on a national scale is already quite high. However, on a global scale further examination of this sources would be very valid, especially in relation to the food composition in different regions of the world. So far, it has not been able to extrapolate national knowledge on this item successfully.

Grasslands. The results presented here are mainly preliminary results of measurements and analysis. Only parts of the annual flux courses have been established yet, for one base year only. The monitoring started in September 1993 and has to be continued for a longer period to be able to indicate the annual contribution of peat soils to the emission of CH_4 to the atmosphere. Also, proper

interpretation and integration of results has not been done yet, due to the phase the research projects were in at the time of the preparation of this report. This has to be accomplished to yield maximum profit of the work done until so far. Especially, the flux modelling can prove to be an important tool for the data processing of these projects. The water dynamics at Zegveld could serve as input for a gas transport model for organic grassland soils. With this gas transport model oxygen dynamics and methane transport can be described. The oxygen dynamics can be used as input for the methane production model. Results of an integrated model of methane production, consumption and transport van be compared with measured field fluxes. This comparison in combination with a sensitivity analysis of the model will show, which aspects need most attention in further research. When the major processes are incorporated in a realistic way, it will be possible to test two hypothesis: during wet periods in situ methane emission is low, because production is limited by the short duration of the anaerobic periods; during dry periods methane uptake by the soil is controlled by transport from the atmosphere to the methanotrophs. These results may be interpreted in relation to future management ideas for the grassland area in The Netherlands.

Traditional pig farming. In the definition phase of this methane cluster, emissions from traditional pig farming systems were not considered as an important contributor to total national CH_4 emissions. Lately, there have been several indications that such systems might be important point sources. As these pig systems are quit abundant in The Netherlands, especially, in a few areas, it is to be considered to at least perform a pilot study to the relative importance of this source type.

Anthropogenic sources
Landfills. The level of uncertainty in emission estimates from landfills still is high. Especially, the limited knowledge of the oxidation processes in the top-layer and the impact of waste composition on formation processes determine this uncertainty. Further research on these items will reduce uncertainty in emission estimates from landfills considerably.

Oil & gas exploitation/transportation. During NRP phase-I it was clearly indicated which sources were of major or moderate importance for the exploration, exploitation and transportation of oil and natural gas. However, these sources have been far from quantified yet. further research of the major sources is required to obtain better quantification. At least, first order estimates and uncertainty ranges will be required to use data on this source for regional extrapolation purposes.

4. NITROUS OXIDE (N_2O)

4.1 Preparatory studies and organization

The programming of the N_2O cluster was based on two preparatory studies. One study involved comparison of measurement techniques (TNO; project no. 850012). The second study was the inventory of emissions of N_2O (and CH_4) from the Dutch territory based on literature (RIVM; project no. 850019).

Comparison of measurement techniques
The aim of this study was to compare and assess the applicability of incubation techniques, static and dynamic enclosure techniques and the gradient method, and to assess the spatial and temporal variability of N_2O fluxes and soil N-contents in peat soils under grassland. Five research groups participated. Two comparison campaigns indicated that static enclosure techniques show large spatial and temporal variability, probably caused by the variability in soil parameters and processes. Static enclosures are, therefore, most useful in combination with studies of the regulating factors and processes of N_2O production and consumption. The gradient technique integrates fluxes over larger spatial scales and this method is most suitable for determining the mean flux from a field plot.

Inventory of Dutch emission
The inventory by Van Den Born et al. (1991) was a first attempt to quantify Dutch N_2O emissions and to assess the uncertainty in estimates. The major conclusions of this study summarized in Table 4.1 show that only a few sources may dominate the Dutch N_2O emissions. Grasslands, particularly those on peat soils, are major sources of N_2O in The Netherlands, even if the low end of the range is considered (Table 4.1). The tentative estimates suggested that other potential major N_2O sources are transportation, arable lands, fresh water and marine aquatic systems and sewage water treatment.

Organization of the N_2O cluster
In the programming of the N_2O cluster the source strength played an important role. Other criteria were the uncertainty in the source estimate, the available expertise, the availability of technical options to achieve emission reductions (policy relevance), and expected future developments in society that will have an effect on emissions.
Grasslands were given a high priority, because they cover about 30% of the Dutch land area, and because of the high expected emission rates (Table 4.1). Detailed studies of N_2O from arable lands and forests were not included in the N_2O cluster. A great number of studies of N_2O fluxes from arable lands were done in the period 1979-1986, mostly in the U.S.A., U.K., and Germany. Therefore, only exploratory measurements in arable lands were included (project 852096). Forests were given a low priority because they cover less than 10% of the Dutch land area. This does not imply that forests may not be a potentially important source, since in The Netherlands N-inputs from deposition are among the highest worldwide.
Transportation, aquatic systems and waste water treatment are research topics in the N_2O cluster, because of the uncertainty regarding their source strength. One project investigating N_2O from combustion processes was included because of the uncertainty that exists since the discovery of sampling artifacts in studies performed before 1988 (Muzio et al., 1988).
Current knowledge was synthesized in a global inventory emphasising biogenic soil emissions, but also including inventories for all other sources of N_2O identified so far.
All components of the cluster aim at verifying the results of the preparatory study, reducing the uncertainty and assessing options for reduction of emissions. The following components are part of the N_2O cluster:

F. Fossil sources

FT. Traffic. Investigation of the contribution of traffic to the N_2O emissions both now and in the future (IMW-TNO; project no. 850030)

FC. Combustion. N_2O emissions in combustion processes with respect to the generation of electricity (KEMA; project no. 850006)

B. Biogenic sources

BAQ. Aquatic systems, including:

BAQ1. Water/atmosphere exchange of nitrous oxide in marine systems (NIOZ; project no. 850027)

BAQ2 Measurements in fresh water systems (IMW-TNO; project no. 852096)

BST. Sewage treatment. The generation of N_2O in sewage treatment plants (BKH; project 853133)

BGR. Grasslands. Research was organized in the integrated N_2O grassland project, consisting of 4 sub-projects investigating soil fluxes:

BGR1. Measurement of the atmospheric concentration of N_2O from biogenic surface sources in general and grassland ecosystems in particular (TNO-IMW; project no. 852096)

BGR2. Factors affecting the emission of nitrous oxide from grasslands in the removal of nitrate from soil by denitrification (LUW-TPE; project no. 852074)

BGR3. The effects of nitrogen fertilization and grazing on the emission of N_2O from grasslands (NMI; project no. 852073)

BGR4. The emission of N_2O from grasslands (IB-DLO; project no. 852078)

G. Global inventory

Modelling of soil emissions of nitrous oxide for global studies (RIVM; project no. 852079).

