GLOBAL CHANGE

Dutch National Research Programme on Global Air Pollution and Climate Change

EVALUATION AND VALIDATION OF METHANE EMISSIONS IN THE NETHERLANDS AND THE CONTRIBUTION FROM VARIOUS SOURCES

Report no.: 410 100 040 (1996)

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Evaluation and validation of methane emissions in The Netherlands and the contribution from various sources

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Nationaal Onderzoek Programma Mondiale Luchtverontreiniging en Klimaatverandering (NOP)

Het Nationaal Onderzoek Programma Mondiale Luchtverontreiniging en Klimaatverandering (NOP) bevindt zich thans in de tweede fase (1995 - 2001). De eerste fase, waarin 150 projecten zijn uitgevoerd, liep van 1990 tot 1995. Naar verwachting zullen in de tweede fase uiteindelijk circa 80 projecten worden uitgevoerd. Gezien de aard van het klimaatprobleem is een multi-disciplinaire benadering binnen het NOP noodzakelijk. Het programma is onderverdeeld in vier thema's:
I Gedrag van het klimaatsysteem als geheel en in onderdelen
II Kwetsbaarheid van natuurlijke en maatschappelijke systemen voor klimaatverandering
III Maatschappelijke oorzaken en oplossingen
IV Integratie en assessment

Het primaire doel van het NOP, als strategisch en lange termijn onderzoekprogramma, is te voorzien in de behoefte aan beleidsrelevante informatie voor de ontwikkeling van het nationale en internationale klimaatbeleid. Naast het bereiken van dit inhoudelijke doel, wordt er ook veel belang aan gehecht dat het onderzoek op de langere termijn verankerd zal blijven in de Nederlandse onderzoeksstructuur.

Door het NOP wordt twee maandelijks de (gratis) onderzoeksnieuwsbrief "CHANGE" uitgegeven. Voor meer informatie over het NOP kunt u zich richten tot:
Programmabureau NOP, Postbus 1 (pb 59), 3729 BA, Bilthoven
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National Research Programme on Global Air Pollution and Climate Change (NRP)

The National Research Programme on Global Air Pollution and Climate Change (NRP) is currently in its second phase, 1995-2001. The first phase, in which 150 projects were carried out, ran from 1990 to 1995. About 80 projects are expected to be finally realised in the second phase.
The nature of the climate problem warrants a multi-disciplinary approach within the NRP. The programme is categorised into four themes:
I Dynamics of the climate system and its component parts
II Vulnerability of natural and societal systems to climate change
III Societal causes and solutions
IV Integration and assessment

The primary objective of the NRP as a strategic and long-term research programme is to meet the demand for policy-relevant information for the development of national and international climate policy. Besides realising this substantive objective, a great deal of importance is attached to the long-term anchoring of the research within the Dutch research structure.

The NRP Programme Office publishes a (free) research newsletter called CHANGE every two months. For more information on the NRP please contact:
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Summary

Within the National Research Programme on Global Air Pollution and Climate Change several research projects have been carried out which have investigated the methane emissions of various source categories. Together with an inventory of sources these investigations provide the basis for an account of the total Dutch methane emissions.
In order to validate and integrate the results of these subprojects on emissions additional studies were initiated in the integrated project cluster: "Validation, integration and scaling of methane emissions from various sources". Projects within this latter cluster aimed at studying methane emissions in the Netherlands at a higher aggregation level to provide a more general validation of the total Dutch emissions and the uncertainty therein.

The research strategy adopted for the study described in this report was to collect data on ambient concentrations of methane and to compare these data by atmospheric modelling with present estimates of Dutch emissions from emission inventories.

To this end hourly concentrations of methane have been measured at three monitoring sites during a period of one year. Additional mobile measurements, though on a smaller scale, have been carried out to supplement the information from the stationary monitoring points.
A detailed analysis of the concentration data has provided the source contributions of emissions in the Netherlands to ambient concentrations.
By atmospheric modelling, using a box model, emissions have been derived from these source contributions and compared with the results of recent emission inventories.

Ground level concentrations of methane at all three monitoring sites generally range from 1.8, (the northern hemisphere background concentration) to 2.5 ppm, with an average close to 2.0 ppm.
The variation is mainly governed by dispersion conditions and wind direction.
The variation with wind direction reflects the presence of upwind source areas.
Largest source contributions (0.5 ppm) were found for continental air masses originating from Central Europe, and in decreasing order in air masses arriving from the northern, southern and most western (coastal) parts of the European continent and in air masses from marine origin (0.1 ppm).
Contributions from Dutch sources were relatively small, between 0.04 and 0.1 ppm.

By atmospheric modelling these source contributions from Dutch emissions were converted to emission densities, which ranged from 10 to 43 $g/m^2.yr$ for various parts of the Netherlands.

The overall average emission rate amounts to 27 g/m^2.yr, which leads to a total emission in the Netherlands of 1.080 kton per year. The uncertainty of this total emission is estimated to be +/- 30%. This total emission is very similar to the total of 1.130 kton per year, derived (in a second NRP project within the same cluster) from trajectory analysis of one year methane measurements in the same period at the 200 m high meteorological tower at Cabouw.
The total emission estimates derived from concentration measurements and atmospheric modelling compare favourably with the results of two recent emission inventories of 760 to 1.230 kton per year, with a central value of 1.230 kton per year (1991) and 1.090 kton per year (1993).

The agreement between the central values of the total emission estimates from ambient concentration measurements combined with atmospheric modelling on one hand and emission inventories on the other hand provides firm confidence in the present total emission estimate.
However, there is still a wide range of uncertainty in the derived totals.
Further research will be required to narrow this range. The strategy of combined ambient monitoring and atmospheric modelling has shown to be a very valuable tool.
Improvement of the data base of ground level concentrations, advanced data processing and improved modelling techniques will be able to provide more detailed insight in the emissions and thus narrow the uncertainty in their estimates.

1. Introduction

Theme B "Causes" of the Dutch National Research Programme Global Air Pollution and Climate Change (NRP) includes studies on the cycle of greenhouse gases but also on anthropogenic activities contributing to changes in atmospheric concentrations. The objective of these studies 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. To structure the programme a first order analysis of the uncertainty in cycles of carbon dioxide (CO_2), methane (CH_4) and nitrous oxide (N_2O) was made. For methane as well as carbon dioxide it was concluded that more knowledge on the process level was clearly needed. It was also concluded that the process of integrating the results of local studies to regional and global scale introduced large uncertainties. For N_2O the situation was even worse; information on important mechanisms was not really available.
On the basis of this information it was decided that coherent programmes were to be formulated in which, on one hand, typically Dutch aspects of the greenhouse gas cycles would be studied and on the other hand the research would contribute to international programmes as well.
Based on the aforementioned considerations three clusters of projects were designed (Berdowski et al., 1995):

CO_2-cluster
Study of CO_2 exchange between grasslands and the atmosphere, and development of a methodology to validate CO_2 exchange models for larger areas.

N_2O-cluster
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.

CH_4-cluster
Investigation of CH_4 emissions of selected sources, thought to contribute significantly and with a large uncertainty, and development of a methodology to validate CH_4 exchange models for large areas.

Within the framework of the CH_4 cluster several projects were formulated in which important sources in the Netherlands are investigated. These include studies on emissions from landfills, oil and gas extraction, gas distribution and emissions from biogenic sources. These investigations coupled with an emission inventory provide an account of the total Dutch CH_4 emissions. However, because of the limited number of actual sample surveys of sources, and limited knowledge of the variability of source strengths within each source category and emissions by unknown sources the uncertainty in the estimates is considerable.

In order to obtain a more accurate estimate of total magnitude of Dutch CH_4 sources and the uncertainty therein a project "Evaluation and validation" was formulated. In this project three sub-projects were formulated:

1 Validation of sources strengths of atmospheric CH_4 using carbon isotope ratios. Project 852097 carried out by The Netherlands Energy Research Foundation ECN.
2 Methane emission of the Amsterdam urban area. Project 853125 carried out by Wageningen Agricultural University.
3 Evaluation and validation of methane emissions in the Netherlands and contributions from various sources" carried out by the Netherlands Organisation for Applied Scientific Research (TNO) and KEMA Environmental Services (project no. 853124).

In this report the results of the third project carried out by KEMA and TNO are reported. The aim of this project was to estimate Dutch emissions using measurements of CH_4 concentrations in air at several locations in the Netherlands. The results of these measurements are interpreted and generalized using a simple box model. This approach provides an independent estimate of Dutch sources and at the same time it contributes to validation of estimates based upon research carried out in the other subprojects. In the sequel the results of the measurements are reported and discussed.

2. Project methodology

2.1 Estimates of methane emissions

In 1990 IPCC estimates were given for the most important global sources and sinks of methane. Wetlands and rice paddies are among the most important sources whereas atmospheric oxidation by OH radicals is the most important sink. It was concluded that although the uncertainties were significant the estimates were fairly well established. In a later stage the uncertainties in these estimates were criticized (Slanina et al., 1994). It was stressed that the uncertainties were underestimated. It was also noted that extrapolations of measurements on a local scale to regional, continental and global scale is probably one of the main sources of errors and uncertainties.

A first estimate of Dutch CH_4 emissions was made by Van den Born et all. (in: van Amstel,et all., 1993) . The major conclusions of this work including an estimate of the uncertainty are presented in Table 1.

Table 1: *Estimated emissions of methane in The Netherlands (in kton CH_4 per annum) (van Amstel et al., 1993)*

	Van den Born et al. '88-'89			RIVM 1990			RIVM 1991		
	Low	Median	High	Low	Median	High	Low	Median	High
Gasproduction	40	55	70	38	52	67	43	60	76
Gastransport	2	7	12	2	6	12	2	7	12
Gasdistribution	60	70	80	61	72	83	71	84	97
Oil production	0	18	35	1	19	37	1	17	34
Combustion[1]	22	27	32	14	21	28	16	23	31
Wetlands	40	70	120	40	70	120	40	70	120
Inland and coastal waters	24	35	60	24	35	60	24	35	60
Small water bodies	11	17	23	8	16	24	8	16	24
Ruminants	295	379	462	290	414	538	297	424	551
Animal waste	10	15	20	74	106	138	76	109	142
Landfills	190	255	320	178	377	576	178	377	576
Water purification	3	3	3	3	3	3	3	3	3
Drinking water production	2	2	2	2	2	2	2	2	2

[1] combustion of gas and oil by (mainly small) end users.

This inventory showed that enteric fermentation, landfills, the oil and gas industry and organic soils are responsible for 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. Since 1991 new information on methane emissions has become available and new guidelines to estimate anthropogenic methane emissions were drafted by IPCC.
Based on these new guidelines new estimates were made for the Netherlands by RIVM. These estimates are shown in table 1 as well. The estimates for landfills and

animal manure have increased considerably while others did not change significantly. From table 1 it can be concluded that uncertainties are large even on a national scale.
It is important to note that these estimates are all extrapolations of a limited number of experiments carried out on a small scale. Larger scale evaluations have hardly been carried out. Therefore independent evaluation of these estimates on the basis of measurements could contribute considerably to the aimed reduction of the uncertainty in Dutch emissions.

2.2 Research method

According to recent measurements (Simmonds et all., 1991, 1993; Bosinger et all., 1993) northern hemisphere background concentrations in air are slightly below 1.8 ppm. When emissions from natural and anthropogenic sources add to this background concentrations, yearly average concentrations up to or even in excess of 2.0 ppm are observed, depending on the upwind source areas contributing to the average concentration. Thus measured ambient concentrations provide information on the source contributions to the methane concentrations, which relate by atmospheric dispersion to emissions of methane.
The research strategy adopted for this study was to collect data on ambient air concentrations of methane and compare these data by atmospheric modelling with present estimates of Dutch emissions from emission inventories.
To this end the activities described below were planned and carried out.

