Background Concentrations for Use in the Operational Street Pollution Model (OSPM)

NERI Technical Report No. 234

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Ministry of Environment and Energy
National Environmental Research Institute
June 1998

Data sheet

Title:	Background Concentrations for Use in the Operational Street Pollution Model (OSPM)
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Department:	Department of Atmospheric Environment
Serial title and no.:	NERI Technical Report No. 234
Publisher:	Ministry of Environment and Energy National Environmental Research Institute ©
URL:	http://www.dmu.dk
Date of publication:	June 1998
Referees:	Ole Hertel and Ruwim Berkowicz
Layout:	Helle Rosvang Fomsgaard
Please quote:	Jensen, S.S. (1998): Background Concentrations for Use in the Operational Street Pollution Model (OSPM). National Environmental Research Institute, Denmark. 109 pp. - NERI Technical Report, No. 234 Reproduction is permitted, provided the source is explicitly acknowledged.
Abstract:	A background model has been developed for application in the Operational Street Pollution Model (OSPM) in context of long-term exposure modelling. The back ground model is based on a semi-empirical method founded on a few monitor stations that estimates standardised one hour time-series of urban and rural back ground concentrations of NO_2, NO_x, O_3 and CO for different geographic regions in Denmark. The annual mean of selected monitor stations is used as a reference year and the development in estimated traffic emissions as an index is used to establish a historic trend. As an exception ozone trends are based on measurements. The temporal variation is represented as indices for the monthly variation and the monthly diurnal variation. In this way concentration levels can be estimated on an hourly basis from 1960-95.
Keywords:	Urban and rural background concentrations, standardised temporal variation, inputs to OSPM, long-term exposure modelling
ISBN:	87-7772-392-9
ISSN:	0905-815X
Paper quality:	Cyclus Offset, 100% recycled paper
Printed by:	Hvidovre Kopi ApS
Circulation:	200
Number of pages:	109
Price:	DKK 125,- (incl. 25% VAT, excl. freight)

For sale at:

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Preface

The Operational Street Pollution Model (OSPM) was developed at the National Environmental Research Institute (NERI) for calculations of air pollution levels in urban streets. The OSPM is a comprehensive and useful model tool for traffic planning and for different types of assessment studies.

The model requires as input: street configuration data, emission factors and on an hourly basis data about traffic, meteorology and background concentrations. A methodology for generation of the required hourly traffic input data for OSPM based on available traffic data has been developed (Jensen 1997).

The present report describes the development of a semi-empirical background model based on standardised urban and rural background concentrations of NO_2, NO_x, O_3 and CO for different regions of Denmark for use in the OSPM in context of long-term exposure modelling.

The generated background concentration profiles has been established with the specific purpose to be used for long-term exposure assessment in the "Childhood Cancer Project" under the Danish National Environmental Research Programme. The profiles will also be used in the authors Ph.D. study concerning human exposure modelling. The semi-empirical background model is a first attempt to provide background concentration for the OSPM model for any location in Denmark.

Chapter 1 describes the context of the developed background model and chapter 2 the overall methodology. In chapter 3 an outline of the physical and chemical processes that influence background concentrations is presented to be able to understand the spatial and temporal variation in concentration levels. Chapter 4 describes the monitoring programmes that the background model is based on. Chapter 5,6 and 7 describes the background model for CO, O_3, NO_x/NO_2, respectively. Chapter 8 discusses the assumption of the background model and its potentials and limitations and in chapter 9 future research requirements are discussed. The appendices describe what is common for several of the compounds and they are equally important in understanding the presented methodology as the chapters in the main report.

Senior researchers Ruwim Berkowicz and Ole Hertel from NERI are acknowledged for valuable suggestions concerning the presented methodology. Hans Eerens from RIVM in the Netherlands is acknowledged for making available data concerning the historic development in rural and urban CO concentrations, Torben Melms from the Municipality of Copenhagen for data concerning the historic development in traffic in Copenhagen, and Spencer Sorenson from the University of Denmark for data concerning the historic development in traffic emission factors.

Summary

The background model

A background air pollution model has been developed for application in the Operational Street Pollution Model (OSPM) in context of long-term exposure modelling in the "Childhood Cancer Project". The Childhood Cancer Project is a large-scale epidemiological study of Danish children investigating the relationship between development of cancer and exposure to traffic air pollution during their childhood (Raaschou-Nielsen et al. 1996). The children's addresses may be at any location in Denmark.

The background model is a semi-empirical method based on measured urban and rural background concentrations of NO_2, NO_x, O_3 and CO from a few monitoring stations representing different geographic regions in Denmark.

The historic trends in concentration levels are estimated based on traffic emissions as an index and a reference year with the annual mean concentration level of a specific year. As an exception O_3 trends are based on measurements. The temporal variation is represented as an index where the seasonal variation is given by monthly factors and the diurnal variation by factors month by month. In this way concentration levels can be estimated on an hourly basis from 1960-95.

Taking into account only the simple photo-chemistry between NO, O_3 and NO_2, levels of NO_x are calculated in the rural setting based on measured concentration profiles of NO_2 and O_3. NO_2 and O_3 are calculated in the urban setting based on measured concentration profiles of urban NO_x and measured concentration profiles of rural NO_2 and O_3. A validation of these assumptions proved good results. CO concentrations in rural areas are estimated as a fraction of concentration levels in Copenhagen and the ratio between urban and rural background was determined based on Dutch monitoring data.

To determine concentration levels in smaller cities where monitoring data are not available a simplified dispersion formula was applied to establish a reduction factor with reference to the number of inhabitants in a city to down-scale observed concentration levels in Copenhagen to smaller cities. This method was validated for Odense and Aalborg with good results. Within a city a simple empirical formula was used to calculate the decline in concentration levels from the city centre to the outskirts.

Impact of background concentrations on street concentrations

The OSPM calculates the concentration levels in the street as a contribution from traffic emissions in the street and a contribution from the background concentrations. This implies that the impact of background concentrations will be higher for streets with little traffic compared to busy streets. Since most rural areas will be characterised by low traffic levels the impact of background concentrations on predicted street concentrations will generally be higher in rural areas compared to urban areas.

A test of the modelled background concentrations was carried out for a busy street in Copenhagen (Vignati et al. (1997)). The relationship between measured and calculated street concentrations was good (r^2=0.84). However, the model underestimates the highest hourly concentrations because the background model is based on average profiles of monthly variations and monthly diurnal variations that will not account for extreme situations. For monthly means of NO_x and NO_2 the differences between the model and measurements were less than 10 and 15 per cent, respectively. For annual means the differences for NO_x and NO_2 were less than 1 and 3 per cent, respectively. The uncertainty of the model increases for shorter averaging times (annual mean to monthly mean) and the uncertainty would increase further for even shorter averaging times (weeks, diurnal and hourly).

Application in exposure assessment

The background model has been used as part of a model system in the Childhood Cancer Project. This study also used other crude inputs like street configuration data generated from a questionnaire, and standardised traffic profiles. An evaluation of the ability of the OSPM based model system to predict observed concentration levels of NO_2 and benzene at the front-door of selected children in Copenhagen and rural areas outside Copenhagen, showed that the background model can be applied in epidemiological studies as the Childhood Cancer Project, which considers long-term exposure on at least a monthly basis (*Raaschou-Nielsen et al.* (1998)).

The relation between front-door concentrations and personal exposures to NO_2 and benzene has also been evaluated in the Childhood Cancer Project (Raaschou-Nielsen et al. 1997a, b). These studies show that the front-door NO_2 concentration is a fairly good indicator of personal exposure especially in urban areas but also in rural areas. Apart from front-door concentrations the personal exposure was also influenced by bedroom concentrations, time spent outdoors, use of gas appliances, passive smoking and burning candles. Front-door benzene concentrations was a less good indicator for personal exposure for urban areas when compared to NO_2 and misclassification would occur if applied for rural areas. Personal exposure of children was also influenced by riding in cars, exposure to gasoline vapours like motocross, moped driving and refuelling of cars.