In Section 4.2 the methods and research activities of individual projects are discussed. Section 4.3 is a summary of the results of the individual projects and Section 4.4 describes the integration of the results over the Dutch territory. Section 4.5 discusses future research needs.

Table 4.1
Emission of N_2O in The Netherlands for 1990 made by Van den Bom et al. (1991) and estimates based on results of NRP-I.

	Emission (Gg N y^{-1})	
	Van den Bom et al. (1991)	NRP-I[1]
Fossil sources		
energy sector	0.2-1.2	0.1-0.3
industry	0.1-0.6	-
commercial & residential	0.1-0.6	-
transportation	1.4-4.1	3.4
Biogenic sources		
grassland, mineral soils	2.8-5.7	NE
grassland, peat soils	3.0-11.8	NE/C
arable land & horticulture	2.2-4.1	-
natural soils	0.0-0.4	-
fresh water systems	0.7-2.2	NE
coastal waters	2.2-2.8	NE
sewage treatment	1.6-3.2	<0.3
Total	14.4-36.8	

4.2 Methods

Validation of results
In the preparatory comparison of measurement techniques the concentration measurements by various research groups was compared. In order to compare methods two standard gas mixtures were distributed to all participants involved in N_2O measurements. The results of this comparison will be analyzed according to ISO standards.

Fossil sources
Traffic (FT). On the basis of the available literature emission factors for various types of automobiles and types of engines were estimated. On the basis of this future emissions from transportation were assessed.

Combustion (FC). Measurements were carried out at power plants, chemical industries, an oil refinery and at a waste incineration plant. A sampling method was developed to prevent N_2O formation in the sample caused by the above mentioned "sampling artefact". Measurements were carried out within 24 hours after sampling. The N_2O emissions were obtained from N_2O concentration measurements in stack gases and total stack flow calculated from the fuel throughput.

Biogenic sources

Aquatic systems (BAQ)

BAQ1. Measurements were carried out in the Northwest Indian Ocean, the open North sea and the Dutch coastal zone. The measurements in the Indian Ocean were conducted in 1992 during cruises in the Somali Basin, the northern part of the Arabian Sea, and a cruise between both areas. North Sea measurements were made during a cruise in the central and northern North Sea in 1990. During other North Sea cruises discrete samples were collected. Measurements in the Scheldt river and estuary were done in autumn 1993 and spring 1994.
The N_2O measurements included analysis of discrete samples from various depths and monitoring of surface waters and atmospheric concentrations. In addition, oxygen and nutrient concentrations were determined to obtain information about the relation between local N-cycling and N_2O formation. For the calculation of concentration anomalies and air-sea fluxes, water temperature, salinity and wind speed were measured. The N_2O concentration measurements were done with a computerised gas chromatograph.

BAQ2. A measurement campaign with the gradient technique of TNO was done performed along the Dutch Waddenzee. Fluxes from the fresh water of the Ketelmeer were determined in spring 1994. The N_2O flux was calculated from the concentration gradient measured over the water surface and wind speed.

Sewage treatment (BST). This project involved a literature study and measurements in sewage treatment plants. The objective of the literature survey was to assess the potential for N_2O production and emission in the different types of treatment plants in The Netherlands as a function of specific process variables, as well as an inventory of the nitrogen influent and removal for each plant type in The Netherlands.
Exploratory measurements were done in installations in Capelle a/d IJssel (covered carrousel with point-aeration) and Alblasserdam (installation with separated nitrification and denitrification), in combination with measurement of oxygen concentrations in the water and determination of the amount of influent water and nitrogen.

Grasslands (BGR). The integrated grassland project includes 4 projects carried out by different research groups, that focused on flux measurements, modelling N_2O production and consumption in grassland soils and assessment of the effect of different agronomic practices on N_2O emission. The soils studied included peat, sand and clay soils. Measurements were made at different levels of integration, with soil incubation techniques, enclosure techniques and atmospheric measurements with the concentration gradient technique to cover the field scale. Parallel to the integration levels of measurements, the modelling approaches cover the micro-scale to understand the processes at the chemical physical level, the rhizotron level, and the field scale.

BGR1. This project consisted of two parts:
A) Comparison of measurement techniques in two campaigns as a continuation of the work in the preparatory study, to investigate the spatial variation of

N_2O fluxes measured with the static enclosures of NMI and dynamic enclosures and the gradient method of TNO.

B) Continuous measurements of N_2O fluxes from grasslands with the gradient technique, daily in the afternoon from April 1993 onwards. Additional measurements in arable lands were made in spring 1994, but results are not yet available.

BGR2. This project concentrated on the processes affecting N_2O emission from grasslands by denitrification. The levels of integration range from the chemical/physical level via the rhizotron scale to the field scale. Incubation experiments were conducted with peat samples collected at Zegveld to investigate the effect of sample location and depth, temperature, initial incubation condition (anaerobic/aerobic), and kind and amount of fertilizer applied. The rhizotron experiment was designed to study the effect of the depth of the ground water and fertilizer application on N_2O losses from a sand soil covered by grass. The N_2O loss to the atmosphere was determined with the closed chamber technique by a photo-acoustic spectroscopic infra-red gas analyzer and a multi-sampler described by Velthof and Oenema (1994). Subsoil N_2O profiles were determined with a gas chromatograph provided with an Electron Capture Detector (ECD).