Measurement at fixed monitoring sites.
Hourly concentrations of methane in ambient air have been measured continuously over a period of more than one year (oct. 1993 to jan. 1995) at three monitoring sites: Arnhem, Kollumerwaard (KEMA) and Delft (TNO). By their geographical location these monitoring sites provided quantitative data on Dutch source contributions and in addition on transboundary contributions.

Mobile measurements.
Mobile measurements were planned to supplement the information from the fixed monitoring sites.
Only a limited amount of such measurements were carried out, which mainly focused on the Groningen gas production area, which was suggested by occasional high concentrations measured at the monitoring site Kollumerwaard.
Airborne measurements were also planned but due to the inaccessibility of flight facilities this activity was, after consultation with the NRP management, deleted from the programme.

Diverse activities.
Since within the project cluster: "Evaluation and Validation" ambient concentration measurements were carried out by various research institutes (ECN, KEMA, LUW

and TNO), calibration cross checks were organized between all parties concerned, to establish traceability of all monitoring results to a common reference.

Interpretation of concentration measurements.
A detailed analyses has been made of the combined data from the three fixed monitoring sites, by selecting and grouping concentration data according to meteorological dispersion variables (wind speed and direction, mixing height) and comparison of data from the monitoring sites under selected conditions.
This analysis finally resulted in the derivation of the contributions from Dutch sources to the concentrations.

Atmospheric modelling.
The aim of the modelling exercise is to couple the measured concentrations, or more precise the measured contributions to the concentrations in excess over the northern hemisphere background concentration to emissions.
There are basically two ways to achieve this goal. With the present knowledge on emissions as an input to the model, the calculated and measured contributions to the background concentrations can be compared to verify the correctness of the emission input. This route however requires a detailed (source strengths and locations) emission data base, which was not available.
Therefore the alternative route was used of calculating the emissions from the source contributions obtained experimentally and to compare the total emission thus obtained with present estimates from emission inventories.

3. Measurements

3.1 Stationary measurements

Methane concentrations were measured over a period of one year at three fixed monitoring sites. The locations of the monitoring sites are shown in figure 1. The geographical coordinates of the monitoring sites are:

Arnhem	:	51°59' N, 05°52' E
Delft	:	52°00' N, 04°23' E
Kollumerwaard	:	53°20' N, 06°17' E

Short monitoring site descriptions and details of the experimental methods used are given below.

Figure 1: *Monitoring sites for continuous methane measurements: Arnhem (A), Delft (D) and Kollumerwaard (K).*

3.1.1 Arnhem and Kollumerwaard

Kollumerwaard
Station Kollumerwaard is located in an agricultural area. The nearest town is the city of Groningen at a distance of about 25 km in the ESE direction. Kollumerwaard is located in a relatively non polluted, rural part of the Netherlands. At the

Kollumerwaard site measurements are also performed by RIVM and TNO within the ozone research program of the National Research Program.

Since July 1991 ambient air measurements are performed for CO_2, CH_4 and CO. Except for some minor disturbances the instrumentation performed very well during the whole period. Every half year, reports are published concerning the measurements of the greenhouse gases as well as the meteorological measurements (Vosbeek, 1993). These reports give a complete image of the performed measurements, disturbances, calibrations and results.

Sampling takes place using a stainless steel sampling line. The main specifications of the sampling line are given below. The samples for the measurements of the different components are taken from the continuous flow of ambient air through the sampling line. The main specifications of the sampling line are:

- Sampling inlet height: 8.6 m above ground level.
- Inner diameter: 47.8 mm.
- Material of the sampling line: Stainless steel 316.
- The inside surface has been electro-polished.
- The pipes are coupled with Swagelok connections; all connections are weldless.
- Flow: 12 l/s (turbulent flow: Reynolds number 22.000).
- Distance between inlet and sampling devices: 7.5 m.
- Sample velocity in the sampling line: 6.7 m/s.
- Residence time: 1.1 second.

Arnhem

Station Arnhem is located at the KEMA site west of the city of Arnhem. Arnhem is located in a typical urban area.

Since April 1990 ambient air measurements are performed for CO_2, CH_4 and CO. For 1993 the first half year of measurements is missing due to a hard disk crash. In 1994 the months February, March and April are missing due to some major disturbances of the instrumentation. Every year, reports are published concerning the measurements of the greenhouse gases as well as the meteorological measurements (KEMA 1993a, KEMA 1993b, KEMA 1993c, KEMA 1994a, KEMA 1994b). These reports give a complete image of the performed measurements, disturbances, calibrations and results.

From 1990 till May 1991, sampling took place at a height of 1.5 metres above ground level. From May 1991 on, the sampling takes place at about 15 metres above ground level. The main specifications of the sampling line are:

- Sampling inlet height: 15 m above ground level.
- Inner diameter: 2 inch.
- Material of the sampling line: Stainless steel 316.
- The inside surface has been electro-polished.
- The pipes are coupled with Swagelok connections; all connections are weldless.

- Flow: 12 l/s.
- Distance between inlet and driers: 0.5 m.
- Sample velocity in the sampling line:
- Residence time: 1.2 second.

The measurements in Arnhem en Kollumerwaard are both performed with a Chrompack CP 9000 GC. Therefore the measurement method is only described once.

Method
Concentration measurements of the trace gases CH_4, CO_2 and CO in ambient air are performed semi-continuously at 15 minute intervals. Using the multi component capabilities of gas chromatography (GC) as an analytical technique, three real time comparable time series of concentrations are produced. The complete sampling system is schematically shown in figure 2.

Figure 2: *Sampling system.*

A Chrompack CP 9000 GC is operated with three columns in series, packed with Hayesep (1 and 2) and Molsieve (3) respectively. Following sample injection, backflush and heart-cut techniques are performed using two 10 port switching valves. After separation, components are led to a hydrogen-fed Ni catalyst (methanizer) where CO_2 and CO are converted to CH_4. Final detection is done by means of a Flame Ionisation Detector (FID).

Separation of a composite peak (containing N_2, O_2, CO, CH_4, CO_2 and any available higher hydrocarbons) is achieved on the first HayeSep column. When the CO_2 peak has passed the splitting point between first and second HayeSep columns, the first 10 port valve is switched to backflush, thus leading any available Non Methane HydroCarbon (NMHC) to the FID (the backflush channel has its own detector). Further separation of composite peak and CO_2 is achieved on the second HayeSep column and carrier gas flow is directed on to the Molsieve column. The outlet of the Molsieve column is vented to ambient air to prevent oxygen from entering the methanizer and thus disturbing its performance. The second 10 port valve is switched before the CO_2 peak arrives, thus preventing it from entering the Molsieve column where CO_2 would be permanently retained. CO and CH_4 are led through the methanizer to the FID, bypassing the Molsieve column. Total separation time is 14 minutes.

Equipment used

- Columns:

  Two columns each 1.5 meter 1/8 inch filled with HayeSep C 80-100 mesh and one 1.9 meter 1/8 inch filled with Molsieve 13 x 80-100 mesh. Operation isothermal 50 °C.

- Detection:

  Flame ionisation detection on both frontflush and backflush channels. Detector temperature 200 °C. The frontflush channel for CO, CH_4 and CO_2 contains a methanizer (Ni-catalyst) operated at 375 °C to convert both CO and CO_2 to CH_4 prior to detection.

- System gases:

Carrier:	N_2 from bottles, Air Products quality 5.0
FID supply:	H_2 produced by Packard Hydrogen generator
FID supply:	Air, compressed ambient air from Chrompack Air-generator.

- Valves:

  Two pressure actuated 10 port Valco valves (1/16 inch ports) for sample injection and column switching. Fixed sample loop 1 cm^3, all lines 1/16 inch stainless steel.

Sampling

From the 2 inch sample intake air is led through 1/4 inch SS tubing at a flow rate of 5 l/min using critical orifice and a vacuum pump to a dual permapure dryer system.

The dryers (PermaPure type MD-125-12S) have an outside diameter of 1/8 inch and are operated at flow rates of 50 ml/min for both sample and purge lines. Air flows are created by applying pressure drop over stainless steel capillaries. To prevent capillaries from clogging, sample and purge flows are filtered through 2µm SS filter (Nupro). The dew point is decreased to far less than - 25 °C depending on air moisture content. Dried air is drawn permanently at a flow rate of 10 ml/min through the internal piping (1/16 inch SS) of the GC to the injection valve. Shortly before the final injection of 1 ml air the sample flow is cut off to equilibrate sample pressure to ambient pressure level.

Sampling frequency
The sampling frequency is once every 15 minutes.

Main system performance

• Lower detectable limit	CO:	0.05	ppm
	CO_2:	5	ppm
	CH_4:	0.05	ppm
• Measuring range	CO:	0.05 -1.5	ppm
	CO_2:	5 - 800	ppm
	CH_4:	0.05 - 15	ppm
• Precision (std.dev.)	CO:	0.01	ppm
	CO_2:	1.8	ppm
	CH_4:	0.01	ppm
• Analysis time	CO:	15	min
	CO_2:	15	min
	CH_4:	15	min
• Interferences:	none		

3.1.2 Delft

The monitoring station Delft is located at the TNO premises at the east border of the city, along the motor way A13. Delft is situated in the densely populated province of South Holland between Den Haag and Rotterdam at some 10 to 25 km north of the large urban and industrial Rijnmond area including Rotterdam and its harbours with a large concentration of chemical and petrochemical industries.

Sampling
Ambient air is drawn from the roof top of the building, 20 m above ground level, through 10 mm inner diameter pvc tubing. This tubing serves as a primary air intake which is vented with 2 l/min using a membrane pump (Austin, type F85 DE) and critical orifice, resulting in a residence time of about 45 s. The pressure drop

over this length of tubing is less than 0.1 kPa and continuously monitored to assure unobstructed air intake of ambient pressure.
Ambient air is sampled continuously, at a rate of 5 ml/min., from this primary intake through 1/8" stainless steel tubing via a stainless steel three way valve to feed a 2 ml sample loop. The flow rate is adjusted and maintained by a needle valve.
The content of the sample loop is analyzed on line with 15 minutes intervals.
The three way valve is periodically (automated) switched to sample from a certified standard gas mixture to provide daily calibration of the monitoring instrument.

Analysis
Methane is analyzed by an automated gas chromatograph (Air Instruments, model 755) using flame ionization detection (FID).
A back flush system is used to avoid cumulative contamination and overloading of the gas chromatographic column by non-methane hydrocarbons.
The chromatographic system is specified and operated as follows:

Gas Chromatograph:	Air Instruments, model 755
Detector type:	FID
Sample injection:	Ten way valve and sample loop
Sample loop volume:	2 ml
Sample flow rate:	5 ml/min
Column:	Stainless steel, 1/8", 1.5 m
Column packing:	HayeSep Q, 80-100 mesh
Column temperature:	50 °C, isotherm
Detector temperature:	ca. 140 °C
Carrier gas/flow rate:	Helium, 1.00 bar, 10.6 ml/min
Back flush flow rate:	12 ml/min
Flame Hydrogen:	1.6 bar, 33.1 ml/min
Flame Air:	1.4 bar, 250 ml/min

Under these conditions the retention time of methane is 1.5 min.
Total cycle time for the analyses is 7.5 minutes.

3.2 Mobile measurements

A way of gaining additional information about the distribution of methane concentrations in the Netherlands, is by performing a mobile measurement campaign. Furthermore, mobile measurements can play a role in the quantification of incidental emissions. In the preparatory study in 1993 (Vosbeek, 1993a) it was investigated what the boundary conditions are for the mobile measurement campaigns. Besides this, possible measurement equipment was studied. In the preparatory study we concentrated on the Groningen gas production area and this study learned that for a successful mobile measurement campaign foreknowledge of activities in the natural gas production area is essential. Without knowledge of exploration and exploitation activities and, therefore, of possible emissions, even mobile measurements are not able to measure elevated concentrations.

Several attempts have been made to obtain cooperation and thus information from the oil and gas industry. Consultation with the Dutch State Supervision on Mines (Staatstoezicht op de Mijnen (STOdM)) delivered information on drilling activities performed in the period July 1991 till August 1994. This information was however not available when the first mobile measurement campaign started.