Future research needs

A discussion of possible refinements of the presented semi-empirical background model is given. These refinements would probably improve the estimation of annual and monthly means for long-term exposure assessment.

Since the model is partly based on standard concentration profiles it will smoothen out the extreme variation in levels. This limitation needs to be improved to be able to estimate reliable time-series during shorter time periods for prediction of short-term exposure. To overcome these shortcomings, an outline for another approach is also discussed that is based on application of an existing regional background transport model (ACDEP) but with a higher resolution in the emission inventory.

1 Introduction

Childhood Cancer Project

The Childhood Cancer Project is an epidemiological study of 7,500 Danish children investigating the relationship between development of cancers and exposure to traffic air pollution during childhood (Raaschou-Nielsen et al. 1996). The project is headed by the Danish Cancer Society. Questionnaires were issued to the municipalities to provide part of the required input data for calculations with the OSPM (The Operational Street Pollution Model) about the surrounding street and traffic environment of the children's home address. The addresses may be at any location in Denmark.

OSPM

The project includes calculation of the exposure of children at their home address using the OSPM and pre-processer models for street configuration data (Vignati et al. 1997), traffic variation (Jensen 1997) and background concentrations. This part of the project is carried out by NERI.

OSPM calculates concentrations of traffic emitted pollutants in a single street. In the model, pollution concentrations are calculated as a sum of two contributions: one due to emissions from the local street traffic and one due to the pollution from other sources. Concentrations are calculated hour-by-hour as a function of the actual emissions and meteorological conditions.

Output from the model is in the form of time series of calculated hourly mean concentrations at the pavements in the street. From these time series, different statistical values can be derived: annual mean values, maximum hourly mean concentrations, percentiles etc. (Hertel and Berkowicz, 1989a,b,c; Berkowicz et al., 1997). A short description of the OSPM is presented in Vignatti et al. (1997) together with the calculation procedure for exposure calculations for the Childhood Cancer Project.

The model structure is shown in Figure 1.1,

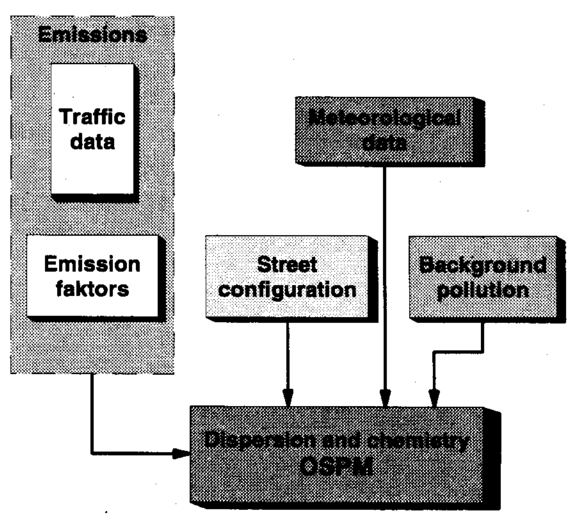


Figure 1.1 Model structure of the OSPM for calculation of air pollution levels in a street. The present report describes how background concentrations are generated hour-by-hour for any location in Denmark

Emission data are calculated in a separate subroutine, as a function of traffic and emission factors. Diurnal traffic profiles are used with differentiation between passenger cars, vans, trucks and buses. For each vehicle category an emission factor (g/km) must be specified. Emission factors depend on travel speed.

Street configuration data covers information about the street geometry and the surrounding buildings. The data required are: width of the street, orientation of the street (with respect to North), height of the buildings along the street specified for wind direction sectors with respect to the calculation point, and the length of the street.

Meteorological data are required on an hourly basis and include: wind speed, wind direction, temperature, and global radiation at roof level. Temperature and global radiation are used for calculation of NO_2.

Background concentrations are required for all the pollutants for which calculations are performed.

Scope

The scope of the present report is to describe a procedure for generation of standardised profiles on an hourly basis of urban and rural background concentrations for CO, NO_2, NO_x and O_3 for use in the OSPM. Further, the historic development of these pollutants is estimated as the Childhood Cancer Project includes cases during 1960-95.

2 Methodology

Background concentrations

Background concentrations are defined as concentrations that represent an area that is not directly influenced by the contribution from any local sources. Background concentrations have been subdivided into rural and urban backgrounds. The rural background is dominated by long-distance transport. The urban background is characterised by high local emissions and less domination of long-distance transport. The rural background concentrations are measured at ground level (approx. 3 m) and the urban background concentrations at roof top level (approx. 20 m).

Monitoring programmes

A comprehensive analysis of the spatial and temporal variation of background concentrations of CO, and NO_x, NO_2 and O_3 in Denmark has been carried out based on the available measurements in the Danish urban and rural air quality monitoring programmes managed by NERI. Annual means, trends, and monthly, weekly, and annual diurnal and monthly diurnal variation were analysed to derive at an appropriate representation of the spatial and temporal variation of background concentrations for long-term exposure assessment.

Overall methods

The following methodology has been applied to generate background concentrations:

- to account for the geographical variation between various regions in Denmark the annual means measured at selected monitor stations in the Danish Air Quality Monitoring Programmes have been used to represent different regions. The annual levels of 1994 and/or 1995 have been established as a base year for estimation of annual levels during 1960 - 1995

- to account for the development in annual concentrations since 1960 a trend factor has been established. Trends in concentration levels have been assumed to follow the development in emissions since monitoring only has been carried out in recent years. However, a trend for ozone has been established based on monitoring data

- to account for the temporal variation of concentration levels standardised monthly and diurnal profiles have been generated. The monthly variation is taking into account as the variation in monthly means represented as a factor of the annual mean. The diurnal variation is taking into account on a monthly basis and represented on an hourly basis as a factor of the monthly mean. The temporal variation is assumed to be the same for all years.

$$[C]_{hour} = [C]_{Annual} * f_{trend} * f_{month} * f_{diurnal} \qquad (1)$$

The methodology is illustrated in Figure 2.1.

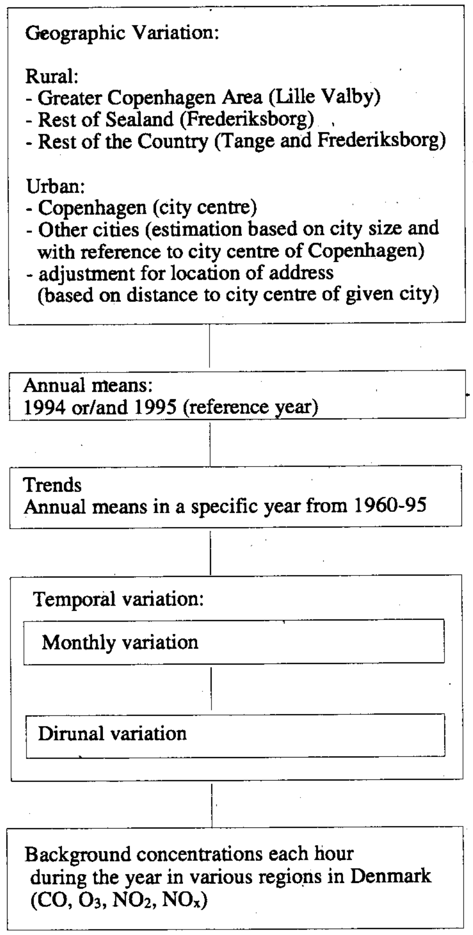


Figure 2.1 Diagram of the methodology for estimation of background concentrations

Geographical areas

The Questionnaire of the Childhood Cancer Project. Question No. 9 (category No. 1) specifies rural areas, and categories 2 - 7 specific urban areas of different city sizes (inhabitants).

Urban areas

For urban areas, Copenhagen has been singled out and the geographical extension has been defined as the area within the motorway ring. The area is defined by zip codes which are given in the Questionnaire. The calculated annual mean at the city centre of Copenhagen was used as the reference for Copenhagen.