BGR3. The effects of N-fertilization, grazing cattle and soil type on the N_2O emission from grassland were investigated on the basis of field measurements, including:

A) Monitoring study during a period of 2 years at 4 sites, including two peat soils with different ground water levels (Table 4.3) in Zegveld, a sand soil in Heino and a clay soil in Lelystad. At each site N_2O fluxes were measured weekly with 6 replicates in unfertilized and mown, N-fertilized and mown, and N-fertilized and grazed grassland. The N-fertilizer applied was calcium ammonium nitrate. Additional measurements were done of soil mineral N, herbage yield and N-uptake, soil organic N-mineralization rates and N-inputs from cattle excreta. At all sites denitrification potentials and mineralizable C content were determined at 5 depths between 0 and 60 cm. At the Zegveld sites denitrification rates were monitored. Temporal and spatial variability were assessed with detailed measurements on the sand, clay and peat soils. In the sand soil and one of the peat soils N_2O concentration profiles in the soil were determined regularly during 1 year. The atmospheric N_2O loss was determined with the closed chamber technique, a photo-acoustic spectroscopic infra-red gas analyzer and a multi-sampler described by Velthof and Oenema (1994).

B) Assessment of the effect of type and level of N-fertilizer application on N_2O emission. Fertilization was done with ammonium (NH_4^+), nitrate (NO_3^-), combinations of NH_4^+ and NO_3^-, urea, NH_4^+ + nitrification inhibitor, and injected and surface applied cattle urine and cattle slurry.

C) The development of a management scheme to minimise N_2O losses, based on the above experiments and literature data.

BGR4. This project concentrated on the effect of urine spots on N_2O emission at the field scale, and consisted of an experimental and a modelling component. In the experimental part oxygen and N_2O profiles were measured in homogeneous grassland soils as a function of soil depth and distance to field drains. These

simultaneous oxygen and N_2O profiles provide information on the relation between N_2O production and soil moisture. The spatial variability at the 0.001-0.1 m scale was studied in undisturbed soil columns. An experiment with application of urine to undisturbed sandy grassland columns was done in autumn 1993. The results of another identical experiment are not yet available.
A model is being developed that describes the controls of N_2O fluxes to the atmosphere. The model is based on the field scale model originally developed by Verberne (1992), and it describes C and N transformations in the soil in four interacting sub-models representing:

i) grass development including C and N turnover and effects of grazing;
ii) transport of water and dissolved N;
iii soil microbial C and N transformations;
iv) gaseous transport.

Global inventory. A preliminary global inventory of N_2O emissions from soils was compiled. The objective was a first validation of global soil emissions. It became clear that for that purpose all sources need to be known. Therefore, grid based emission estimates derived from the EDGAR database (chapter 5) and from the IMAGE project (NRP Theme "Integration of climate change research)" were used. For N_2O emission from mineral N-fertilizer and animal excreta a preliminary estimate of 1% of the N-input from was used, based on literature and supported by results of the integrated grassland project. These inventories were tested against inverse atmospheric modelling results for 4 latitudinal zones covering the globe.
An attempt was made to explain high observed N_2O fluxes after forest clearing observed in the tropics. Soil and plant samples were taken in a tropical rain forest zone in Costa Rica. A model developed by Wageningen Agricultural University, Department of Soil Science and Geology will be used to assess the amount of N liberated by mineralization of soil organic matter in the course of time after forest clearing and pasture development.

4.3 Results

Fossil sources

Traffic (FT). Emissions are based mainly on data from the Institute Français du Petrole (IFP). The major factors influencing N_2O emissions are

(i) the presence and type of catalyst;
(ii) the age of the catalyst;
(iii) the air-fuel ratio;
(iv) the temperature of the exhaust gas and
(v) other factors including tuning and maintenance of the engine.

The emission factors derived (Table 4.2) show that aged catalysts give rise to emission factors exceeding those for new catalysts by a factor 3. The study indicated that it is difficult to give uncertainty ranges for these emission factors.

Table 4.2
Emission factors for N_2O from different types of engines

Diesel, light	0.031
Diesel, heavy	0.2
Otto, without catalyst	0.015
Otto, with new catalyst	0.035
Otto, with aged catalyst	0.12

Combustion (FC). The observed stack gas concentrations of N_2O found for power plants fired with coal, oil and gas were close to ambient air concentrations (0.3 ppmv N_2O). The fluxes estimated on the basis of stack gas concentrations were low and in most cases negligible. For some types of gas turbines the concentrations in stack gases were somewhat higher, while the N_2O concentration for an oil refinery with oil as fuel, vacuum residue and refinery gas was 0.3 ppmv.
The N_2O concentrations in stack gases from boilers and gas turbines fired with high caloric gas and methane gas in the chemical industry were below 0.2 ppmv. Depending on the temperature in the combustor, the N_2O loss from fluidized bed combustors fired with several types of coal was found to be 4.5-49 g N_2O-N GJ^{-1}, with higher N_2O concentrations at lower temperatures.
The N_2O concentration at a domestic waste incinerator averaged 2.4 ppmv with peak values of 43 ppmv. The estimated emission from this incinerator was 13 mg N_2O-N kg^{-1} domestic waste.

Biogenic sources

Aquatic systems (BAQ)
BAQ1. In the Somali Basin N_2O concentrations in water showed a strong negative correlation with O_2 concentrations. Supersaturations of up to 700% were observed in the oxygen minimum zone. The calculated N_2O fluxes to the atmosphere ranged from 1.1 to 3.1 kg N_2O-N ha^{-1} y^{-1}. The water column of the Arabian Sea showed zones of extremely low oxygen concentrations, giving rise to N_2O consumption by denitrification. As this N_2O consumption zone was surrounded by water masses with high N_2O production by both nitrification and denitrification, surface waters were on balance supersaturated. The N_2O supersaturations in the water column ranged from almost zero to 1300% in surrounding waters.
In the North Sea the measured N_2O saturations varied from 98 to 105%, yielding an atmospheric flux of between -0.04 and 0.2 kg N_2O-N ha^{-1} y^{-1}. In the Scheldt river and estuary a close correlation was found between N-concentrations and N_2O production.

BAQ2. Measurements carried out along the Dutch Waddenzee showed high surface water N_2O concentrations compared to the open sea, with values of around 1000 ng l^{-1}. The observed emission from the Waddenzee was equivalent to about 1 kg

N_2O-N ha^{-1} y^{-1}. This is similar to fluxes observed from agricultural lands as shown in the integrated grassland project for mineral soils.