3.2.1 Measurement method

The mobile measurements were performed by using a photo acoustic spectrometer of Bruel & Kjaer (Verhage *et all.*, 1992; Vosbeek, 1993a). The accuracy of the spectrometer is not very high (0,1 ppm), but it is possible to measure methane background concentrations (1,8 ppm - 2,0 ppm). The detection limit of the photo acoustic spectrometer is 300 ppb.

As was already mentioned the information obtained from Staatstoezicht op de Mijnen was not available when the first mobile measurement campaign started. Information about the location of natural gas production sites was therefore obtained from the Dutch Topographical Atlas (1987). These locations were consolidated with information of the Dutch Photographic Service in Emmen. This service receives every year a new photographic update of the Netherlands. Several photographs have been studied to consolidate the locations found in the Topographical Atlas. The locations of the production sites were still the same, there was however no time to identify possible new sites (after 1987).

During the campaigns it was agreed upon measuring only production sites with burning flares. The fact that a flare is burning indicates towards production activities, since a flare is often used as a kind of a pilot-flame.

3.2.2 Measurement campaigns

Campaign 28-06-1994
West from Veendam three natural gas production sites are situated. At all three the sites triple measurements were performed, first upwind and then downwind. The meteorological conditions for this day were sunny, with a weak south-east wind (3 m/s). In Veendam two sites had a burning flare, at the third site the flare was out. The measured concentrations at all three the sites did not differ significantly from background concentrations.

In the neighbourhood of Hoogezand at least 15 production sites are situated, however only few sites had burning flares. At two sites measurements were performed and again no differences were measured with respect to background concentrations.

In the surroundings of Midwolde two natural gas production sites are located, but at only one a burning flare was observed. At this location measurements were

performed but again with the same conclusion as at the other sites: no increased concentration levels of methane with respect to background concentrations.

Campaign 19-07-1994
The first measurement campaign learned that in the area of production sites similar CH_4 concentrations are measured as are measured at Kollumerwaard. Thus confirming the homogeneity of the CH_4 concentration in that particular area. Before a new measurement campaign was carried out information was obtained from STOdM concerning the drilling activities performed at that moment in the north of the Netherlands. Very close to the measuring site of Kollumerwaard two drilling activities were executed: in Munnekezijl and in Anjum. At both locations concentration measurements were carried out and the Kollumerwaard site was also measured twice.

Weather conditions were fair: sunny, clear weather at the beginning of the day and in the course of the day somewhat cloudier. Wind came from the north-east (5 m/s). During this campaign methane concentrations were measured similar for the drilling sites and the Kollumerwaard site. No indication of incidental emissions of CH_4 was found.

Campaign 26-07-1994
For the beginning of the third campaign again the Dutch State Supervision on Mines was consulted. The drilling at Anjum and Munnekezijl were still active and at the time a work-over (maintenance activities) took place at 't Zandt. The plan for this campaign was a visit to some of the production sites already visited in the first campaign, then a visit to the drilling sites of the second campaign and finally a visit to the work-over at 't Zandt.

The weather conditions were again sunny, the wind direction in Veendam, Scheemda and 't Zandt was stable south-west, with wind speeds of 3 to 5 m/s. During the measurements at Anjum and Munnekezijl there was almost no wind.

In Kollumerwaard the daily average concentration during this campaign was 1,9 ppm. The production sites as well as the drilling sites as the work over in 't Zandt showed no relevant increased CH_4 concentrations with respect to the concentration measured at Kollumerwaard.

3.3 Quality Assurance and Quality Control

3.3.1 Calibration Arnhem and Kollumerwaard

Calibration for Arnhem and Kollumerwaard is identical. Calibration is integral in a control routine of the long term stability of the chromatographic system. Every

three hours one of four standard gas mixtures each containing CO, CH_4 and CO_2 is measured. The concentrations of the three components in the four standard mixtures cover a concentration range according to expected ambient concentration levels. This control routine daily produces two results for each of the four existing concentration levels per component. These results yield the response function for each component using first order regression. Calculated slopes and intercepts are used to produce concentration values for ambient measurements for the respective day. Absolute concentration levels of the four different gas mixtures are verified twice per year using a comparison method with certified reference materials according to ISO 6143.

The calibration scheme is shown below:

Every 3 hours:	Injection of standard gas mixture (CO, CH_4 and CO_2) to establish long term stability and daily response functions.
Daily:	Control of system operation by viewing status files in PC using modem communication from home lab (manual).
Monthly:	Control and registration of system parameters (e.g. sample flows, vacuum pump performance, H_2-generator status, gas supplies for N_2 and air, detector base line levels, GC-temperatures).
Semiannually:	Validation of concentration levels of control and calibration gas mixtures using NIST reference materials (manual).
	Check of performance of Perma Pure Dryer system using Karl Fischer technique to determine absolute moisture content of incoming and outgoing air.
Occasionally:	Participation in Round Robin tests for gas mixtures.

In addition all non regular phenomena or specific events regarding performance of equipment, meteorological situation, visits of personnel are recorded in log-books.

3.3.2 Calibration Delft

The performance of the monitoring method was evaluated using commercially obtained (Scott Specialty Gases) certified standard gas mixtures of 3.14 and 99.2 ppm methane in air and dilutions of the latter in the range 1 to 10 ppm, prepared by dynamic volumetric methods.
From these the following performance specifications were derived:

Lower detection limit:		0.060 ppm
Non-linearity (0-10 ppm):		< 0.020 ppm
Repeatability stand. dev.	(at 2.0 ppm):	0.016 ppm
	(at 3.0 ppm):	0.020 ppm
	(> 3 ppm):	< 0.5 % (relative)
Reproducibility stand. dev.	(at 3 ppm):	0.025 ppm
Span stability, systematic component:	0.008 ppm / month	
	random component:	0.008 ppm

During these experiments it became clear that the two commercially obtained certified standard gas mixtures did not match (discrepancy ca. 12 %). The results of an intercomparison exercise (ECN, 1994) confirmed the largely erroneous certified value of the "3.14 ppm" standard. Recertification by the supplier of both gas mixtures did wipe out the mismatch only partially. Final analysis by the "Nederlands Meetinstituut (NMI)" confirmed the concentration ratio of the two mixtures as found by our laboratory and gave absolute values for their concentrations which brought our results in the intercalibration in line with the results of other participants. The NMI values were therefore adopted for all further calculations of concentrations.

Given the linear response of the FID, and the insignificant non-zero intercept of the calibration function, a single point calibration was used as a daily routine. The "3.14 ppm" (after correction 2.71 ppm, +/- 2%) standard mixture was used for these daily calibrations. Calibration was performed automatically at intervals of 23 hours, by consecutive duplicate injections of the standard gas mixture.

Duplicate calibrations with larger differences than 0.07 ppm (related to repeatability) were rejected. Further two and four weekly variances of the calibration data were checked against the reproducibility and possible outlying results eliminated. This procedure resulted in only incidental rejection of calibration results. The average values of the calibration data (over two and four weeks) were further regressed against time to reveal any systematic change with time. For a period of six months in which no adjustments were made to the chromatographic system, a minor systematic drift of 0.3 % per month was observed, while the random part of the instability with time was of the same size as the reproducibility of the measurement. From this analyses it was concluded that the raw data could be corrected to concentrations applying a monthly average calibration factor for periods of uninterrupted operation. Changes in instrumental parameters (2 occasions) also required adjustment of the calibration factor.

Including the reproducibility of the ambient air and calibration gas measurements and the (small) systematic part of the instability with time, total accuracy of the data (relative in the time series) is estimated to be about 1.1%. For the accuracy of the absolute values of ambient air concentrations the accuracy of the calibration standard (+/- 2%) should be included. The absolute values of ambient air concentrations are therefore probably accurate to +/- 3 %.

3.4 Intercalibrations

KEMA and TNO have participated in a number of intercalibration exercises. The results are reviewed here in short.

In april 1992 KEMA participated in an international intercomparison organized, within the framework of the TOR project, by Hahn (Hahn, 1993) of the Fraunhofer Institute at Garmisch Partenkirchen.
There were only a few participants in this programme and they reported CH_4 concentrations which differed from the reference concentrations between +0.66 and + 2.31 %. The result reported by KEMA showed a deviation of +2.05 %.

In june 1993 and may 1994 intercalibrations were organized by ECN within the frame work of the Dutch National Research Programme (NOP) on Global Air Pollution and Climate Research. All research institutes carrying out ambient air measurements within the NOP participated:ECN, KEMA, LUW and TNO.
The results are summarized in table 2.

Table ?: *Results of the 1993 and 1994 intercalibrations. All data in ppm. Relative standard deviations (%) in parenthesis.*

	june 1993	may 1994		
		mix 43	mix 44	mix 45
ECN-C	1.848 (0.17)			
ECN-P	1.847 (0.49)	1.973 (0.10)	2.219 (0.23)	2.616 (0.16)
ECN-P	1.844 (0.36)	1.973 (0.15)	2.215 (0.40)	2.611 (0.11)
KEMA-A	1.953 (1.30)	2.018 (0.15)	2.258 (0.27)	2.646 (0.19)
KEMA-K	1.954 (1.59)	2.014 (0.89)	2.290 (0.70)	2.696 (0.41)
LUW	1.989 (0.05)	2.221 (0.09)	2.600 (0.10)	
TNO	1.784 (2.04)	2.016 (0.43)	2.252 (0.38)	2.630 (0.66)
Average	1.872 (3.63)	1.997 (1.08)	2.243 (1.32)	2.633 (1.32)

The results show that the laboratories have improved their repeatability and that comparability between laboratories has also improved. The participants are generally able to produce results within 0.5 % (within laboratory standard deviation) and the between laboratory standard deviation is about 1% for the concentration range of interest.

3.5 Meteorological measurements

3.5.1 Arnhem and Kollumerwaard

Arnhem
From 1990 till 1994 station Arnhem was equipped with sensors for 5 meteorological parameters: wind speed, wind direction, relative humidity, temperature and

global radiation. The site where these measurements took place was about 100 m from the site where the trace gases were measured. Air pressure measurements could not be performed at the Arnhem site and were requested from the KNMI-measurement site Deelen (about 9 km from Arnhem). The main specifications for the meteorological measurements are given in Table 3. The sampling frequency of the meteorological data is once every 2 minutes and the data are processed to hourly values.

Since January 1994 till now all the meteorological parameters are derived from KNMI measurement site Deelen.

Table 3: *Meteorological parameters in Arnhem*

parameter	unit	type sensor	location sensor
wind direction	degrees	weather-vane	mast H = 7 m
wind speed	m/s	cup anemometer	mast H = 7 m
relative humidity	%	solid state sensor	H = 1.5 m above ground level
temperature	°Celsius	PT-100 sensor	H = 1.5 m above ground level
global radiation	$W.m^{-2}$	PAR[1] pyranometer	roof cabin H = 2.5 m
air pressure	hPa	vacuum membrane	KNMI measurement site Deelen

[1] PAR = Photosynthetic Active Radiation

Kollumerwaard

Measurement site Kollumerwaard is equipped with six meteorological sensors. Table 4 shows the parameters with type of sensor and the height of the measurements above ground level.

Table 4: *Meteorological parameters in Kollumerwaard*

parameter	unit	type sensor	location sensor
wind direction	degrees	weather-vane	RIVM mast H = 10 m
wind speed	m/s	cup anemometer	RIVM mast H = 10 m
relative humidity	%	solid state sensor	sampling inlet height H = 8 m
temperature	°Celsius	PT-100 sensor	sampling inlet height H = 8 m
global radiation	$W.m^{-2}$	pyranometer	roof cabin H = 2.5 m
air pressure	hPa	vacuum membrane	in cabin near the GC

The sampling frequency of the meteorological data is once every 5 minutes and the data are processed to hourly values.