All other urban areas are defined by the city size as given in question No. 9 categories 2 - 7.

Rural areas

Rural areas are further subdivided into the Greater Copenhagen Area, the rest of Sealand, and the rest of the country. The rest of the country includes Jutland, Funen and Bornholm. The areas are defined by zip codes. Figure 2.2 illustrates the extension of the different areas and zip codes are given in Appendix A.

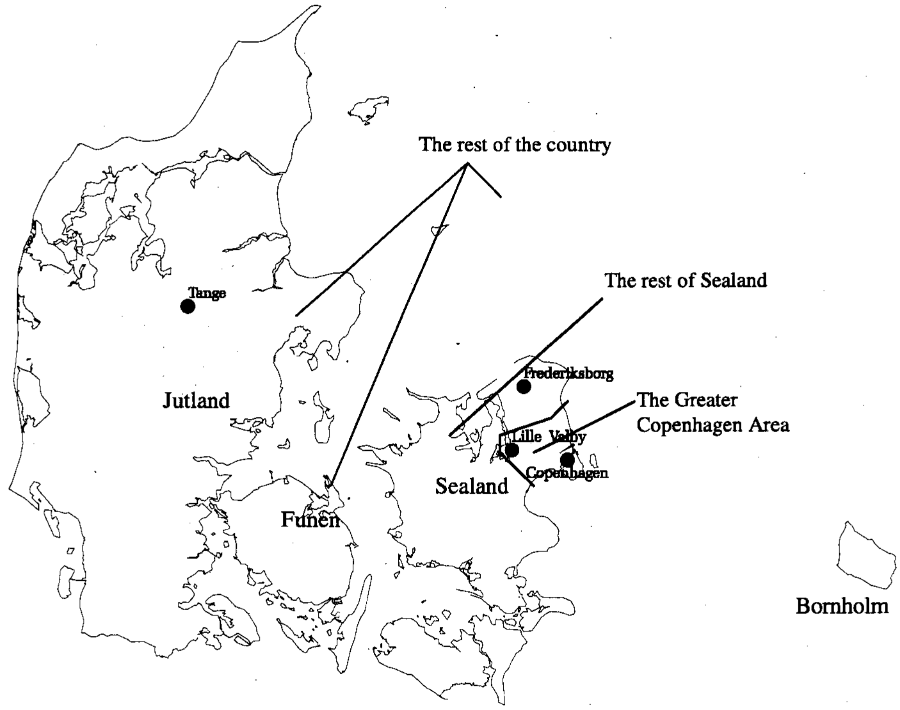


Figure 2.2 Division of the country into four regions: Copenhagen, the Greater Copenhagen Area, the rest of Sealand, and the rest of the country defined by zip codes

Calculated concentrations

NO_x in rural areas has been calculated assuming a steady state between NO, NO_2 and O_3. Details are given in Appendix B.

The urban monitoring programmes includes: Copenhagen, Odense and Aalborg but not smaller cities less than about 150.000 inhabitants. Therefore, background concentrations in other cities than Copenhagen is calculated as a fraction of the concentration levels in Copenhagen. The fraction is dependent on the No. of inhabitants. Appendix C gives details.

Measurements indicate that the urban background concentrations are higher the closer an address is located to the city centre (Raaschou-Nielsen et al. 1997). This observation is taken into account in the calculation of the urban background concentration at an address using the distance from the address to the city centre given by the Questionnaire. The method is outlined in Appendix D.

NO_2 and O_3 have also been calculated for urban areas based on the rural background concentrations of NO_2 and O_3 and assuming a steady state between NO, NO_2 and O_3, as well as, mass conservation of NO_x (NO + NO_2). Details are given in Appendix E.

The above mentioned calculations take place hour by hour in the OSPM model system for estimation of concentrations at any given address in Denmark as part of the Childhood Cancer Project.

Representative monitor stations

Table 2.1 Regions Represented by Measurements (M) and Calculations (C)

Areas:	Areas:	Station	CO	NO_x	NO_2	O_3
Urban	Copenhagen	Copenhagen (1259)	M	M	C	C
	Other cities	Fraction of Copenhagen	C	C	C	C
Rural	Greater Copenhagen Area	Lille Valby (2090)	C	C	M	M
	Rest of Sealand	Frederiksborg (2002/2082)	C	C	M	M
	Rest of the country	Frederiksborg (2002/2082)	C	C	M*	M

* Tange (6083) for annual mean of NO_2

SAS system

The SAS System, version 6.11 was used for downloading and statistical analysis of monitoring data stored in the databases of the National Environmental Research Institute. Quality assured monitoring data up to 1994 or 1995 were analysed.

Time notation

The time notation in the monitoring data is Danish Normal Standard Time (Greenwich plus one hour). The notation of hours run from 0 to 23 where 0 is from 0 to 1.

An adjustment of the normal standard time has been carried out during the summer time period to reflect the actual time since emissions follow actual time. The summer time period varies a few days from year to year. However, a standard summer period has been applied defined as the 28th of March to the 26th of September.

3 Influence of Emission and Atmospheric Processes on Background Concentrations

The concentration of a specific substance in space and time is determined by the production and loss of the substance. Concentration levels have a high spatial and temporal variation governed by emissions and atmospheric processes (physical and chemical prosesses). The interaction between these processes is complex. A brief description of the main processes is outlined based on Fenger and Tjell (1994) and Hertel (1995).

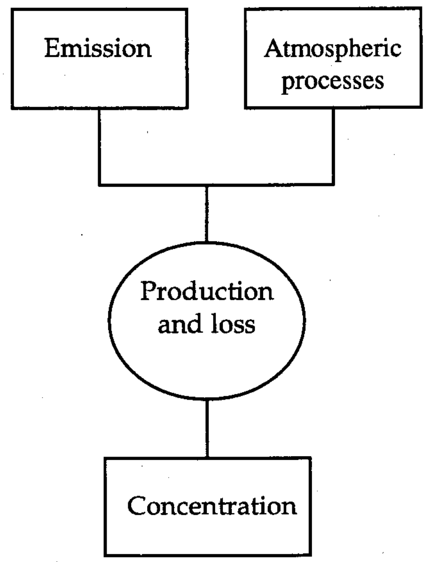


Figure 3.1 Illustration of the main factors that influence concentration levels

3.1 Emissions and Sources

Concentration levels are influenced by the geographical and temporal variation in the emissions.

In urban areas CO and NO_x ($NO + NO_2$) emissions are dominated by local road transport. The urban background consists of two contributions: local emissions and regional emissions. Concentration levels will be influenced by the emission density and the extension of the city and the transport of air pollution from the rural areas.

In rural areas concentration levels originate from regional emissions like urban areas, power plants, traffic, residential heating and other combustion processes. The emission density in rural areas is usually low and concentration levels are dominated by long-range transport.

Development in emissions from year to year depends on the economic and technological development of the society. Emissions typically exhibit a seasonal, weekly and diurnal variation due to the temporal variations in economic and social activities.

3.2 Dispersion and Transport

The meteorology influences the dispersion and dilution of emissions. The wind direction and the wind speed are the main factors. The transport distance of a pollutant depends on the lifetime which is governed by chemical and deposition processes. Depending on these factors the transport distance may be on a local scale (10 - 20 km), meso scale (- 1.000 km) or global scale (several thousands km).

Mixing layer

An important parameter that to a high degree determines the dilution is the mixing layer height. During summer days with sunlight, the thickness of the mixing layer is built up during the day from typically about 200 m in the morning to about 1,000 - 1,500 m in the afternoon. In the late afternoon it collapses. During winter days with clouds the thickness of the mixing layer is more constant over the diurnal cycle and thinner.

Turbulence

The mixing layer height is determined by the mechanical and convective turbulence. Mechanical turbulence is generated by the surface braking the wind and it is mainly depending on the wind speed and the surface roughness. The connective turbulence is generated due to heating up of the air masses close to the surface which leads to large connective eddies. The connective turbulence is mainly depending on the temperature gradient determined by the solar radiation. When atmospheric air flows are characterised as turbulent it means that the variation is chaotic and it is impossible to predict the variation from the mean to a given time.