Sewage treatment (BST). The N_2O emission from treatment plants from various studies reported in the literature ranges between 0.01 and 6% of the total N-influent. The highest N_2O formation occurs in systems where nitrification and denitrification occur simultaneously, such as in aeration tanks, or if conditions for nitrification and denitrification are sub-optimal. During denitrification N_2O formation is inevitable. The N_2O formation during nitrification can be prevented if process conditions are optimal. Creation of conditions that are optimal for both denitrification and nitrification is not possible. The conclusion of the literature survey was that about 0.4% of the influent-N evolves as N_2O averaged over all types of installations. However, the exploratory measurements indicated a loss of N_2O of only 0.01% in two installations (Capelle a/d IJssel and Alblasserdam), both having a low influent load and very efficient N-removal.

Grasslands (BGR)

BGR1. There was a wide range in observed fluxes in two measurement campaigns of the method comparison of BGR1 and the preparatory study. No firm conclusions could be drawn regarding systematic differences between the various methods. The reason may be that methods representing smaller areas tend to give a larger variation than methods for larger regions, reflecting the scale of N_2O production processes that occur in so-called "hot spots". The measurement comparison campaigns in BGR1 showed that fluxes measured with static chambers vary at least one order of magnitude within a field.

Observed night-time fluxes were lower than daytime fluxes. The fluxes measured with the gradient method showed that the flux was not constant with height, possibly due to inhomogeneity of the source area. In the method comparison study the 24-hours measurements with the static enclosure method seemed to give somewhat lower fluxes, although the order of magnitude was similar to fluxes determined with the gradient method. The results from the monitoring studies with both methods give similar annual results.

Continuous measurements from April 1993 onwards showed that during rainy periods the flux determinations are more variable than during dry periods. Shortly after rain events the N_2O flux may increase rapidly by accelerated denitrification and nitrification and then decrease again. The N_2O pulses may also be caused by soil air that is forced out of the soil pores by infiltrating rain water and by negative fluxes by deposition of N_2O on wet surface soils, observed occasionally with both the gradient method and static chambers. However, during the second half of 1993 no strong N_2O pulses were measured, because of the high precipitation and high frequency of rainfall events. The estimate based on the gradient method for the flux from fertilized peat soils at Zegveld is 25 kg N_2O-N ha^{-1} y^{-1}.

BGR2. The peat soil incubation experiments indicated that net N_2O production by denitrification occurs during part of the incubation period for all treatments. No net N_2O loss occurs during nitrification after initial aerobic conditions. Relatively high amounts of N_2 evolved during the incubations. The differences found between soils in different plots and locations were only minor. The denitrification activity decreased with depth and increased with temperature.

In the rhizotron experiments the highest N_2O fluxes of 17 ± 2 g N_2O-N ha^{-1} d^{-1} were observed for the highest ground water level and highest N-applications, fluxes being mostly higher after a grass cut than before. Low N_2O fluxes were found for high ground water levels in containers where no N was applied (4 ± 2 g N_2O-N ha^{-1} d^{-1}) and for intermediate N-application (4 ± 3 g N_2O-N ha^{-1} d^{-1}). For these cases the fluxes before and after the cut were not different. No significant N_2O loss (0 ± 5 g N_2O-N ha^{-1} d^{-1}) was observed in containers with low ground water level and no N applied. Moisture content was found to be the major factor affecting N_2O concentration in the soil atmosphere.

Table 4.3
Estimated losses of N_2O for all treatments on sand and clay soil and two peat soil with different ground water levels. The % N_2O loss of the N-applied as calcium ammonium nitrate is calculated as the different between the N_2O loss from the fertilized plot and the unfertilized plot, presented as a % of N-fertilizer application. The fertilizer induced loss for grazed grassland is calculated for 1992 as the difference between the N_2O loss from fertilizer and animal N-excretion. Amounts of N excreted via urine and dung for 1992 were calculated using standard procedures

1994 Soil input treatment	1992 N-input	1992 N_2O loss	1992 % of N-input	1993 N-input	1993 N_2O loss	1993 % of N-input	1992-1994 % of N-input
Sand soil							
unfertilized; mown	0	1.2	-	0	1.0	-	
N-fertilized; mown	313	3.1	0.6	426	6.6	1.3	1.0
N-fertilized; grazed	743	7.3	0.8	426[1]	13.2	-	
Clay soil							
unfertilized; mown	0	1.0	-	0	0.5	-	
N-fertilized; mown	277	5.0	1.4	437	2.6	1.5	0.9
N-fertilized; grazed	557	10.6	1.7	437[1]	16.1	-	
Peat soil, grondwatertrap II[2]							
unfertilized; mown	0	2.1	-	0	1.8	-	
N-fertilized; mown	266	8.0	2.2	464	9.6	1.7	1.9
N-fertilized; grazed	521	11.9	1.9	464[1]	17.3	-	
Peat soil, grondwatertrap III[3]							
unfertilized; mown	0	12.9	-	0	4.2	-	
N-fertilized; mown	161	20.2	4.5	323	15.9	3.6	3.9
N-fertilized; grazed	356	36.0	6.5	323[1]	41.0	-	

[1] The N-input for 1993 does not include N from animal excreta; therefore the N_2O-loss as % of the N-input is not calculated.

[2] Grondwatertrap II = means annual low ground water level at 50 - 80 cm below surface.
[3] Grondwatertrap III = means annual low ground water level at 80 - 120 cm below surface; the mean level was about 20 cm lower than for the site with grondwatertrap II

BGR3. The results of the monitoring study showed peak fluxes of N_2O during the first 1-2 weeks after fertilization and grazing, except during dry conditions. High fluxes were observed during the wet period from July till the end of September 1993. Fluxes from the peat soil with low ground water level were much higher than from the other soils, and for all soil types the fluxes were higher for grazed plots then for ungrazed plots. Winter fluxes were low in all treatments, except those from the peat soil with low ground water level (grondwatertrap II, Table 4.3), where occasionally significant N_2O fluxes were observed in winter. Fluxes of N_2O from the clay soil in Lelystad were low during the wet period between July and September 1993, while fluxes from the other sites were high during this period.