3.5.2 Delft

Hourly values of wind speed and wind direction were obtained from the national meteorological institute (KNMI). The data originate from the nearest meteorological monitoring station Zestienhoven, about 7 km south east of Delft.

3.6 Data Processing

3.6.1 Arnhem and Kollumerwaard

Analog FID-signals are processed using a two-channel Nelson intelligent interface (RS 232 version) and a commercial Nelson software package on an HP Vectra computer. Concentrations are calculated by the Nelson software using peak height and regression function of daily performed calibrations. Validated and checked concentration values are corrected to standard ambient conditions P = 1013.25 mbar and T = 25°C according to WMO recommendations using locally measured meteorological information.
Dedicated customized software is linked to Nelson after each run, performing several functions.

1. data control regarding retention times, over range peaks and ratio of peak area for last and previous run.
2. data reduction and storage (on hard disk) after acceptance of testing criteria.
3. steering of injection of standard gas mixture at 3-hour intervals.
4 creating control files for remote check of system performance by modem communication.
5. resetting the general system alarm.

Data are copied on floppy disk on station visits necessary for calibration and taken to home lab.

3.6.2 Delft

Chromatograms were recorded and on-line integrated using a SP 4400 Chromjet integrator. Peak integrals were immediately converted to concentrations, using the nominal value of the calibration standard. Ambient concentration and calibration data where then stored on a PC hard disk.
Ambient air concentrations were corrected afterwards by application of monthly (or shorter in case of instrument readjustments) average correction factors derived from the calibration data as described above.
Hourly concentrations of methane were calculated by averaging the concentration values obtained for each hour, with the restriction that at least two measurements were available.

4. Results of stationary methane measurements

Below, data are presented from stationary measurements at the monitoring sites Arnhem (A), Kollumerwaard (K) and Delft (D).
By analyzing various cross-sections of the data set, the main variables are derived, which govern the concentration levels and their variation in space and time.
Since the main emphasis of this study is on the contribution of various sources to ambient concentrations in the Netherlands, the amount by which concentration levels exceed the background concentration level, more precisely the Northern Hemisphere (NH) background concentration, are of particular interest.
Taking into account a NH background concentration of about 1.8 ppm as compared to an average ambient concentration level of 2.0 ppm, it is clear that any source contribution is derived by subtracting two large numbers to yield only a relative small contribution to which any bias in the background concentration is transferred, making up a substantial part of the source contribution.
Rather than taking NH background concentrations from literature, it is therefore better to derive these from the measurements themselves in order to avoid any bias. Natural variation in time and space as well as any trend in the NH background concentration may be not insignificant with respect to source contributions. Moreover, when using data from various monitoring sites or literature, where measurements have been carried out independently by various organisations, any bias in these measurements will also seriously effect differences calculated from these data sets. In order to illustrate the problem we recall from chapter 3.4 the results of intercomparison giving differences between laboratories which range between 0.04 and 0.09 ppm, which is between 2 and 4 % of the measured concentration, but would be translated (by subtracting a fixed background for all measurements) to between 20 and 40 % of the concentration in excess of the background value.

4.1 Determination of background concentration.

At monitoring sites which are not characterised as "remote", background concentrations are only expected to be measured incidentally under specific conditions, such as wind directions from non-source areas and high atmospheric dilution. Especially at coastal stations in the Netherlands (Kollumerwaard and with some limitations Delft) such conditions occur with north-western marine winds of atlantic origin, with high wind speed and good vertical mixing. In the Netherlands high wind speeds generally occur at NW-wind directions and vertical mixing is generally high at such meteorological conditions, especially at mid-day.

Several methods can be used to derive the background concentration from the data sets:

- from inspection of windrozes and diurnal variation of the concentration for minimum values;
- from extrapolation of concentrations as a function of wind speed;

- from lower percentiles of the measured concentrations;
- from minimum concentration(s) measured.

An investigation of these various methods showed:

- Minima of the windrozes and diurnal variation resulted in a relatively high (with respect to recent literature data of 1.75-1.80 ppm) estimate of the background concentration. Such a high estimate can be expected due to the inclusion of data collected under non-background conditions. A more detailed break up of the data set would be required.
- Concentrations at wind speeds above 3 m/s follow closely a proportional relationship with the inverse of the wind speed, facilitating easy extrapolation. Extrapolation to infinite wind speed results in rather low estimates, as at the highest wind speeds measured there still is a considerable slope in the function. Concentrations at the highest wind speeds measured are still relatively high, probably due to mixing with data from wind directions wich do not give real background concentrations.
- Using a lower percentile of the concentration data seems attractive, but the choice of that percentile (somewhere below 2) is rather arbitrary and the background value derived will strongly depend on the percentile chosen.
- The use of the single minimum value in a data set could very much be subject to experimental error, as one only relies on a single measurement. Such could be overcome by using the average of the minimum values measured each month, taking into account the consistency of these data.

The latter method was adopted for deriving the background concentration. Table 5 shows the values including their standard deviation.

Table 5: *Estimated background concentrations and standard deviations. All data in ppm.*

	Kollumerwaard	Arnhem	Delft
1990	1.72 ± 0.05		
1991	1.73 ± 0.03	1.71 ± 0.11	
1992	1.75 ± 0.11	1.72 ± 0.21	
1993	1.75 ± 0.11	1.58 ± 0.09	
1994	1.74 ± 0.04	1.75 ± 0.04	1.80 ± 0.02

From this table it is observed that in some years the estimate of the background concentration has a rather wide confidence interval. The estimate for 1994 however, is within acceptable limits and shows improved consistency of the data within each subset of data.
In addition it is observed that the fair consistency over a number of years is incidentally broken. The data also indicate some bias between data sets from different origin (1994).

From these data it is concluded that the background concentration derived from these data sets is between 1.75 and 1.80 ppm, in agreement with recent literature data (Bosinger et al., 1993; Simmonds et al., 1991; Khalil et al., 1993). Recent data

from Cabauw (Veltkamp *et al.*, 1995) indicate an average background of 1.80 ppm in agreement with data from a remote oceanic monitoring site.

Additionally it is concluded that the possible bias between the data sets from different monitoring sites is best eliminated (by rescaling) before conclusions are drawn on differences measured, that is on source contributions. After a preliminary comparative analysis of windrozes from the three monitoring sites, it was concluded that most consistent results were obtained by rescaling the Kollumerwaard and Arnhem data by adding 0.06 and 0.05 ppm, respectively.

4.2 Concentration levels

4.2.1 Average concentrations

Average concentrations for the three monitoring sites in 1994 are presented in Table 6. For comparison also data for 1990 to 1993 are included.

Table 6: *Average concentrations of CH_4 (in ppm) for the whole years and for summer and winter half years.*

	year	summer half	winter half
Arnhem 1990 *	1.95	1.90	2.05
Kollumerwaard 1991 **	1.98	1.95	2.00
Arnhem 1991	2.00	1.99	2.02
Kollumerwaard 1992	1.99	2.00	1.98
Arnhem 1992	1.98	1.95	2.00
Kollumerwaard 1993	1.98	1.98	1.99
Arnhem 1993	2.05	1.96	2.16
Kollumerwaard 1994	1.96	1.95	1.96
Arnhem 1994	2.02	2.00	2.04
Delft 1994	2.02	1.98	2.08

* April to december
** July to december

The average concentrations at the three monitoring sites show only small differences to such an extend that possibly experimental error interferes with detecting clear patterns. Nevertheless, the data reveal that average concentrations at Arnhem and Delft are slightly higher than at Kollumerwaard and this is most pronounced in the winter half year, the concentrations in the summer half year being almost equal. In the winter half year the concentrations are higher than in the summer half year, except for Kollumerwaard where they are almost equal.
The differences observed are only a few percent of the total concentration. With respect to the concentrations in excess of the background concentration the differences are up to 50 %.
No trends are observed in the average concentrations at Arnhem and Kollumerwaard.

4.2.2 Frequency distributions

In order to compare the range of concentrations of methane at the monitoring sites the 50-, 95- and 98 percentiles of the frequency distribution are given in Table 7. For comparison also data from earlier years are given at the Kollumerwaard and Arnhem sites.

Table 7: *Percentiles of CH_4 concentrations (in ppm) for the whole years and for winter and summer half years.*

		year			summer			winter		
		50	95	98	50	95	98	50	95	98
Arnhem	1990	1.89	2.42	2.67	1.85	2.26	2.44	1.96	2.66	2.87
K'waard	1991	1.89	2.45	2.76	1.88	2.38	2.69	1.91	2.52	2.80
Arnhem	1991	1.94	2.51	2.70	1.94	2.33	2.81	1.94	2.61	2.81
K'waard	1992	1.94	2.35	2.55	1.95	2.35	2.62	1.93	2.34	2.51
Arnhem	1992	1.94	2.55	2.70	1.87	2.33	2.51	2.00	2.64	2.86
K'waard	1993	1.94	2.34	2.60	1.94	2.34	2.56	1.94	2.34	2.57
Arnhem	1993	1.98	2.59	2.88	1.92	2.34	2.50	2.08	2.82	3.11
K'waard	1994	1.90	2.35	2.57	1.90	2.30	2.47	1.89	2.40	2.68
Arnhem	1994	1.96	2.47	2.69	1.96	2.39	2.58	1.95	2.56	2.90
Delft	1994	1.94	2.51	2.76	1.91	2.35	2.55	1.98	2.67	2.94

First it is noted that the frequency distribution of the concentrations is almost log-normal. It is noted that the cumulative distributions generally show a "knick" at about 2.0 ppm (the 70-75 percentile), suggesting two modes in the distribution separating concentrations below and above 2.0 ppm. This effect is however not discussed further.
The log-normal distribution is characterized by its median (50-percentile) and its standard geometric deviation (SGD).
Typical for the frequency distributions of the CH_4 concentrations is the very small SGD of about 1.2. This is due to the constant background concentration of 1.80 ppm, making up a large part of the overall concentration. The SGD of the concentrations in excess of the background value is about 2.5, which compares with the SGD's of other pollutants not largely determined by their background values.
As a result of the small SGD the 50-percentiles are only slightly lower than the average concentrations. The typical patterns in the 50-percentiles are therefore very similar to those of the averages discussed above. It is only noted that the 50-percentiles, being more robust statistical parameters, more consistently show the typical patterns as displayed by the averages.

The higher percentiles illustrate the range of the CH_4 concentrations generally found. Only occasionally concentrations above 3.0 ppm are found (0.5 to 1% of time). Maximum concentrations measured were up to 5 ppm.
Despite the intrinsic higher variability of the higher percentiles, they even stronger than averages and 50-percentiles show typical patterns very similar to the latter. Due to the fact that we deal with higher concentrations, differences are more significant and not obscured by the margins of experimental error.

For the winter half year the 95- and 98-percentiles at Arnhem and Delft are very similar, but distinctly higher than in Kollumerwaard. This is not the case in the summer half year where the higher percentiles at all three sites are very similar. The 95- and 98-percentiles in the winter half year are higher than in the summer half year, except for Kollumerwaard, where they are almost equal.
No typical trend in the higher percentiles is observed at the sites Arnhem and Kollumerwaard.

4.2.3 Time series of concentrations

Figure 3 shows the 50- and 95- percentiles of the methane concentrations for each month (1994) in a time series plot. Figure 4 shows the ratio's of 98- and 95-percentiles to the 50-percentile and the ratio of the upper to lower quartile of the concentrations, which illustrate the variation in the dynamics of the concentrations over the year. With the lower limit of the concentrations bound by the background concentrations, any increase of the ratios is due to the occurrence of higher concentrations.

The interquartile range (the mid 50 % of the concentrations) only shows small changes over the year and between monitoring sites, but notably increases in the winter months, also showing more divergence between the monitoring sites in the winter months. The 95- and 98 percentiles alter strongly from one month to the other. The higher percentiles strongly increase in the winter months, most notably for Arnhem and Delft.