Stability

One way to characterise the vertical movements of air masses in the mixing layer is by the stability of the atmosphere. There are three basic conditions: stable condition which decreases vertical movements, unstable condition which increases vertical movements, and neutral condition. The most unstable conditions with heavy turbulence occur with high solar radiation and low wind speeds and the most stable condition occurs during the night with low wind speeds and a clear sky with no clouds.

Impact of meteorology

Denmark covers a relatively small geographical area and has a reactively flat terrain. Therefore, the meteorological parameters like wind speed, distribution of wind direction, solar radiation etc. are rather homogeneous in time. However, the actual wind speed and direction, solar radiation etc. may vary substantially when compared hour by hour between different regions. In general, meteorological parameters can not explain variations in aggregated levels like annual means and monthly means within different regions of Denmark.

Meteorological conditions varies from year to year but annual mean concentrations do usually not vary more than 10 per cent due to meteorological conditions.

The seasonal and diurnal variation in concentrations are influenced by the meteorological conditions due to seasonal and diurnal variation of wind speeds (e.g. usually higher in spring and autumn), the thickness of the mixing layer (e.g. lower during winter and higher during summer).

Maximum concentrations are mainly determined by the meteorology and may occur under very stable conditions with little wind and solar radiation.

Special meteorological conditions

The local topography may cause special meteorological conditions. Valleys surrounded by high mountains may cause very low wind speeds and trap air pollutants. Such phenomena are not observed under Danish topographical conditions due to the flat terrain.

The topography of Denmark is characterised by large areas of waters, many islands and a long coast line. The wind speed will be higher in coastal areas compared to inland areas because the reduction of the wind speed is higher over land compared to water. Furthermore, the sea breeze phenomenon will cause coastal areas to be more windy than inland areas since air masses from the sea is transported towards the coast due to heating up of the air masses over the land by the sun. Other things equal, the nearby coastal areas will tend to have lower annual concentrations than inland areas due to the special meteorological conditions of the coastal areas.

3.3 Chemical Processes

Chemical processes may take part in the production and loss of a given substance. NO, NO_2 and O_3 take part in a complex chemistry where processes lead to both production and loss.

Ozone

The formation of O_3 is a large-scale phenomenon that takes place on a European scale. Danish annual rural and urban background levels are more or less the same and dominated by long-range transport. The seasonal variation of O_3 is therefore dominated by changes in long-range transport. Ozone levels are highest during summer.

O_3 is not directly emitted but is a secondary pollutant formed by photochemical processes with NO_x and volatile organic compounds (VOCs). Photochemical reactions in which VOCs take part in the formation of O_3 and NO_2 are of minor importance under Danish meteorological conditions.

NO, NO_2 and O_3

Under Danish meteorological conditions with relatively high wind speeds, little sunshine and low temperatures the most important reactions are between NO and O_3 forming NO_2 (1) and the photochemical dissociation of NO_2 forming NO and O_3 (2). The chemical reaction between NO and O_3 takes place within a few seconds and the photochemical dissociation takes place within minutes and is depending on solar radiation and temperature. A steady state between production and loss of NO, NO_2 and O_3 is reached in a few minutes. NO and NO_2 are produced and lost in reactions with O_3 but the sum of NO and NO_2 (NO_x) is preserved. These reactions influence the diurnal variation of NO_2 and O_3.

$$NO + O_3 \xrightarrow{k} NO_2 + O_2 \quad (1)$$

$$NO_2 + hv \xrightarrow{J} O_3 + NO \quad (2)$$

were k and J are the reaction and photodissosiation coefficients, respectively.

Reaction (2) consists of two reactions:

$$NO_2 + hv \xrightarrow{J} NO + O^{\bullet} \quad (2.1)$$

$$O^{\bullet} + O_2 \longrightarrow O_3 \quad (2.2)$$

where $O^{\bullet}$ is the oxigen radical that reacts quickly with O_2 to produce O_3.

Only a minor fraction of NO_x ($NO + NO_2$) is emitted as NO_2. From traffic it is about 5 per cent (Hertel and Berkowicz 1989b).

In rural areas, the majority of NO_x can be expected to be on the form of NO_2 as most NO has been oxidised to NO_2 in reactions with O_3. Even more important is the formation of NO_2 due to reactions between NO and various organic radicals. There is also a low NO_x emission density and therefore little NO is available for formation of NO_2. During summer the ozone levels will be high and the NO_2 levels low whereas it is the other way around in winter.

NO_2 is also lost in reactions with hydroxides (OH) that form nitric acid (HNO_3). These reactions are another reason why NO_2 levels are low during summer in rural areas as the concentration of OH various with solar radiation.

In urban areas, the NO_x emission density is higher and NO_2 is formed in reactions between NO and O_3. In the urban background the availability of O_3 will be the limiting factor for formation of NO_2. The seasonal variation in urban areas will be similar to rural areas for O_3 and NO_2. The diurnal variation of NO_2 will be more influenced by the variation in NO_x emissions which follows the traffic with morning and afternoon peaks. The variation of ozone will show the opposite pattern with high concentrations during the night, low in the morning and high during the day.

CO

CO has a long lifetime in the atmosphere (several months) but is eventually oxidised to CO_2. Chemical processes do not influence the seasonal and diurnal variation of CO.

3.4 Deposition

Substances are lost to the surface due to dry and wet deposition. Dry deposition is depending on the meteorology, characteristics of the substance and the type of deposition surface (vegetation, water etc.). Wet deposition is depending on precipitation, frequency, intensity and amount, concentration and characteristics of the substance and the presence of other substances in clouds and droplets. For wet deposition two processes are important in-cloud and below-cloud scavenging.

The wet deposition of the substances NO_2, NO_x, O_3 and CO is very limited due to the low solubility of these gases. NO_2 and O_3 are dry deposited to some extent on mainly vegetation, O_3 more than NO_2, where as the dry deposition of NO and CO is very modest. The deposition of NO_2 and O_3 is most profound on vegetation compared to urban surfaces and deposition on forest land is higher than on agricultural land.

The deposition is a minor factor in explaining differences in spatial and temporal differences between urban and rural areas. Deposition may to some extent influence difference in annual levels between forest and agricultural land.

4 Rural and Urban Monitoring Programmes

Four air quality programmes are in operation in Denmark. In the following a brief description of the programmes is given with an emphasis on the stations and pollutants of interest in this report.

LMP

The Danish Air Quality Monitoring Programme - "Det Landsdækkende Luftkvalitetsmåleprogram (LMP)" - started in 1982 and covered seven cities in the beginning. The main purpose of the programme is to monitor and carry out research related to the air quality in urban areas. The programme is co-ordinated and undertaken by NERI (Kemp et al. 1996).

Initially the programme included monitoring of pollutants (SO_2, Total Suspended Particulates (TSP)) from power plants, industry and residential heating were measured and some pollutants from traffic (NO, NO_2, lead and other elements). In recent years measurements include: O_3, SO_2, NO, NO_2, elements, meteorological parameters and campaign measurements of i.e. Polycyclic Aromatic Hydrocarbons (PAHs) and Volatile Organic Compounds (VOCs). Measurements are stored in a the so called Monitor Database. Fine particles and a number of other pollutants will be monitored in the coming years due to the implementation of EU-directives on revised limit values and monitoring requirements.

In 1996 three cities are included in the programme: Copenhagen, Odense and Aalborg and a "city-near" background site in a rural area outside Copenhagen (Lille Valby). In each of the cities measurements are carried out in one heavy trafficked street and one near by roof top location.

The urban background concentrations represented by measurements at roof tops include: O_3, meteorological parameters and campaign measurements of NO, NO_2 and CO. Measurements at the rural background station of Lille Valby include: O_3, NO and NO_2.