The results of the monitoring study between March 1992 and March 1994 show losses of N_2O of up 36 (in 1992-1993) to 41 (in 1993-1994) kg N_2O-N ha^{-1}, observed in grazed plots on peat soils with deep ground water tables, while the losses for the corresponding periods for high ground water levels were 12 and 17 kg N_2O-N ha^{-1} (Table 4.3). These results are confirmed by the estimated 25 kg N_2O-N ha^{-1} y^{-1} based on the concentration gradient method of project BGR1. The N_2O losses from the grazed grassland on clay soil in Lelystad were also high: 11 and 16 kg N ha^{-1} for 1992-1993 and 1993-1994, respectively. For the grazed sand soil in Heino the losses were 7 and 13 kg N ha^{-1} for 1992-1993 and 1993 and 1994, respectively (Table 4.3). In fertilized and mown plots the N_2O losses for the 2-years period were about 1% of the N-input from synthetic fertilizer for the sand and clay soils, and 2% and 4% for the peat soils with respectively high and low ground water level (Table 4.3). In fertilized and grazed plots the N_2O loss for 1992 - 1993 amounted to 0.8% of the total N-input from synthetic fertilizer and animal excreta for sand soils, 1.7% for clay soils, 2% for the peat soil with high ground water level, and 6.5% for the peat soil with low ground water. In the calculation of the % losses no correction was made for mineralized N.

BGR4. Losses from urine spots calculated from field measurements appear to be less than 1% of the amount of urine N-input, even for doses of up to 1300 kg N ha^{-1}. The N_2O loss was promoted by high concentrations of mineral N and by wet soil conditions, during at least 2 months after the urine application. Direct damage of the grass by urine (urine scorch) led to less re-growth, lower N-uptake and less transpiration at the urine spot, and these conditions are favourable for N_2O losses via nitrification and denitrification.

Global inventory. The global estimated N_2O emissions for different sources were calculated for 4 latitudinal zones covering the globe from the original 1° x 1° grid. The results were compared with inverse modelling estimates for these zones. The sources included: soils under natural vegetation (4.3-4.5 Tg N_2O-N y^{-1}), grassland soils (1.4-1.5 Tg), cultivated soils (1.8-1.9 Tg), direct emissions from savanna

burning and deforestation (0.1 Tg), post burn effects on N_2O emissions caused by deforestation (0.4 Tg), agricultural waste burning (0.1 Tg), emissions from animal excreta (1 Tg), commercial energy use (0.2 Tg), fuelwood combustion (0.1 Tg), adipic acid production (0.2-0.4 Tg), nitric acid production (0.2 Tg), and oceans (2.0-3.8 Tg).
The calculated ratio northern hemispheric : southern hemispheric emissions of 1.5-1.8 is consistent with results obtained in other studies. The comparison with results of inverse modelling techniques showed that an estimation of 1% of N_2O loss induced by synthetic fertilizer inputs is probably more appropriate than the lower N_2O loss rates assumed by IPCC (1991). The results of this study also show that animal waste and tropical land clearing are significant N_2O sources. The calculation of the effect of land clearing accounted for generally observed gradually declining N_2O fluxes along with the age of the clearing, resulting in a lower estimate than that of IPCC (1990). A higher global oceanic emission than the 2 Tg N_2O-N y^{-1} assumed by IPCC (1990) is probable, with high contributions from the Antarctic ocean and the 0°-30°N tropical zone. The latter conclusion is supported by oceanic measurements of project BAQ1 and literature.

4.4 Integration of results

Fossil sources

Traffic (FT). The 1990 emission may be 3.4 Gg N_2O-N y^{-1}, based on the emission factors in Table 4.2 and statistics on Dutch transportation activity levels, (RIVM, 1993). An increase for this source to 9.7 Gg N_2O-N y^{-1} was projected as a result of increasing mobility and the further penetration of catalyst-equipped vehicles. It is difficult to determine the uncertainty of this estimate, since the published emission factors in literature vary by more than a factor 10. However, it is obvious that the current emission is close to the high end of the range given in the preparatory inventory by Van Den Born et al. (1991) (Table 4.1), and that this source will increase in the future.

Combustion (FC). The N_2O emission from fossil fuel combustion may amount to only 0.1-0.2 Gg N_2O-N y^{-1}, based on the emission factors presented in Section 4.3. The emission from waste incineration is an insignificant amount of less than 0.05 Gg N_2O-N y^{-1}. This confirms recent publications (Dlugokencky et al.) showing that fossil fuel combustion is an insignificant source of N_2O. These estimates are lower than those made in the preparatory inventory of Dutch emissions (Table 4.1).

Biogenic sources

Aquatic systems (BAQ). The N_2O fluxes of 1.1-3.1 kg N_2O-N ha^{-1} y^{-1} observed in the Somali Basin and in the Arabian Sea are much higher than the mean global oceanic flux of 0.04-0.2 kg N_2O-N ha^{-1} y^{-1} (Butler et al., 1992). This indicates the importance of these waters for regional emissions. This conclusion is supported by the global inventory (see below). The North Sea, excluding the coastal regions, may be a net source of N_2O of about the same magnitude as the global mean. Although fluxes have not been calculated yet, the emissions observed in the Scheldt river and estuary must be higher than those in the North Sea, as the N_2O concentration increased one order of magnitude from the mouth of the estuary upstream towards

Antwerp. Here a close correlation has been found between the N-loading and N_2O fluxes.

Extrapolation of the emission rate of 1 kg N_2O-N ha^{-1} y^{-1} to the area of 250000 ha of the Dutch Waddenzee yields a figure of 0.25 Gg N y^{-1}. The observed emission rate is much higher than the 0.0042 kg N_2O-N ha^{-1} y^{-1} from earlier measurements in intertidal sediments in the Western part of the Waddenzee (Kieskamp et al., 1991). It is not clear what causes this difference. The gradient method applied by TNO is an actual measurement of fluxes, while Kieskamp et al. (1991) calculated the flux on the basis of N_2O concentration in the surface water. Assuming that the gradient technique yields the actual flux, further work on this subject may yield insight in the relation between the total water soluble N-concentration in the water and N_2O fluxes.

Sewage treatment (BST). The total Dutch emission from sewage treatment plants may be 0.33 Gg N_2O-N y^{-1}, based on a total of 80 Gg y^{-1} of N-influent to sewage water treatment plants in The Netherlands and a N_2O-N loss rate of 0.4% of the N-influent derived from the literature. The exploratory measurements indicate a much lower N_2O emission of 0.01%. The number of measurements was, however, too limited to use this number for extrapolation. They indicate, however, that the estimate based on literature data is possibly an overestimation, and that sewage water treatment plants are of relatively minor importance (Table 4.1).