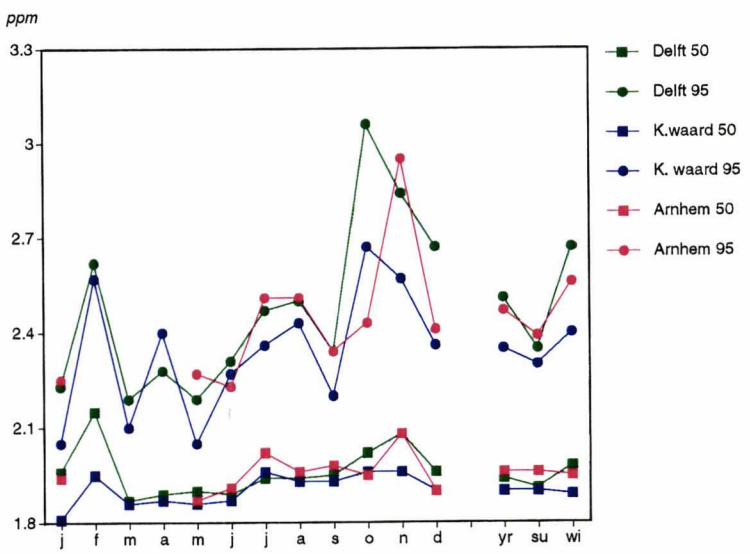


Figure 3: *Time series of 50- and 95 percentiles of methane concentrations (ppm) in 1994 for Arnhem, Delft and Kollumerwaard.*

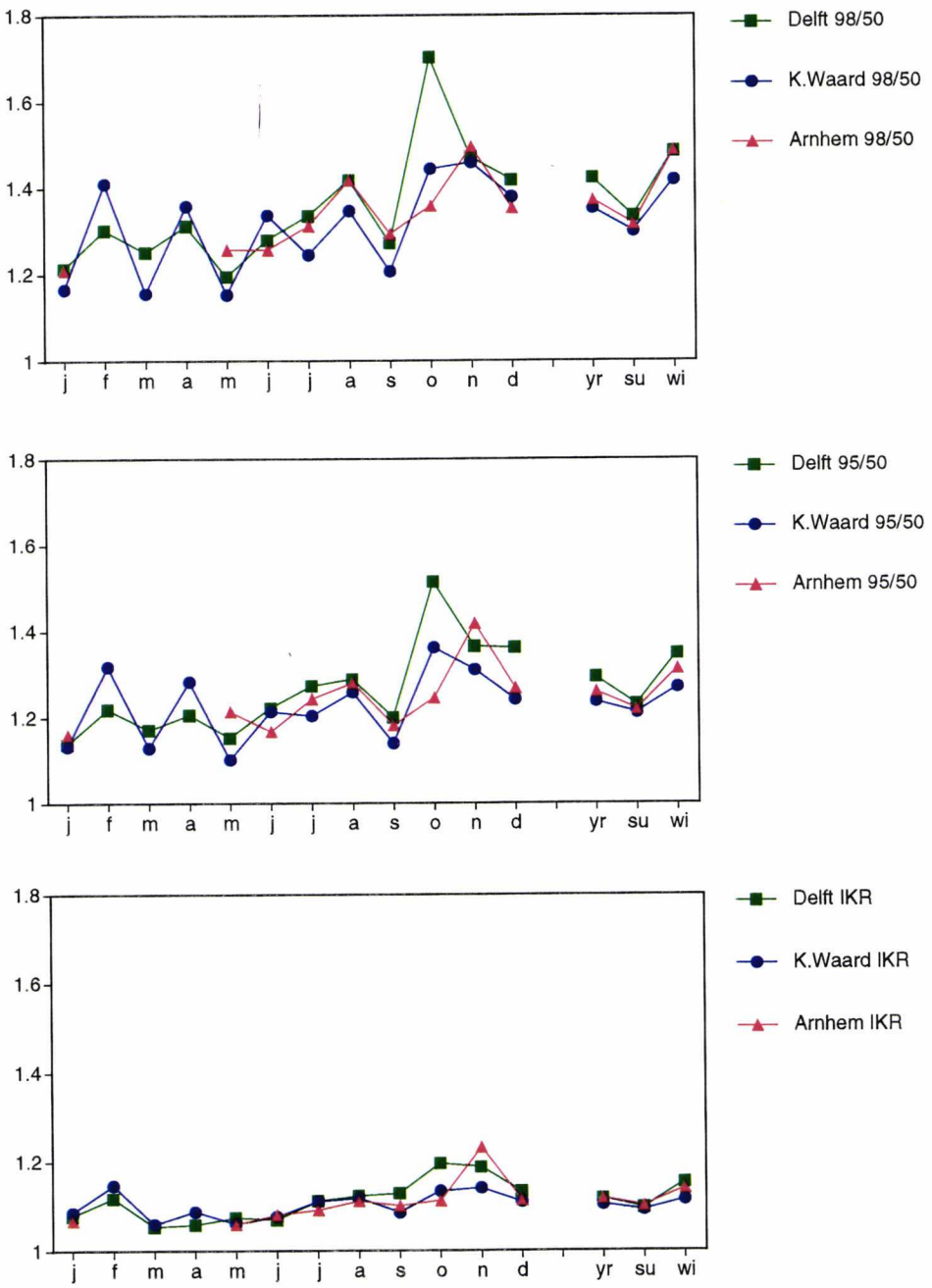


Figure 4: *Time series of ratios between 95/50, 98/50 and 75/25 (interquartile range) percentiles for Arnhem, Delft and Kollumerwaard.*

From these figures it is clear that variation over the year follows the same general pattern at all three monitoring sites, but the amplitude of the variation with time differs for each station.

4.3 Diurnal variation

The diurnal variation of the methane concentrations is presented in Figure 5. Some characteristic concentration levels and ratio's derived from these diurnal variations are given in Table 8.

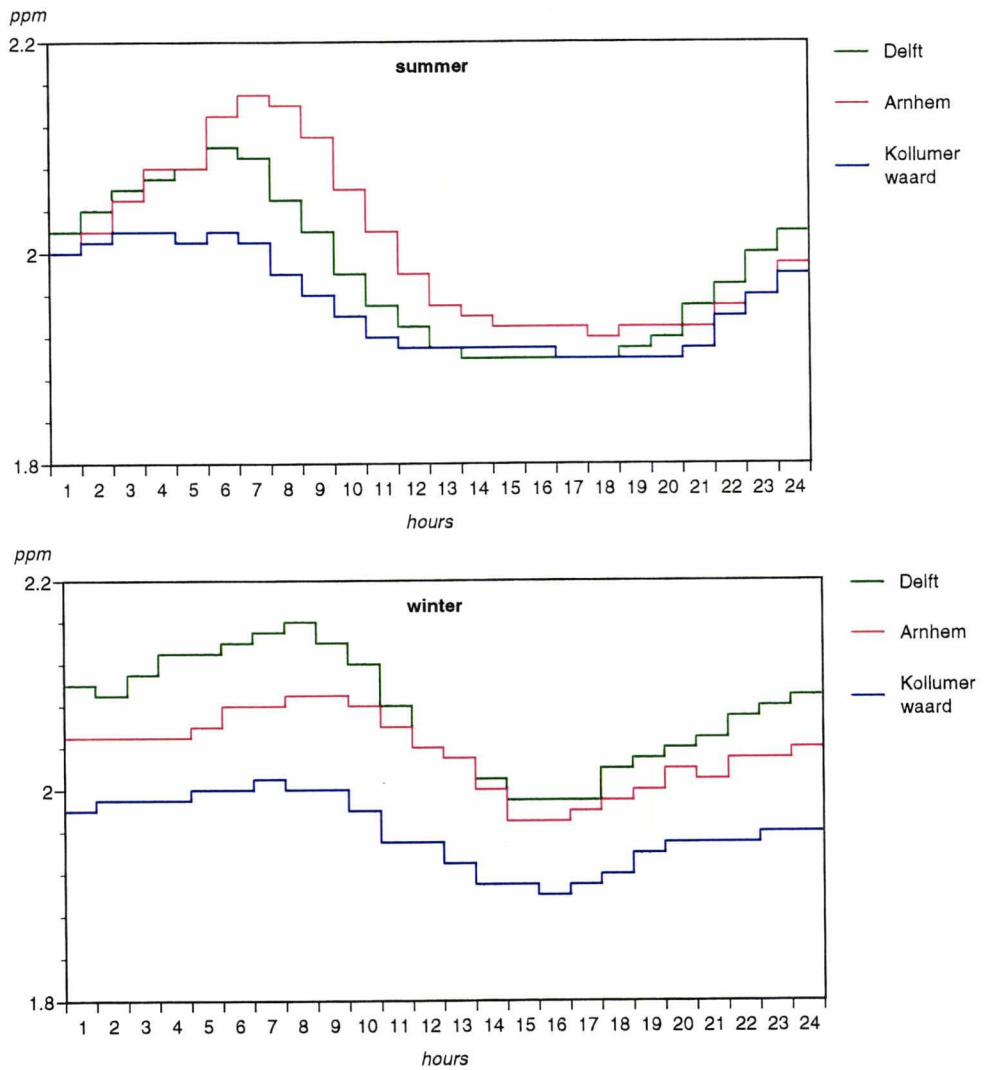


Figure 5: *Diurnal variation of the methane concentrations in 1994 for Arnhem , Delft and Kollumerwaard. a: summer half year. b: winter half year.*

The diurnal variation typically runs parallel with the diurnal variation of meteorological variables, such as mixing hight, wind speed and temperature, all reaching maximum values at about mid day. Taking also into account that most sources emit methane at or close to ground level are ground sources, understandably lowest concentrations are observed around noon when atmospheric mixing is maximal and highest concentrations at night (early morning) when mixing is minimal and low inversions may occur.

Table 8: *Characteristics of the diurnal variation. Concentrations in ppm. Concentrations in excess of background in parenthesis.*

	morning decrease max.		noon min.		max/min min. ratio	
	time	conc.	period	min. conc.	tot.	exc
Summer						
K'waard	6.00	2.02 (0.28)	11.00-21.00	1.90 (0.16)	1.06	1.75
Arnhem	7.00	2.15 (0.40)	14.00-21.00	1.93 (0.18)	1.11	2.22
Delft	6.00	2.10 (0.30)	13.00-18.00	1.90 (0.10)	1.11	3.00
Winter						
K'waard	9.00	2.01 (0.27)	13.00-17.00	1.90 (0.16)	1.06	1.69
Arnhem	9.00	2.09 (0.34)	14.00-16.00	1.97 (0.22)	1.06	1.55
Delft	8.00	2.16 (0.36)	14.00-17.00	1.99 (0.19)	1.09	1.90

Some typical differences, however, are also observed between the monitoring sites, which relate both to the time scale of the patterns and the span of the concentrations observed.

On the time scale, most notable are in the summer half year the long period of minimum concentrations at Kollumerwaard and the early rise in the afternoon of the concentrations at Delft.

In the winter half year the period of minimum concentration in Arnhem is very short.

This may as well relate to typical meteorological differences for coastal and more inland monitoring sites as well as to the presence of sources with specific diurnal patterns in their emissions.

Minimum concentrations in the summer at noon are very similar, with Arnhem slightly higher than Delft and Kollumerwaard.

At Kollumerwaard the minimum concentration in the winter is equal to that in the summer. At Delft and Arnhem minimum concentrations in winter are higher than in summer.

The (early) morning maxima in the diurnal patterns differ substantially between sites.

The lowest maximum is found at Kollumerwaard, where summer and winter maximum are almost equal. Maximum concentrations in Delft and Arnhem are larger, but also change order in summer and winter.

The amplitude of the diurnal variation can be expressed by the ratio between the maximum and minimum concentrations for each site and season (Table 8).

The differences are most clearly seen when not the ratio of the total concentrations (for a large part due to background contribution) is calculated, but the ratio of the concentrations in excess over the background concentration related to source contributions. These ratio's vary between 1.5 and 3.0.