NO, NO_2, O_3 and CO are measured with monitors and data is stored in the so-called Monitor Database.

All stations have been used in the geographical and temporal analyses. The locations of the stations are shown in figure 4.1.

BOP and Ion Balance

The Nation-Wide Danish Monitoring Programme - "Baggrunds Overvågnings Programmet (BOP)" and "Ion Balance Programmet" - started in 1988 and covers 18 rural background stations located in different areas like forests, agricultural lands, coasts, lakes and peninsulas around the country. A few stations were in operation before 1988. The purpose of the programme is to monitor the atmospheric nutrient deposition to especially the inner Danish waters to evaluate the effect of the Danish Action Plan for the Aquatic Environment (Vandmiljøplanen) and to assess the deposition to forest and natural areas. The programmes are managed by NERI (Skov et al. 1995), (Hovmand et al. 1994).

Measurements include: atmospheric precipitation chemistry estimating various nutrients (i.e. ammonium (NH_4^+), nitrate (NO_3^-), sulphate (SO_4^{--})), gases (i.e. NO_2, ammonia (NH_3), O_3) and particles (elements). Measurements are stored in the so called Chemistry Database.

Only the stations where measurements of O_3 and NO_2 have been carried out are used in the analyses. The rural background stations include: Frederiksborg, Anholt, Tange, Ulborg and Keldsnor. The locations of the stations are shown in Figure 4.2.

Detailed information about the monitoring programmes is available on the NERI homepage at:

http://www.dmu.dk/AtmosphericEnvironment/netw.htm.

HLU

Air Quality Monitoring in the Greater Copenhagen Area is carried out by the "Hovedstadsregionens Luftovervågningsenhed (HLU)" which presently represents the counties of Copenhagen, Frederiksborg and Roskilde and the municipalities of Copenhagen and Frederiksberg. The EPA of the Municipality of Copenhagen (Miljøkontrollen) is in charge of the operation of the network. The purpose of the programme is to monitor the air quality in the Greater Copenhagen Area including Copenhagen.

Measurements started in 1967 but the number of stations, measurement techniques and selected pollutants have changed over time. For shorter or longer periods measurements have included: SO_2, NO, NO_2, O_3, CO, soot, TSP and selected elements (HLU 1990).

Presently the programme includes three street stations in Copenhagen and one rural and one urban background station (HLU 1996). The rural station is located in the north part of Sealand (Frederiksværk) and the urban station is located in the town of Køge south of Copenhagen. Measurements have included NO, NO_2 and O_3 during 1993-94 and 1991-94 for the urban and rural station, respectively. Furthermore, measurements of CO has only been carried out at the urban station. The rural station is located relatively close to (2 km) a large steel rolling mill (Stålvalseværket). The urban background station is located at a parking site close to (250 m) a large industrial and harbour area in Køge (Junckers Industrier). Meteorological parameters are measured in the town of Gladsaxe north of Copenhagen and in Køge, south of Copenhagen .

The rural station could have been used to represent rural background concentrations but the nearby stations of Frederiksborg and Lille Valby have been used for analyses of the temporal variations of concentrations. The urban station has not been used for temporal analysis since it is likely to be influenced by the nearby industrial sources.

Scope of analysis

Table 4.1 summaries the stations used in the analyses of the geographical and temporal variation of NO_2, NO_x, O_3 and CO. The type of station, database, time resolution, analytic method and monitoring periods are given.

Monitoring periods, time resolution and accuracy

Data from the Monitor Database is stored as ½ hour values. For all rural stations except Lille Valby the sampling time is 24 hours. Overall the accuracy of the stored values are within 10 per cent.

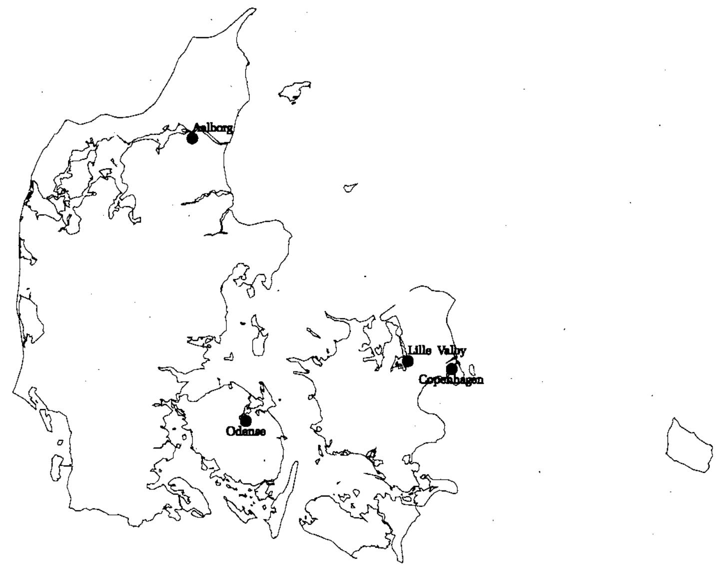


Figure 4.1 Location of monitoring stations operated within the LMP Programme. Copenhagen, Odense and Aalborg are urban background stations and Lille Valby is a rural background station

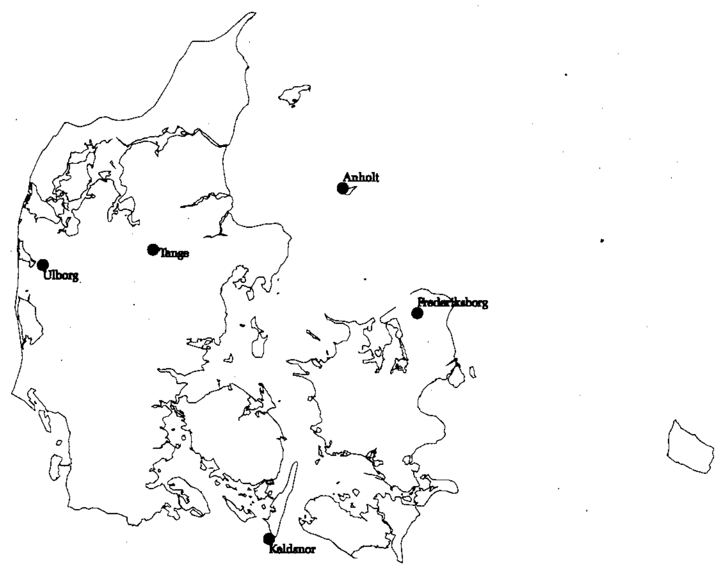


Figure 4.2 Location of selected stations from the BOP Programme. All stations are rural background stations

Table 4.1 Characteristics of the stations used in the analyses of the geographical and temporal variation of NO_2, NO_x, O_3 and CO

Substance	Station	Inhabit. x 1000	Type	Data Base	Mean values	Analytic Method	85	86	87	88	89	90	91	92	93	94	95
Rural:																	
NO_2	Frederiksborg/2082	-	Forest	Chem.	24h	KI Method					x	x	x				
	Lille Valby/2090	-	Agriculture	Moni.	½h	Chemilum.							x	x	x	x	
	Anholt/6081	-	Coast	Chem.	24h	KI Method					x	x	x		x	x	
	Tange/6083	-	Forest	Chem.	24h	KI Method						x	x				
	Ulborg/7081	-	Forest	Chem.	24h	KI Method					x	x	x	x	x	x	
	Keldsnor/9085	-	Coast	Chem.	24h	KI Method						x	x				
NO_x	Lille Valby/2090	-	Agriculture	Moni.	½h	Chemilum.							x	x	x	x	
O_3	Frederiksborg/2002	-	Forest	Moni.	½h	UV Absorption				x	x	x	x	x	x	x	
	Lille Valby/2090	-	Agriculture	Moni.	½h	UV Absorption							x	x	x	x	x
	Ulborg/7001	-	Forest	Moni.	½h	UV Absorption	x	x	x	x	x	x	x				
	Ulborg/7060	-	Forest	Moni.	½h	UV Absorption							x	x	x	x	
Urban:																	
NO_2	Copenhagen/1259	1300	Roof Top	Moni.	½h	Chemilum.									x	x	x
	Aalborg/8159	155	Roof Top	Moni.	½h	Chemiluminiscence										x	
	Odense/9159	175	Roof Top	Moni.	½h	Chemiluminiscence									x	x	
NO_x	Copenhagen/1259	1300	Roof Top	Moni.	½h	Chemilum.									x	x	x
	Aalborg/8159	155	Roof Top	Moni.	½h	Chemiluminiscence										x	
	Odense/9159	175	Roof Top	Moni.	½h	Chemiluminiscence									x	x	
O_3	Copenhagen/1259	1300	Roof Top	Moni.	½h	Chemilum.									x	x	x
	Aalborg/8159	155	Roof Top	Moni.	½h	UV Absorption								x	x	x	
	Odense/9159	175	Roof Top	Moni.	½h	UV Absorption								x	x	x	
CO	Copenhagen/1259	1300	Roof Top	Moni.	½h	IR Absorption										x	x