Grasslands (BGR). It is interesting to note that field measurements for the peat soils at Zegveld showed highest N_2O losses from plots with deep ground water, while in rhizotron experiments with sand the highest fluxes observed were found for high ground water. This is clearly a result of differences in soil conditions. With low ground water the organic soil material quickly decomposes and huge amounts of N are mineralized. The high microbial activity, relatively poorly drained soils and high available N, created conditions favourable for nitrification and denitrification. In the well aerated sand in the rhizotrons of project BGR2 a high water table also created anaerobic conditions, causing a significant increase in N_2O flux relative to the rhizotrons with low ground water.

The model development in BGR2 was not planned in this phase of NRP, while the model of project BGR4 is not yet completed. First conclusions on the basis of the integrated grassland project are that the N_2O emission from peat soils under grasslands are important in The Netherlands. The measurements are of the same order or even higher as the preparatory estimates by Van Den Born et al. (1991). Another important finding is that urine spots have a long lasting effect on N_2O fluxes and need to be included in field measurements of grasslands. However, it is not yet possible to reduce the uncertainty of the estimate (Table 4.1) and to extrapolate the results (see Section 4.5).
A management scheme for good agricultural practice to minimise N_2O emission should restrict periods with increased mineral N contents in the soil. Possible measures include type of N-fertilizer, timing of application, reduction of the N-input according to the needs of the crop or grass, use of nitrification inhibitors, soil drainage, reduction of the period of grazing and increase of N-use efficiency by animals to achieve a reduction of N-excretion.

Global inventory. The tentative estimates for the various N_2O sources confirm that at the global scale soils are dominant sources of N_2O, seconded by the oceans. Agricultural activities may be responsible for the major part of the observed atmospheric increase. There are many uncertainties to be resolved. Major uncertainties are in the estimates of N_2O emissions from tropical soils and the role of legumes in natural ecosystems and leguminous crops (covering about 10% of the world's cultivated area), N_2O emissions from oceans, inland waters, coastal waters, and continental shelves, and industrial sources. Estimation of these sources is hampered by a lack of measurement data. The global amount of N-fixation by leguminous crops may be of the same order of magnitude as synthetic nitrogen fertilizer use and N from animal excreta. Aquatic systems may be very important sources of N_2O that are increasing due to ever increasing anthropogenic N-inputs in the environment, such as leaching of N from agricultural lands and human waste.

The N_2O loss of 1% of the N-input used for N-fertilizers and animal waste seems to be a reasonable assumption, and is in agreement with the results obtained in the integrated grassland project for sand and clay soils. This suggests that fertilized fields are major sources of N_2O at the global scale, and this offers possibilities for reducing emissions in order to control the atmospheric increase. The tentative estimates of the N_2O from animal excreta and those caused by deforestation show that these sources may be significant.

4.5 Future research needs

Integration and validation

Most research in the causes of climate change of the NRP-phase I involved improvement and reduction of uncertainty of source estimates, and identification of sources. Extrapolation and validation of results were not major research topics. Extrapolation to the regional or national scale require a GIS approach, based on measurement results and, particularly for the biogenic CH_4 and N_2O sources, process and management models. These models were not planned in phase I of NRP, or are not yet operational. The geographic information for extrapolation is available in various locations in electronic form, but not as one complete set of data with common scale and format. A general research need that supports source and sink measurements is, therefore, the development of a database including all basic information required for flux extrapolations.

Validation of source and sink estimates can only be done at regional or national scales, i.e. scales larger than point source scale or field scale. The emission maps produced by extrapolation and emission inventories for sources outside the Netherlands and sources not investigated, would form the basis for such validation efforts.

Specific research needed for N_2O

Fossil sources. Combustion is an insignificant source of N_2O. However, transportation probably contributes significantly to Dutch emissions, and may be an important global source. With further penetration of catalyst-equipped vehicles and with ageing of existing catalysts, this source may show a rapid growth. However, the uncertainties of the emission factors are very high. Measurements

under real conditions of N_2O emission from vehicles with and without catalyst are required to verify and improve the estimates presented in Table 4.2. In addition, this research would have explore ways to prevent these N_2O emissions.

Aquatic systems. The preliminary measurements in the Dutch Waddenzee, the North Sea and the Indian Ocean, and the global inventory, indicate that the knowledge on fluxes from aquatic systems is still inadequate to produce reliable estimates. However, N_2O fluxes from the Waddenzee may be of the same order of magnitude as those from heavily fertilized agricultural lands. These relative high fluxes are probably caused by human activities. Part of the NO_3- and other N-compounds that are lost from soils by leaching finally end in surface waters. Further inputs of N, and of organic C, to aquatic systems include human waste, effluent from sewage water treatment plants and industrial waste. This indicates that research is required into the relation between organic C- and N- input into in rivers, coastal systems and estuaries, and CH_4 and N_2O fluxes.

Sewage treatment. The exploratory measurements indicated that sewage treatment may have only a minor contribution to the N_2O emission from The Netherlands, and that possible technical measures to reduce emissions are relatively well known. Further study into this source is not an urgent research priority.

Grasslands. In the first phase of NRP an attempt was made to refine the estimates of CO_2, CH_4 and N_2O fluxes from grasslands as a function of soil properties and physical/chemical factors in the soil, fertilization, grazing, and soil and water management. Part of the modelling work needs to be completed and continued in order to make better extrapolations of the results. This is needed in order to investigate the effect of land use transformations on fluxes of greenhouse gases. In Europe important changes in land use are envisaged, particularly affecting arable lands and grasslands. Conversion of grasslands to arable lands, as well as rotations with 7 years of grassland followed by some years of maize or fodder beets is increasingly applied to obtain "virgin" soils. Such conversions may have effects on fluxes of CO_2, CH_4 and N_2O. Fluxes of one gas may be reduced while fluxes of another gas may increase at the same time. Because of the complex interrelations between controlling factors of CO_2, CH_4 and N_2O production and consumption, future research on trace gas exchange between grasslands and the atmosphere can best be done as a combined research effort. Extrapolation of the results for grasslands requires models in combination with a GIS with information on soils, ground water tables, land use, N-fertilizer inputs, animal densities, N-excretion by animals, length of grazing periods, etc. Currently these data are not readily available for such calculations.