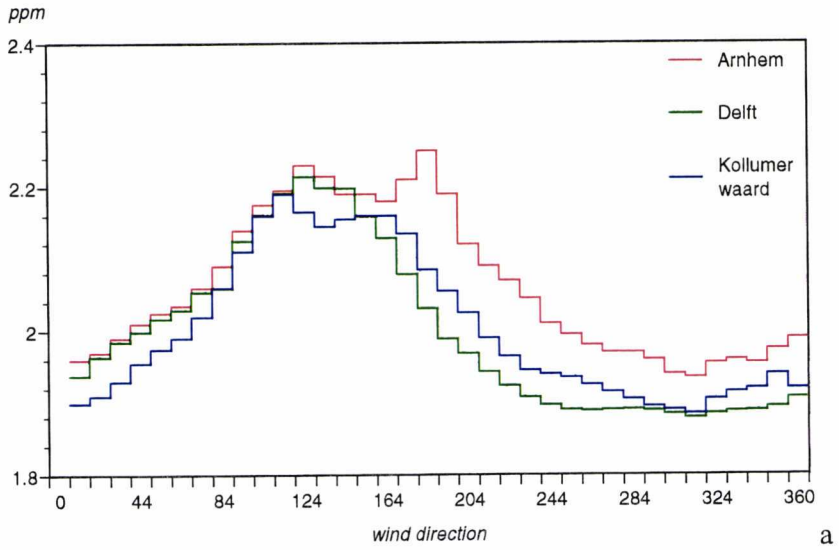
Although inevitably influenced by meteorological factors, the variation in the minimum/maximum ratio's indicates different source contributions for the monitoring sites and in the summer and winter half year.

4.4 Variation with wind direction

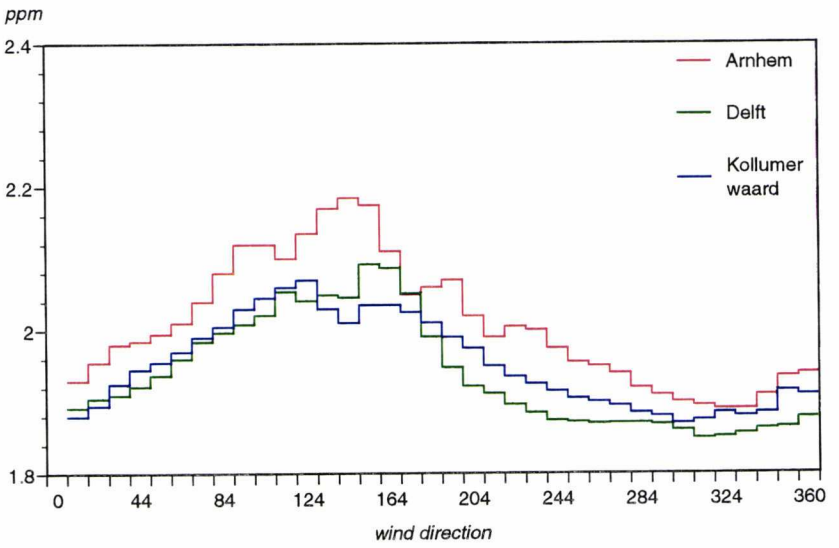
The variation of the concentrations with wind direction is most easily discussed at the hand of so called windrozes, in which concentrations are displayed as a function of wind direction. Locally measured wind directions are used to construct such diagrams. For the data analyses two- (KEMA data) and three-sector (TNO data) moving averages have been used in order to handle some differences in the systematics of the reporting of meteo data.
In order to satisfy meteorologists, it is noted that air parcels not necessarily travel in straight lines, especially not over large distances. The use of moving averages mitigates this problem to some extent.
Where windrozes are used to indicate the directions of the presence of major sources, this is thus more justified for sources at short distance than at long distances. Where our intention is not to pin point very precisely (individual point) sources at large distances, but merely to distinguish between transboundary source contributions and Dutch ones, the latter to be identified more precisely, the analyses of windrozes and differences between windrozes at the three monitoring sites is a very useful tool.

4.4.1 Windrozes

Windrozes for the summer and winter half years at the three monitoring sites are shown in figures 6 and 7.



a



b

Figure 6: *Windrozes of methane concentrations in 1994 for Arnhem, Delft and Kollumerwaard; summer half year. a: all data; b: windspeed ≥ 4 m/s*

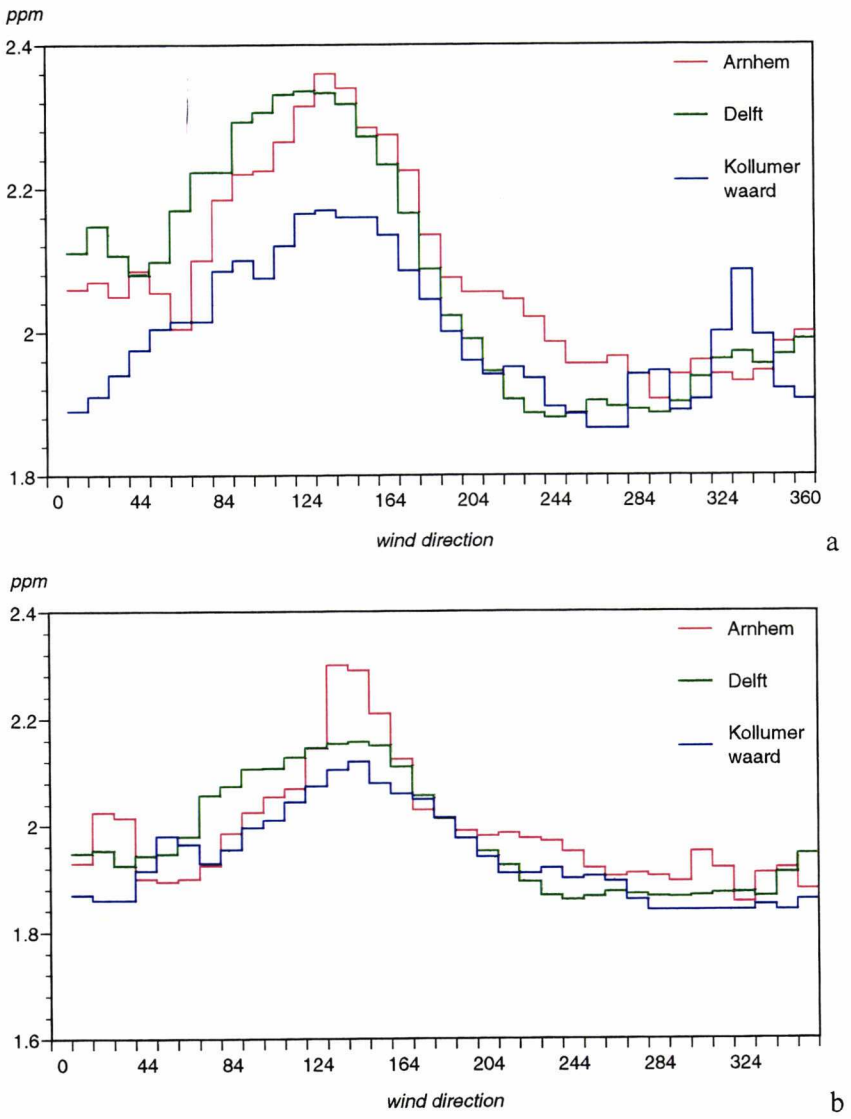


Figure 7: *Windrozes of methane concentrations in 1994 for Arnhem, Delft and Kollumerwaard; winter half year. a: all data; b: windspeed ≥ 4 m/s*

The most general and common feature of all windrozes (site and season) is a broad maximum around south-east wind direction and a broad minimum around north west wind direction. Typical differences for the monitoring sites are the average concentrations in the minima and maxima. While the minimum concentrations do not differ very much for the summer and winter half year, the maximum concentrations do, with exception of Kollumerwaard.

The concentrations in excess over the background concentration range from less than 0.1 ppm to 0.55 ppm. The highest values occur with air masses arriving over land. Since the monitoring sites Arnhem and Kollumerwaard are located near the east border of the Netherlands, they relate to emissions in Germany and further upwind in central Europe. The lowest values occur with air masses arriving over sea, where emissions are much lower. Typically the non-coastal site Arnhem has

under this conditions higher concentrations than the other two (coastal) sites, indicating the contributions of Dutch emissions.
It is also quite notable that with south to south west wind directions concentrations in Arnhem are much higher than in Delft. This indicates that the source density in the more continental areas south of the Netherlands are much larger than those in the coastal areas south of the Netherlands.
In the summer half year not much fine structure is superimposed on this general behaviour, except for Arnhem.
The winter half year windrozes show more fine structure. This fine structure occurs for Kollumerwaard and Delft at wind directions between North-West and North and at Arnhem and Delft in the North to North-East sectors. In some cases the observed fine structure is however only due to a small number of high concentrations in a compass sector with only few observations. In such cases the significance of the structure is difficult to estimate.
Taking data for higher wind speeds only (> 4 m/s), some fine structure is lost and some added, while other become more explicit. Such effects indicate remote as well as local source contributions. The differences in the windrozes for all wind speeds and those for high wind speeds only are most notable for the winter half year on all sites and in the summer half year for Arnhem.

Thus the windrozes show almost background concentrations at Kollumerwaard and Delft for north west wind directions, which represent marine air masses, while the somewhat higher concentration at Arnhem with those wind directions must be due to additional contributions from emissions in The Netherlands.
The higher concentrations for south east wind directions, representing continental air masses, relate to large source areas south east of The Netherlands, the contribution of which is somewhat lower at the most northern monitoring site Kollumerwaard, but only in the winter half year. The latter effect may relate to the relative contributions of various source categories which not necessarily are of equal strength in summer and winter. The fine structure in the windrozes indicates that in the winter half year there are more and different contributions from local sources than in the summer half year.
Part of these sources seem to be related to large urban areas.
It has to be noted that the average concentrations as a function of wind direction do not directly reflect upwind source strengths. Emissions relate to concentrations via dispersion characteristics, which vary systematically with wind direction. Generally wind speed and mixing height are larger for western (atlantic) wind directions as compared to eastern (continental) wind directions.

4.4.2 Windroze differences

In table 9 average concentrations and differences between them have been tabulated for compass points in which pairs of monitoring sites line up. All differences are calculated as the concentration at the downwind site minus the concentration at the upwind site.

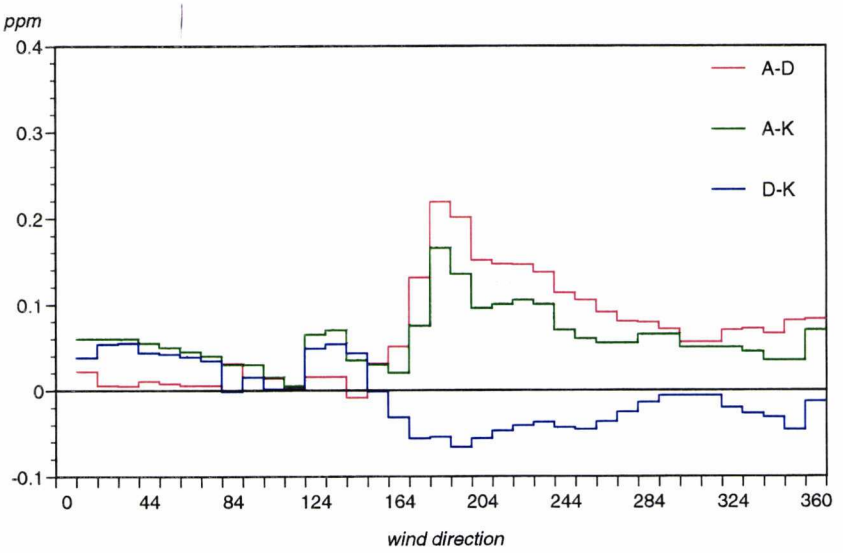
With the monitoring points located north (Kollumerwaard), south west (Delft) and south east (Arnhem), an increase of the concentration (positive values for downwind minus upwind) will be due to emissions in The Netherlands along tracks of 100 to 200 km.