Chemiluminiscence: NO and NO_2 are measured using chemiluminiscent reaction between NO and O_3. The method can only determine NO. NO_x is determined by converting NO_2 to NO. NO_2 is calculated as the difference between NO_x and NO which may cause large uncertainties in conditions with low NO_2 concentrations. Mean half hour values are stored.

UV Absorption: O_3 is measured with monitors using the principle of UV absorption. Monitors are installed in Copenhagen, Aalborg and Odense and in Lille Valby, Frederiksborg and Ulborg. Mean half hour values are stored.

KI Method: NO_2 is sampled on sintered glass filters impregnated with a solution containing KI and $NaAsO_2$ which reduces NO_2 to NO_2^- (nitrite). The concentration of nitrite is determined after extraction with water and reactions with Saltzmann's reagens by spectrometric determination. The sampling period is 24 hours and values are stored as 24 h values.
IR Absorption: CO concentrations are measured with a monitor using the principle of absorption of infra red light. Half hour values are stored.

DOAS: In recent years DOAS (Differential Optical Absorption Spectrometry) has been implemented as an analytic method. DOAS is able to measure many pollutants at the same time taking advantage of the difference in absorption of light of different pollutants. The concentration of the pollutants are measured many times every second over a longer distance using a light beam. Mean half hour values are stored.

5 Carbon Monoxide

In this chapter the annual mean, trends and temporal variation of urban and rural CO background concentrations are derived.

Table 5.1 Regions represented by: Measurements (M) and Calculations (C)

Areas:	Areas:	Station	CO
Urban	Copenhagen	Copenhagen (1259)	M
	Other cities	Fraction of Copenhagen	C
Rural	Greater Copenhagen Area	Fraction of Copenhagen	C
	Rest of Sealand	Fraction of Copenhagen	C
	Rest of the country	Fraction of Copenhagen	C

5.1 Urban Background

Annual Mean

CO has been measured continuously at the urban background station of Copenhagen since January 1994.

Table 5.2 Annual Mean CO Concentration at the Urban Background Station of Copenhagen

Station	Type	Period	Mean (ppm)
Copenhagen/1259	Roof top	1-8,12,1994	0.28
Copenhagen/1259	Roof top	1-12,1994*	0.31
Copenhagen/1259	Roof top	1-12, 1995	0.34

* The annual mean of 1994 has been estimated based on the assumption that the months of September to November constitute the same fraction of annual mean as for 1995.

The Copenhagen monitor station (1259) has been used as indicator station for Copenhagen and the annual level of 0.31 ppm in 1994 as a reference base year.

Dependence on City Size

Since the monitor programmes only cover the largest Danish cities it has been necessary to develop an extrapolation method to estimate the urban background concentrations in smaller cities. The reference for the method is the concentration levels observed in Copenhagen (1259) and this level is scaled down using the method applied in *Hertel and Berkowicz* (1990). Estimated concentration levels have been normalised with respect to the levels in Copenhagen. The method estimates background concentrations for area sources of known emission density based on the assumption that the emissions are evenly distributed and that the dispersion is linearly depending on the dispersion distance (city diameter). The emission density is obtained from *Bendtsen and Reiff* (1996) and the city diameter from maps for selected cities. The method is described in details in Appendix C.

Dependence on Distance from City Centre

Measurements carried out as part of the Childhood Cancer Project indicate that the urban background concentrations are higher the closer an address is located to the city centre. This observation is applied empirically in the calculation of the urban background concentration at an address using the distance from the address to the city centre given by the Questionnaire. The method is based on *Raaschou-Nielsen et al.* (1997) and outlined in details in Appendix D.

Trends

Continuous CO measurements have been performed for Copenhagen since January 1994. Obviously, the record is too short for trends analysis.

Campaigns

Campaign measurements of CO were carried out in backyards to main streets in Lyngby in the north part of Copenhagen during March and April 1986 and in Aalborg during May to July 1986. Mean levels were 0.9 ppm and 0.4 ppm for Lyngby and Aalborg, respectively (Bendtsen 1989, Rokkjær 1986).

An analysis of the monthly variation at the monitor station of Copenhagen (1259) shows that the levels in March and April in 1994 are close to the annual mean, that is, the annual mean for Lyngby is likely to be approx. 0.9 ppm.

Since the annual mean is 0.31 ppm in Copenhagen (1259) in 1994 and it is estimated to be 0.9 ppm in Lyngby north of Copenhagen in 1986 this indicates a drastically decrease from 1986 to 1994. The difference does not reflect differences in meteorology because the variation in e.g. NO_x levels from year to year is minor.

The annual mean in Aalborg is estimated to 0.6 ppm assuming the same monthly variation as in Copenhagen (1259). This level is about 50 per cent lower than the annual mean estimated in Lyngby north of Copenhagen.

The number of Danish CO campaigns is insufficient to establish a trend but the data indicate that CO levels have dropped substantially during the last decade.

Dutch monitoring data

Urban CO background concentrations have been measured for a longer period in the national monitoring programme in the Netherlands. Data were obtained from the RIVM for five urban monitor stations for the period 1989-1995, see Figure 5.1. The trend shows a general increase in levels until 1991 followed by a general decrease which probably is due to increase in catalyst cars. The Dutch data show a decrease in CO levels within recent years although it is less profound compared to Danish data.

The available data are insufficient to generate a realistic trend in background concentrations since 1960. In the absence of measurements, the development in emissions is the best indicator for trends in background concentrations.

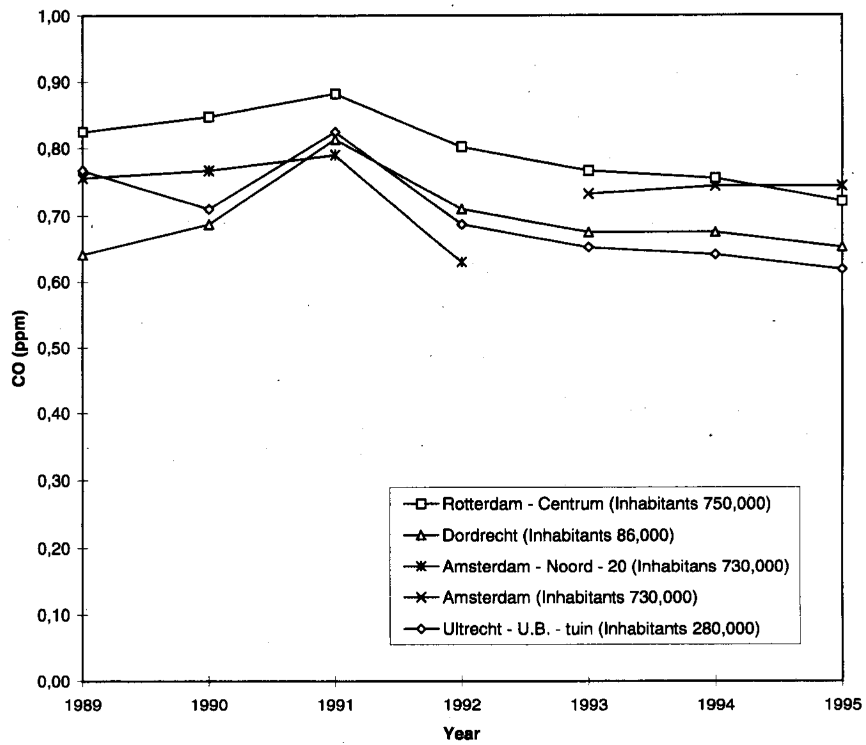


Figure 5.1 Trends in CO concentrations in the Netherlands for urban background stations