In addition there are a number of research topics specific for N_2O emission from grasslands that need research and policy attention, including:

- the effect of techniques to reduce ammonia volatilisation on dairy farms on emission of other compounds; there are indications that reduction of ammonia losses from animal waste may induce increases in N_2O and CH_4 emission;
- nitrate leached from soils may be denitrified in subsurface environments, producing N_2O. Nitrate may also enter surface waters, as discussed below.

Losses of N_2O dissolved in percolating water may be lost to the atmosphere by degassing from e.g. ditches and canals. There is great uncertainty regarding the magnitude at the global scale, but available publications show that this is a potentially important global source. Considering the current excessive inputs of N in Dutch agriculture, this may be very important at the national scale.

Sources not investigated in NRP I

As indicated in 4.1 a number of potentially important sources were not included in the N_2O cluster. These include arable lands, forests and industrial processes.

Arable lands and forested lands. The N_2O cluster of phase I of NRP did not include detailed studies of N_2O losses from arable land. Significant N_2O losses were measured from the mineral soils in the integrated grassland project. In the Netherlands there are no quantitative estimates and good field measurements for N_2O losses from arable lands, including maize land with excessive N-inputs from animal waste, and horticulture.

Recent German research showed that acid forest soil that receive N-inputs from deposition may show emissions in the order of 5-6 kg N_2O-N ha^{-1} y^{-1} (Brumme et al., 1992). Dutch forests receive even higher N-inputs from deposition, and many forest soils are in the process of acidification. Therefore, arable lands and forests may be much more important than suggested by the inventory in the preparatory study.

Industrial sources. IPCC (1992) mentioned two industrial N_2O sources, i.e. production of adipic acid and production of nitric acid, that had not been identified when the preparatory inventory was made. Experts expect that all industrial and chemical processes in which nitrogen oxidation steps in overall reducing conditions are involved, are potential sources of N_2O (Olivier, 1993). For these sources no estimates have been reported in the literature. The Netherlands is an industrialized country with many chemical industries. An inventory of these industries coupled with collection of data on N_2O losses and additional measurements is required to improve Dutch estimates and to develop techniques and policies to reduce these sources.

Global inventory

There are a number of major knowledge gaps in the global N_2O budget. The major topics where research is required include:

- Emissions from soils under natural vegetation and from the world oceans. In particular, there is uncertainty in the emissions from the tropical zones. Furthermore, the contribution of episodic emissions from temperate soils in winter, early spring and autumn is poorly known.
- The contribution to global N_2O fluxes of N-inputs by N-fixing leguminous crops. The global amount of N-fixation by these crops is of the same order of magnitude as mineral N-fertilizer use and N from animal excreta. Considering that a part of this nitrogen ends in soils, this may be an important global N_2O source.

- Part of the N-losses from various sources to ground water and aquifers and via aquifers to surface waters may finally end via rivers in estuaries, coastal waters, continental shelves and oceans. As shown for the Waddenzee, these aquatic systems may be a very important and very local sources. At the global scale knowledge about the amount of nitrogen transported this way is inadequate. Research involving an inventory of global N-transport though aquatic systems will yield the basic information required to extrapolate models of the relation between N-loading in the water and N_2O emission.
- Quantification of N_2O emission from industrial processes.

5. EMISSION DATABASE DEVELOPMENT

5.1 World Inventory of Soil Emission potentials (WISE)

Methods

The aim of this project (project 851039; ISRIC) was to make a global gridded database of the major soil factors that play a role in the production and consumption of greenhouse gases, such as CO_2, CH_4, N_2O and NO, and to apply this database to assess CH_4 emissions from rice cultivation. The project consisted of two phases. In the first phase an inventory based on available literature was made of the soil parameters and processes that determine the production and consumption of CO_2, CH_4, N_2O, and of existing models to describe these processes. On the basis of this inventory a list of soil factors was made that should be included in a global soil database. A workshop was organized with soil and rice experts participants from all over the world. The development of the database in the second phase was based on the $1/2^\circ$ x $1/2^\circ$ version of the FAO/Unesco soil map of the world prepared by FAO-AGL in collaboration with WISE staff. The soil type information was coupled with soil information from representative profiles. In total 3000 detailed soil descriptions from all continents were included with analytical data. Secondary soil characteristics required for soil models will be derived from climate, parent material and land use.

A global inventory of the potential for CH_4 production of rice soils will be made. In collaboration with the International Rice Research Institute (IRRI) and Wageningen Agricultural University, Department of Soil Science and Geology, a model for describing CH_4 emission from wet rice fields will be developed. This model will be adapted to make use of the data available in the soil database. If this model proves a useful tool, it will be applied to make a global estimate of CH_4 emission from rice cultivation and natural wetlands.

Results

The software to import soil profile information into the WISE database is operational. About 3000 profiles have been imported and validated. The final version of WISE will include information for 3500-4500 profiles. A $1/2^\circ$ x $1/2^\circ$ resolution version of the soil map of the world is being linked to the soil profile data. A start has been made with the development of algorithms to derive soil data for modelling purposes. Work on the development of simple models in co-operation with IRRI and Wageningen Agricultural University is ongoing.

There have been unforeseen problems in this project that have caused delays:
- the response of national soil survey institutes to make available representative soil profiles was disappointing;
- the amount of time needed for proper checks of the imported data was underestimated;
- the preparation by FAO of the $^1/_2^\circ$ x $^1/_2^\circ$ database of soil types has been proceeding more slowly than expected;
- development of the models to calculate CH_4 emission from wet rice fields has revealed the complexity of the relationships between the process-regulating factors in wet rice fields, i.e. soils, land use, climate and hydrology.

5.2 Emission Database for Global Atmospheric Research (EDGAR)

Methods
This project (RIVM projectno. 851060) involved the development of a global emissions database with base year 1990 with 1°x 1° spatial resolution and altitude resolution of 1 km for the following compounds: CO_2, CH_4, N_2O, CO, NO_x, non-methane VOC, SO_x, of NH_3, and ozone depleting compounds (halocarbons) from all known sources. The database development consisted of information analysis, system design and software development by means of prototyping.