Table 9: *Average concentrations and differences between average concentrations (Bold printed for compass points connecting the various monitoring sites). All concentration data in ppm.*

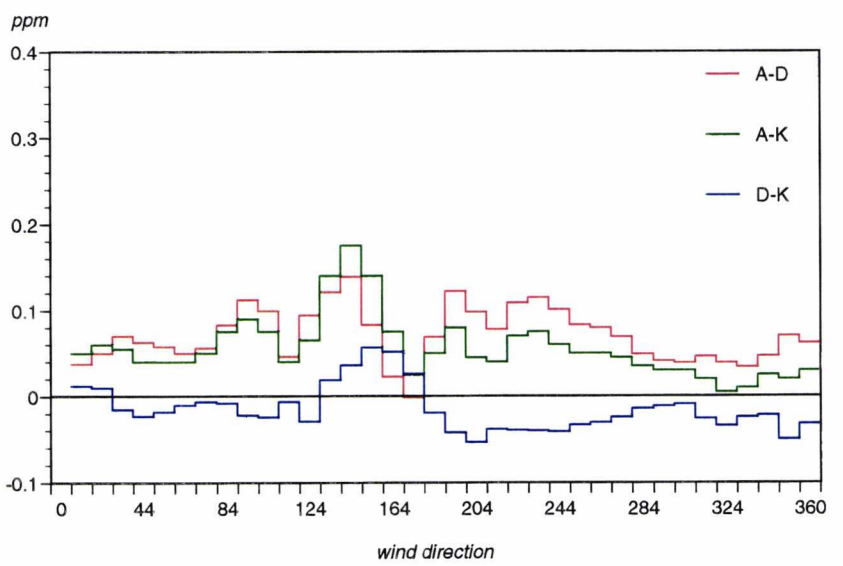
sites, from-to	**D-A**	**A-D**	**K-A**	**A-K**	**K-D**	**D-K**
compass point	270°	090°	010°	190°	040°	220°
distance (km)	100	100	150	150	200	200
Concentrations						
Summer half						
Arnhem	1.97	2.14	1.96	2.19	2.01	2.07
Delft	1.89	2.12	1.93	1.99	2.00	1.92
Kollumerwaard	1.91	2.06	1.90	2.06	1.96	1.97
Winter half						
Arnhem	1.97	2.22	2.06	2.08	2.09	2.05
Delft	1.90	2.29	2.11	2.02	2.08	1.90
Kollumerwaard	1.87	2.10	1.89	2.00	1.98	1.95
Differences						
summer half A-D	**+0.08**	**-0.02**	+0.03	+0.20	+0.01	+0.15
K-D	+0.02	+0.06	+0.03	+0.07	**+0.04**	**+0.05**
A-K	0.06	+0.08	**+0.06**	**-0.13**	+0.05	-0.10
winter half A-D	+0.07	+0.07	-0.05	+0.06	+0.01	+0.15
K-D	-0.03	+0.19	+0.22	-0.02	**+0.10**	**+0.05**
A-K	+0.10	+0.12	**+0.17**	**-0.08**	+0.11	-0.10

In some cases negative differences are found, which are explained by the presence of local sources of methane at short distances upwind of the upwind monitoring site of the pair. This particularly is the case south and east from Arnhem in the summer half year and south from Arnhem in the winter half year.
The fact that differences on paths from and to a monitoring sites are almost similar indicates some consistency of the result.
Where large differences are observed from and to the same monitoring site local sources must be present just upwind of the receiving site. This is the case for Arnhem and Delft in the winter.
Not taken into account the effects of local sources, the path differences are generally in the range of 0.04 to 0.10 ppm, which could be thought of as due to diffuse non point sources along those paths.
Since the most important emissions of methane in the Netherlands (cattle, waste disposal sites, manure, wetlands, gas distribution and gas production) may be regarded as collective emissions distributed over the whole country (with possible some concentration areas) the path average increase of the concentrations will be a measure of the emission density along those paths.

Figures 10 and 12 show the data given in table 9 projected on a map of the Netherlands.

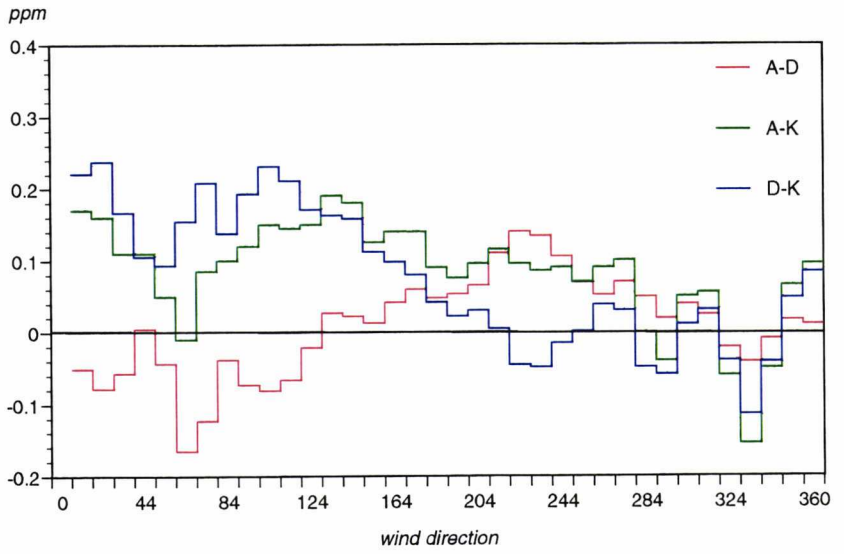


a



b

Figure 8: *Difference windrozes for the summer half year 1994. a: all data; b: windspeed ≥ 4 m/s*



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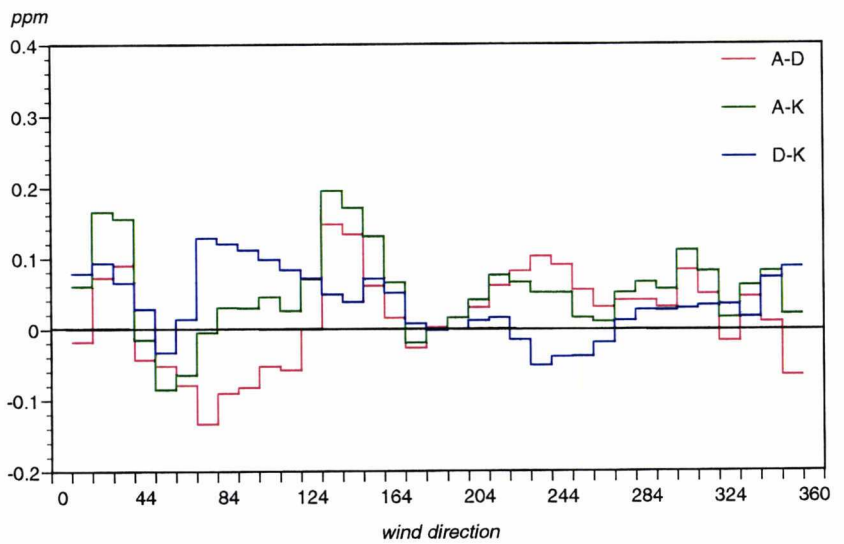

b

Figure 9: *Difference windrozes for the winter half year 1994. a: all data; b: windspeed ≥ 4 m/s*

Differences of the concentrations over the full compass simply obtained by subtracting windsector average concentrations, are shown in figures 8 and 9.
As the windrozes for Kollumerwaard are very similar for the winter and summer half year and generally show the lowest concentrations, these may conveniently be used as a reference.
The difference curve for Delft-Kollumerwaard for the summer half year is easily understood taking into account that air masses having travelled over land have taken up more methane emissions than those which have travelled over sea.

The differences between Delft and Kollumerwaard are generally less than 0.05 ppm. Although such differences could be close to the margin of error, the systematics is such that they have to be regarded real.
For the winter half year the differences are much larger, and the increased fine structure indicates the presence of more local sources.
The differences curve for Arnhem-Kollumerwaard for the summer half year is positive over the whole compass, also about 0.05 ppm, except in the south to south east direction where the difference increases above 0.1 ppm, indicating local sources south to south west from Arnhem. Also in this case the differences for the winter half year are larger and extend over a larger part of the compass.
The difference curve for Arnhem-Delft for the summer half year shows, as in the Arnhem-Kollumerwaard case, the contribution of local sources south to south west of Arnhem. For the winter half year the curve shows more fine structure, which will be due to local sources north to east of Delft.

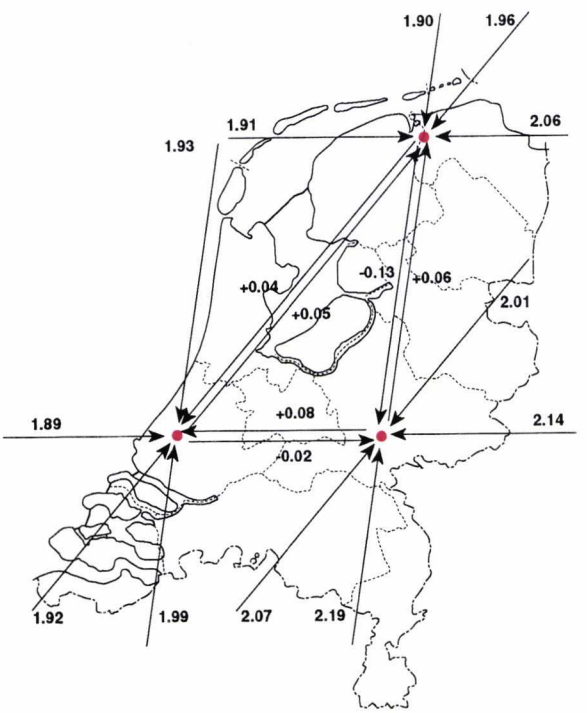

Figure 10: *Local background concentrations and increase (+sign) of concentrations along trajectories between upwind and downwind monitoring sites. Summer half year 1994.*

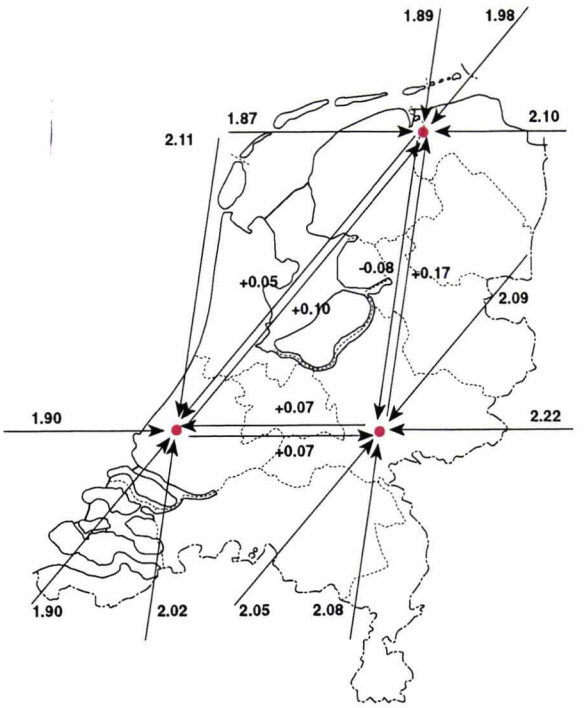

Figure 11: *Local background concentrations and increase (+sign) of concentrations along trajectories between upwind and downwind monitoring sites. Winter half year 1994.*

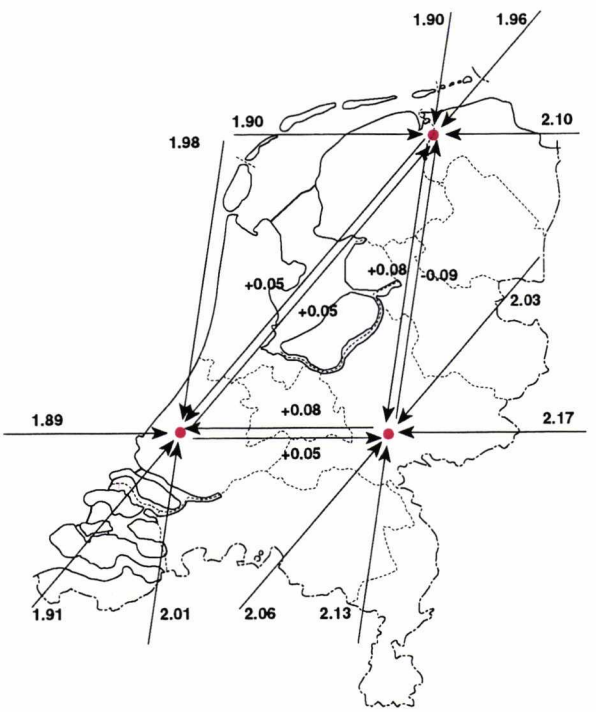

Figure 12: *Local background concentrations and increase (+sign) of concentrations along trajectories between upwind and downwind monitoring sites. Whole half year 1994.*

5. Modelling of methane concentrations

5.1 Model selection

The aim of the modelling exercise is to couple the measured concentrations, or the measured contributions to the concentrations in excess over the NH background concentration to emissions. With the present knowledge on emissions as an input to the model, the comparison of calculated and measured contributions to the background concentrations will verify the correctness of the emission estimate. The other way around, the modelling will lead to an estimate of the emissions required to reproduce the measured contribution to the background concentration. This emission estimate can then be compared with the present knowledge of emissions.