National emission trends

The national CO emission inventory for road transport shows that CO emission has varied moderately from year to year during the period 1972-1990. During the period there has been an increase of 5 per cent. During the same period national emissions from all sources increased by about 30 per cent (Fenhann, Kilde 1994). A decline in CO emissions from road transport of about 15 per cent has taken place from 1990 to 1993 due to the introduction of cars with catalytic converters since 1990 (Danmarks Statistik, 1995). The national inventory does not support the profound decline in background levels indicated by the campaign measurement.

However, the inventory has been established based on the energy consumption and applying constant CO emission factors (CO/energy) during 1972-89 and revised emission factors during 1990-94 (Sorensson, private communication). The assumption that CO emission factors should have been constant for 1972-89 is fare too simplistic.

Further, a national road CO emission inventory is likely to be a poor indicator for developments in CO emissions in urban areas since development in traffic is different in central urban areas and on a national scale. A study of the development in traffic levels of 34 representative urban streets in central parts of cities in Denmark with more than 10.000 inhabitants, indicates that traffic levels in general have been more or less constant in central urban areas during 1985 - 1992. During the same period, traffic levels have increased about 30 per cent on the main road network (Bendtsen et al., 1994). In October

1990 catalytic converters became mandatory on new petrol-powered passenger cars and small vans. The combination of constant traffic levels in central parts of urban environments and more stringent emission standards should contribute to lower CO background levels since 1990. The national monitoring programme in the Netherlands also shows a decrease in urban background concentrations in this period.

During the last two decades district heating has replaced most of local residential and commercial heating in Copenhagen which also contributes to lower background concentrations.

Urban emission trends

The development in CO traffic emissions in central Copenhagen has been estimated and applied as an indicator for the trend in urban background concentrations of CO based on the actual development in traffic and emission factors. Traffic performance and traffic composition are based on data from the Municipality of Copenhagen. As indicators for the development, traffic counts carried out at the arterial roads to the central parts of Copenhagen are used ("Søsnittet" and "Havnebroerne"). The basic CO emission factors for the reference year 1993 have been established based on a study from the Danish Road Directorate (Vejdirektoratet 1992). The development in emission factors is based on data from the Laboratory for Energetics, Technical University of Denmark (Sorenson, private communication). Refer to Appendix F for details.

The development in urban traffic emissions in Copenhagen is shown in Figure 5.2. The trend in emissions is used as an indicator for the development in urban background concentrations for all urban areas.

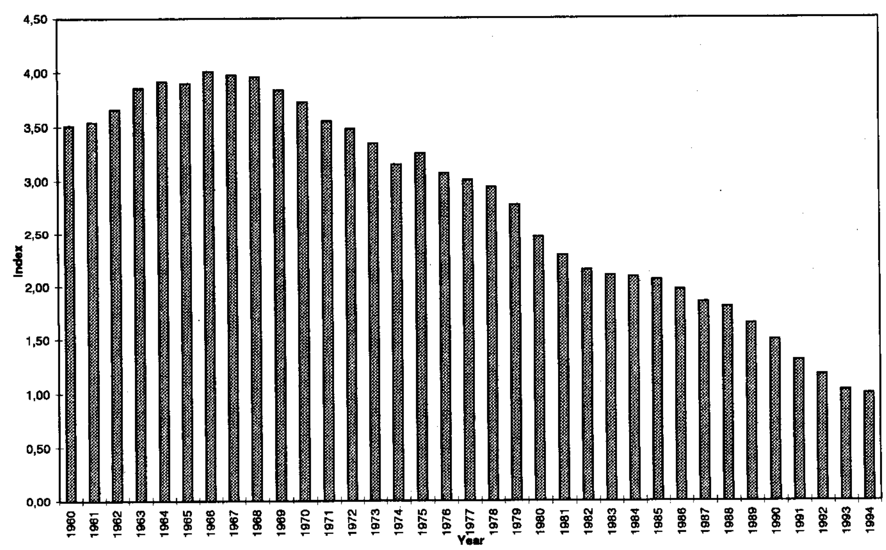


Figure 5.2 Development in CO traffic emissions from 1960 to 1994 in central parts of Copenhagen (index with 1994 as reference year)

CO emissions decreased about 50 per cent from 1986 to 1994, see Figure 5.2. The few Danish campaigns indicate that CO concentrations have dropped about 50-67 per cent during the same period. Based on the limited data it seems likely that the development of CO emissions is a fair indicator of trends in CO concentrations.

Monthly variation

The monthly variation in Copenhagen based on a complete data set from 1995 is shown in Figure 5.3 together with the monthly variation in passenger cars.

There seems to be a distinct seasonal variation in CO concentrations with high levels during winter and low levels during summer.

The seasonal variation in passenger car traffic shows a slightly different pattern with little less traffic during the winter months, more traffic during spring and autumn and a slight drop in traffic during the holiday month of July (Jensen, 1997). Passenger cars have been chosen as an indicator for CO emissions since they dominate CO emissions from vehicles. At national level, sources of CO emissions are dominated by road transport (70 %) and other sources like residential and commercial heating (20 %). In larger urban areas where district heating is wide spread CO emission from road transport is even more dominating.

The different seasonal pattern for CO background concentrations compared to traffic is due to differences in meteorology between winter and summer with typically more shallow mixing layers during winter, and more CO emissions from other sources than traffic during the heating period in the winter months.

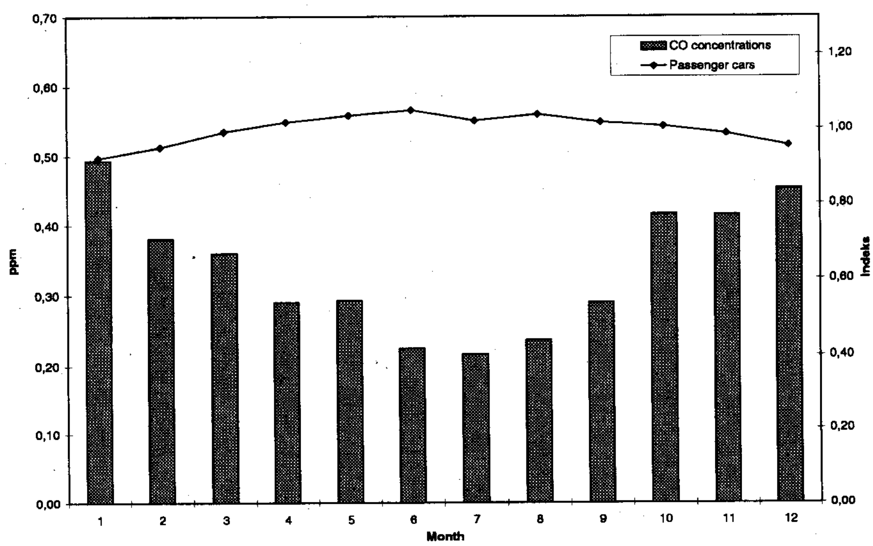


Figure 5.3 Monthly variation in urban CO background concentrations (ppm) in Copenhagen (1259) during 1995 and monthly variation of passenger cars (index)

Monthly variation in diurnal variation

In Figure 5.4 the monthly diurnal variation is shown for Copenhagen. The diurnal variation is similar for the various months although the levels are different. The diurnal variation with high levels in the morning and afternoon is similar to the diurnal variation in traffic , see e.g. (Jensen 1997).