Major processes described in EDGAR are land use, energy consumption, other industrial production and consumption, and waste handling. The emissions from processes are estimated from so-called activity levels and emission factors. Activity levels comprise demographic data, social and economic factors and land use and vegetation distributions. Country emissions are distributed on the basis of the co-ordinates of major point sources or by using so-called allocation functions (maps on grid, such as human population, animal populations and energy consumption).
As far as possible statistical data are from internationally accepted sources to ensure comparability and efficient updating. For emission factors, representativity and availability of data as well as compliance with the Global Emission Inventories Activity (GEIA) (project included in the International Global Atmospheric Chemistry Programme, IGAC), OECD and European emission database systems are important criteria. To guarantee the quality of the data, checks on consistency, anomalies and completeness are performed on the result of the final data processing.

A number of inventories produced by GEIA and other institutes are included in EDGAR. In the framework of GEIA TNO and RIVM co-ordinate the work on global biogenic sources of N_2O and NO_x, and inventories of all sources of NH_3 and VOC emissions.

Results
The first version of EDGAR containing activity levels and emission factors for the above sources and compounds will be completed in December 1994. A preliminary version will be made available in summer 1994. The first version of EDGAR allows for:

- definition of source categories, data sets for variables connected to each category, including allocation functions for distributing emissions to the 1°x 1° resolution, and references for all data included in the database;
- importing, inspection by value and visual checks of activity levels, either by country or on grid, emission factors by country of region, and allocation functions to distribute national emissions to grid cells;
- calculation of emissions on grid and by region of selected source categories and inspection of results through maps (on grid and by country) and tables (by region or country);
- exporting of emission data in a specified format. Although for users only emission estimates for source categories will be made available, which is sufficient to meet the user requirements as expressed during the EDGAR user workshop, a full data source description of the underlying data is available for all data included in EDGAR.

Additional EDGAR results are contributions to:
- GEIA: inventories completed so far of anthropogenic and biogenic sources of N_2O, and of VOC sources.
- IPCC National Greenhouse Gas Inventory programme: a default calculation scheme and default emission factors for CH_4 and N_2O from fuel combustion and industrial processes.
- IMAGE 2.0: aggregated regionally calibrated emission factors.
  For some sources, such as oceans, volcanoes and lightning, the emissions for some compounds cannot be generated in this version of EDGAR caused by lack of reliable data.

5.3 Future research needs with respect to database development

The development of the WISE database and EDGAR has taken place in NRP-phase I, but there has been very limited opportunity for developing applications of the products. For the WISE soils and derived databases, a number of possible research activities are apparent:
- Distributions of soil characteristics and derived properties, including soil texture, soil carbon and nitrogen, soil reaction, soil fertility, presented as $^1/_2°$ x $^1/_2°$ resolution databases, form an important improvement compared to the generally used 1°x 1° soil type information in e.g. the model of the terrestrial carbon cycle. In addition, the derived data form important inputs in dynamic assessments of natural emissions of CH_4 and N_2O.
- The inventory of soil properties important in CH_4 production in wetland rice soils and natural wetlands could be used in attempts to develop temporal emission distributions, and in extrapolations of emissions as done in the EDGAR project.

Possible applications of the EDGAR database are related to validation of the emission inventories. The first version of EDGAR is based on measurements and extrapolated with models or with so-called allocation functions. Each global inventory has its specific uncertainty, related to either the measurements or the methods of extrapolation. Uncertainty analysis and validation can only be done by using atmospheric chemistry and transport models. Such assessments have to be done on the basis of temporal distributions, since atmospheric models all have

their specific requirements in terms of temporal distribution. Hence, the production of temporal distributions of emissions and uncertainty analysis are important research needs related to validation.
Furthermore, there are a number of major omissions in the EDGAR database, such as emissions of ammonia, aerosols and compounds involved in aerosol formation (sulphate and DMS).

6. SOCIO-ECONOMIC CAUSES

6.1 Methods

The objective of the project (project 850019; RIVM) was to develop and apply methodologies to determine national inventories of greenhouse gas emissions. The project focused on methane and nitrous oxide, because least was known about emissions of these greenhouse gases in the Netherlands. In three phases, three reports were planned: a first inventory of Dutch emissions (see Section 4.1), and two subsequent detailed overview reports on the state-of-the-art of knowledge about emissions of methane and nitrous oxide. A secondary objective was the identification of knowledge gaps, which could be used by the NRP for programming experimental emissions research. The main research method was literature survey and discussions with experts. Initially emission estimates were primarily based on international literature, later experimental data from Dutch research could increasingly be used. The focus of the project shifted rapidly from social causes to methodologies for national emissions estimates as a contribution to the IPCC methodology development. One of the contributions consisted of the organization of an international workshop on CH_4 and N_2O.

6.2 Results

The first result was an inventory of the current and an estimate of future Dutch emissions for all greenhouse gases. This inventory was used as a basis for the development of greenhouse gas policies in the 2nd National Environmental Policy Plan and for setting priorities in selecting experimental projects for the NRP. The experience led to active RIVM-participation in the IPCC-OECD Programme on Developing Guidelines for National Emissions Inventories. A background study on methane updated the previous estimate of national methane emissions, partly based on new experimental data. It was found that methane emissions in the Netherlands would probably decrease by more than 10% in 2000 relative to current levels as a result of policies in the area of waste and manure control. Manure related emissions were identified as an important "new" and growing source. Specific policies were identified that could further decrease national emissions.
Currently, a similar report on nitrous oxide is being finalised. In this report, an important discrepancy is identified between the methodology proposed by IPCC and a more detailed method, developed in this project. The main reasons for this discrepancy are differences in emission factors and the omission of a number of significant sources in the IPCC methodology.
Future Dutch emissions of N_2O are believed to increase, while increasing emissions due to the introduction of car catalysts are not fully compensated by decreasing emissions due to limitation of nitrogen deposition (atmospheric and through manure and fertilizer).

6.3 Future research needs

There are two major research topics which merit research attention. First, the background reports on CH_4 and N_2O show that the IPCC methodology for national emission inventories needs to be complemented with methods to derive estimates for a number of identified but unquantified anthropogenic sources. Secondly, the IPCC method leaves a great liberty in using emission factors. The national or regional results need to be compared with estimates based on global methods, such as in EDGAR, or with results of validation efforts with atmospheric models. On the basis of such comparisons the proposed national methods can be revised.

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