Various models are available to carry out such calculations. In hierarchical order these are:

- box models;
- plume models;
- trajectory models;
- 1-2-3 D models.

Which model to use depends on the question to be answered and the complexity of the mass balance. In a first approximation modelling of methane concentrations has to deal with a) background concentrations, b) emissions and c) transport.
Chemistry and deposition can in first approximation be neglected when only transport over areas of the size of The Netherlands is considered. Within these boundary conditions the available monitoring data determine the model to be used best.
The relative coarseness of the horizontal resolution (50 x 50 km or more) of current continental dispersion models (1-2-3 D models) allows for only a small number of grid cells describing the atmospheric conditions above the Netherlands, which makes them less suitable for our purposes.
Trajectory models are typically applied in the comparison of time series of measurements. Moreover such analyses have been carried out in the NOP programme by ECN (Veltkamp et all.,1995) using measurements at the 200 m high meteorological tower at Cabauw.
For the interpretation of long term average concentrations and differences between them measured at a few monitoring points box and plume models are sufficient. As plume models generally are more limited to local scales, a box model applicable to the scale of the distances between the monitoring sites was chosen for application in this case (Derwent *et al.*, 1995.

5.2 Model results

The basic data for application of the model were wind sector averaged concentrations at the three monitoring sites and differences between them for those compass points where pairs of monitoring sites align in an upwind-downwind configuration. The extraction of the relevant data from the measured concentrations is described in chapters 4.4.1 and4.4.2 and summarized in Table 9.
The box model assumes that methane emissions are more or less homogeneously distributed along the path between monitoring sites. Such an assumption seems to be justified by the fact that the majority of emissions in The Netherlands (Table 1) is due to so called collective emissions strongly related to land use and population density.
The application of the box model under average meteorological conditions in The Netherlands (mixing height 500 m and wind speed 4.5 m/s) leads to a parameterized equation relating concentration differences to source strengths:

$$E = 47.3\ dC/L$$

where: E is the emission rate ($g/m^2.yr$), dC the concentration difference (ppb) between downwind and upwind monitoring site and L the distance (km) between the monitoring sites.
In the detailed calculations, the results of which are presented below, the numerical constant in the equation was adjusted to measured wind speeds as a function of wind direction and season.
No such detailed information was available for the mixing height, so only the average value was used for all calculations. As a result of mainly the latter approximation the uncertainty of the derived emission rates is roughly estimated as +/- 30 %.
The uncertainty in the total Dutch emissions (averaging out various factors) may be somewhat smaller. It should however also be taken into account that the area enclosed by the monitoring sites does not cover the whole country. So with some extrapolation included the uncertainty in the total Dutch emissions may well remain about +/- 30%.
From the results it appears that the model works satisfactory for those cases where the upwind monitoring site has a close to background concentration. Where the upwind concentration deviates strongly from the background value, which will be due to upwind sources at short distances, the model does not perform satisfactory, resulting in a negative value for source contributions between the pair of monitoring sites.
In other cases concentration differences along the path from A to B differ from those along the same path in the reverse direction B to A. This will be related to sources close to the downwind monitoring site or to systematic differences in the meteorological conditions connected to the two different wind directions involved.

For the summer half year the calculations result in emission densities of 10, 18 and 43 $g/m^2.yr$ for the western, eastern and southern parts of the Netherlands, respectively.

Generally somewhat higher emissions are found for the winter half year of 17, 33 and 39 g/m^2.yr for the western, eastern and southern parts of the Netherlands, respectively.
Overall this gives an average emission rate of 27 g/m^2.yr and a total emission for the Netherlands (40.000 km^2) of 1.080 kton per year.
From trajectory analyses of one year methane measurements at Cabauw ECN estimated a total Dutch emissions of 1.130 kton per year (Veltkamp *et al.*,1995). This very similar totals derived from concentration measurements and modelling are in fair agreement with total emission estimates from emission inventories. As table 1 shows the 1991 estimate amounts from a low 760 to a high 1.730 kton per year with a median value of 1.230 kton per year. Recent estimates from the Dutch Emission registration give a 1995 total of 1.090 kton per year (Berdowski *et al.*, 1995).
Thus it appears that the trajectory analyses of the time series of measurements at Cabauw, as well as the analyses of concentration differences from measurements at three monitoring sites result in a similar estimate of total emissions in the Netherlands (1.100 ton per year) which is close to the total accounted from recent emission inventories.

6. Concluding remarks

Within the National research programme on Global air Pollution and Climate Change several research projects have been carried out, under theme B: "causes", which have investigated the methane emissions of various source categories. Together with an inventory of sources these investigations provide an account of the total Dutch methane emissions.
This approach results in an uncertainty in the total emission due to the inevitable limited sample survey of sources, the intrinsic variability of emissions within each source category and possible omissions of unknown sources or sources wrongfully estimated to be not important.
In order to validate and integrate the results of the subprojects on emissions additional studies were initiated in the integrated project cluster: "Validation, integration and scaling of methane emissions from various sources". Projects within this latter cluster aimed at studying methane emissions in the Netherlands at a higher aggregation level to provide a more general insight in the total Dutch emissions and the uncertainty therein.

The research strategy adopted for the study described in this report was to collect data on ambient concentrations of methane and compare these data by atmospheric modelling with present estimates of the Dutch emissions from emission inventories. To this end hourly concentrations of methane have been measured at three monitoring sites during a period of one year. A comprehensive analysis of the data has provided the source contributions of emissions in the Netherlands to ambient concentrations.
By atmospheric modelling, using a box model, emissions were derived from these source contributions and compared with the results of recent emission inventories. Additionally mobile measurement campaigns were carried out to supplement the information from the stationary monitoring points.

The mobile measurements were focused on the Groningen gas production area, which was suggested by occasional high concentrations measured at the monitoring site Kollumerwaard. The mobile measurements indicated that methane emissions in the north of the Netherlands are rather homogeneous. Continuous emissions were close to the detection limit of the monitoring instruments. Incidental "large" emissions from individual installation were not detected during the measurement campaigns. From the measurements it is concluded that under regular operational conditions at production and drilling sites of natural gas no particular elevated emissions occur.

The continuous monitoring of methane at three fixed monitoring sites over a period of one year has proved to be a very rich source of information on the behaviour and origin of ambient concentrations.
The major findings of the analysis of the concentration data are described below.

Average concentration levels at all three monitoring sites were 2.0 ppm (as compared to 1.5 ppm in 1979 at Terschelling (Hollander, 1079)). Generally the concentrations are in the range from 1.8 to 2.5 ppm. Incidentally higher concentrations are observed which may reach extreme values of 5 ppm.
From the data a northern hemisphere background concentration was derived of 1.8 ppm, which agrees with recent literature data on monitoring (Khalil and Rasmussen, 1990 and 1993, Bosinger et all., 1993, Simmonds et all., 1991 and 1993) and the results of a recent model calculation on a global scale (Roemer, 1995).

The variation in the ground level atmospheric concentrations is mainly governed by the time of the day and wind direction.
The diurnal variation mainly reflects the effect of wind speed and mixing height on the concentrations resulting essentially from ground level sources and probably to a lesser extent diurnal variations in source strength.
The variation with wind direction mainly reflects the presence of upwind source areas, modified to some extent by systematics in dispersion as a function of wind direction.
Only small source contributions add to air masses from marine or atlantic origin: less than 0.1 ppm.
Source contributions of well over 0.5 ppm are found in continental air masses arriving from central Europe. Source contributions in air masses from more southern and northern parts of the European continent are lower (0.3 ppm) and lowest values are found in air masses from the south west, coastal part of the continent (0.1 ppm).
From a comparative analysis of source contributions at the three monitoring sites, using compass points where the monitoring sites line up in upwind-downwind position, source contributions from only Dutch sources were derived. These range between 0.04 and 0.10 ppm.
In the process, local source contribution were also identified which relate to emissions from densely populated areas.

By atmospheric modelling the source contributions from Dutch emissions were converted to emission densities for various parts of the Netherlands.
This resulted in emission densities ranging from 10 to 43 $g/m^2.yr$, with the lowest values found for the western and northern parts of the Netherlands and the highest values for the southern part of the Netherlands. This is in fair agreement with the totalled emissions per province from the National Emission Registration (Berdowski, 1995).
The overall average emission rate amounts to 27 $g/m^2.yr$. The uncertainty in the latter is estimated to amount +/- 30%.
With a total area of 40.000 km^2 for the Netherlands, this leads to a total Dutch emission of 1.080 kton per year.
From trajectory analysis of methane measurements at Cabauw in the same period ECN estimated a total Dutch emission of 1.130 kton per year (Veltkamp et all., 1995).

This very similar totals derived from concentration measurements and using different model concepts are in fair agreement with total emissions derived from inventories.
According to van Amstel et all. (van Amstel et all. 1993) total emissions for 1991 were estimated between a minimum of 760 and a maximum of 1.730 kton per year with median value of 1.230 kton. Recent estimates according to the Dutch Emission Registration amount to 1.090 kton per year (Berdowski et all. 1995).
The agreement between emission estimates from inventories on one hand and those from ambient concentration measurements combined with atmospheric modelling on the other hand strongly supports the correctness of the present estimate of total Dutch methane emissions.

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Authentication

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M.E.J.P. Vosbeek (KEMA)

Period in which the research took place

September 1993 – April 1996

Signatures

J.C.Th. Hollander (TNO-MEP)
Project leader

Date: 13/6/96

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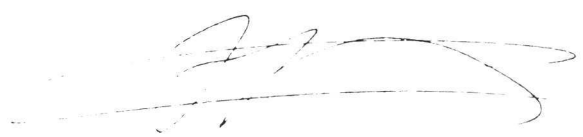

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The following reports document the research carried out in over 140 projects in the first phase of the Dutch National Research Programme on Global Air Pollution and Climate Change (1990-1995). A comprehensive overview of the programme can be found in "Climate change research: evaluation and policy implications". Proceedings of the International Climate Change Research Conference, Maastricht, The Netherlands, 6-9 December 1994 by Zwerver, S., R.S.A.R. van Rompaey, M.T.J. Kok and M.M. Berk. Elsevier Studies in Environmental Science 65A and 65B.

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