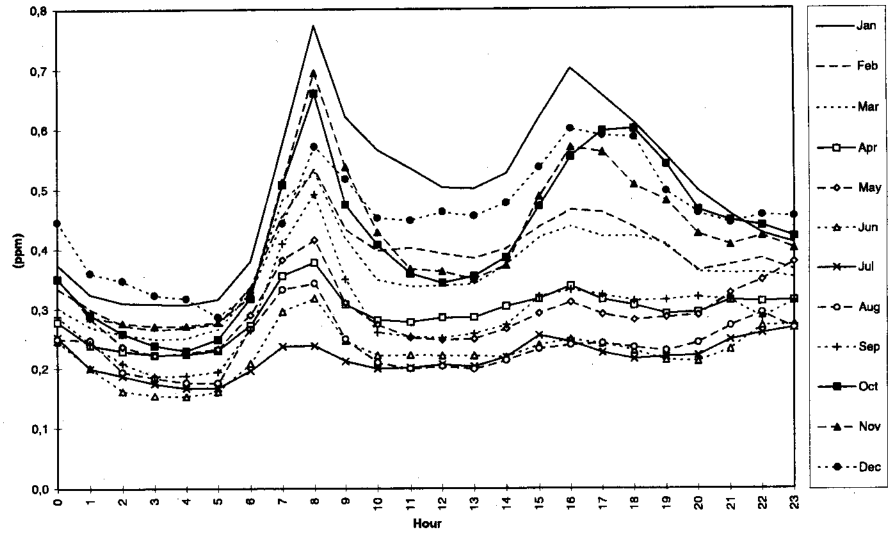


Figure 5.4 Monthly diurnal variation for urban CO background concentrations in Copenhagen (1259) during 1995

Weekly variation

Figure 5.5 shows the weekly variation in CO levels. The levels during the week-end are about 15-20 per cent lower than during the working days probably due to the fact that there is less traffic during the week-end (Jensen 1997). To limit the number of calculations the minor weekly variation has not been taken into account.

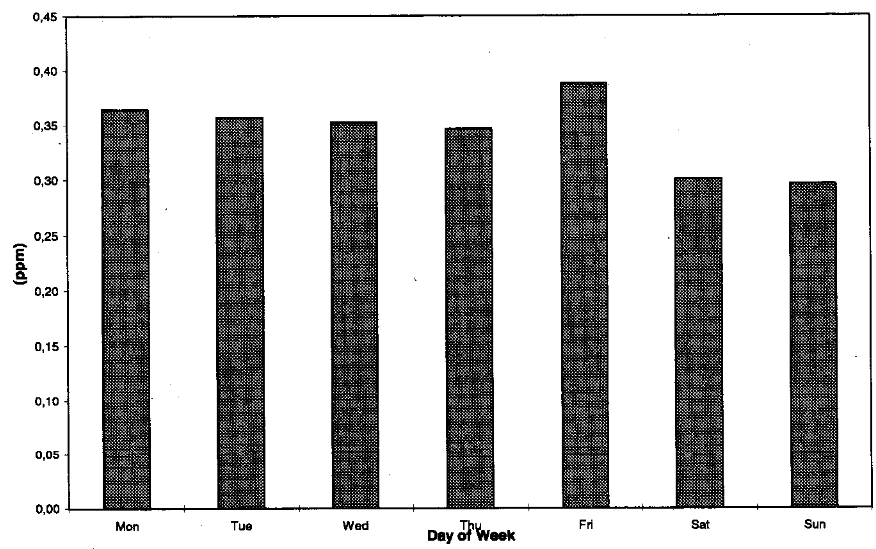


Figure 5.5 Weekly variation in urban CO concentrations in Copenhagen (1259) during 1995

5.2 Rural Background

Geographic variation

No measurements are available for CO in Danish rural areas. In the clean troposphere the levels is about 0.12 ppm which corresponds to very remote rural areas (Seinfeld, 1986). In Denmark the levels in rural areas are likely to be higher due to a relatively high density of roads and other CO emission sources. Furthermore, the lifetime of CO is quite long and long-range transport will dominate in rural areas.

Data from the Dutch monitoring programme showed that rural CO levels are about half of urban levels. The same ratio between urban and rural levels have been applied for the Danish rural background corresponding to 0.15 ppm as an annual level in 1994.

Monthly and diurnal variation

No measurements are available for analysis of the diurnal variation in rural areas. Traffic is the dominating source of CO emissions and a distinct traffic related diurnal variation is seen for the urban background concentrations. However, the influence of traffic emissions will be less pronounced in rural areas and a more level diurnal variation may be expected. Despite these considerations the monthly and diurnal variation in rural areas are simply assumed to be similar to the urban areas.

Trends

Monitoring has only been carried out in recent years, therefore, the development in national road emissions have been taken as an indicator for the trends in concentrations in rural areas. Road CO emissions account for about 70 per cent of national emissions. The development in CO emissions has been calculated from the development in national traffic performance and emission factors (g/km). See Appendix G for details.

The trend in CO emissions is shown in Figure 5.6.

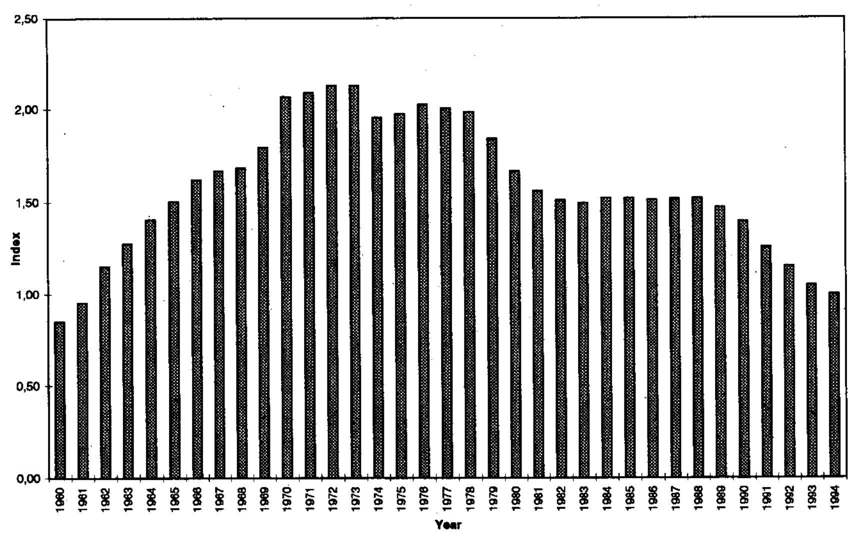


Figure 5.6 The Development in National Road CO Emissions as Indicator for Trends in CO Concentrations in Rural Areas.

6 Ozone

In this chapter the annual means, trends and temporal variation of O_3 are derived.

Table 6.1 Regions represented by: Measurements (M) and Calculations (C)

Areas:	Areas:	Station	O_3	NO_x	NO_2
Urban	Copenhagen	Copenhagen (1259)	C	M	C
	Other cities	Fraction of Copenhagen	C	C	C
Rural	Greater Copenhagen Area	Lille Valby (2090)	M	C	M
	Rest of Sealand	Frederiksborg (2002[b]/2082[c]), Lille Valby (2090[d])	M	C	M
	Rest of the country	Frederiksborg (2002[b]/2082[c]), Lille Valby (2090[d])	M	C	M[a]

a Tange (6083) for annual mean of NO_2
b Used for O_3
c Used for monthly variation of NO_2
d Used for monthly diurnal variation of NO_2

Large-scale phenomenon

Wind direction distributions of concentrations measured at the urban background O_3 monitoring stations are similar for both urban and rural background stations, see Figure 6.1. This indicates that O_3 is a large-scale phenomenon dominated by long-range transport which also accounts for O_3 episodes.

6.1 Urban Background

Geographic variation

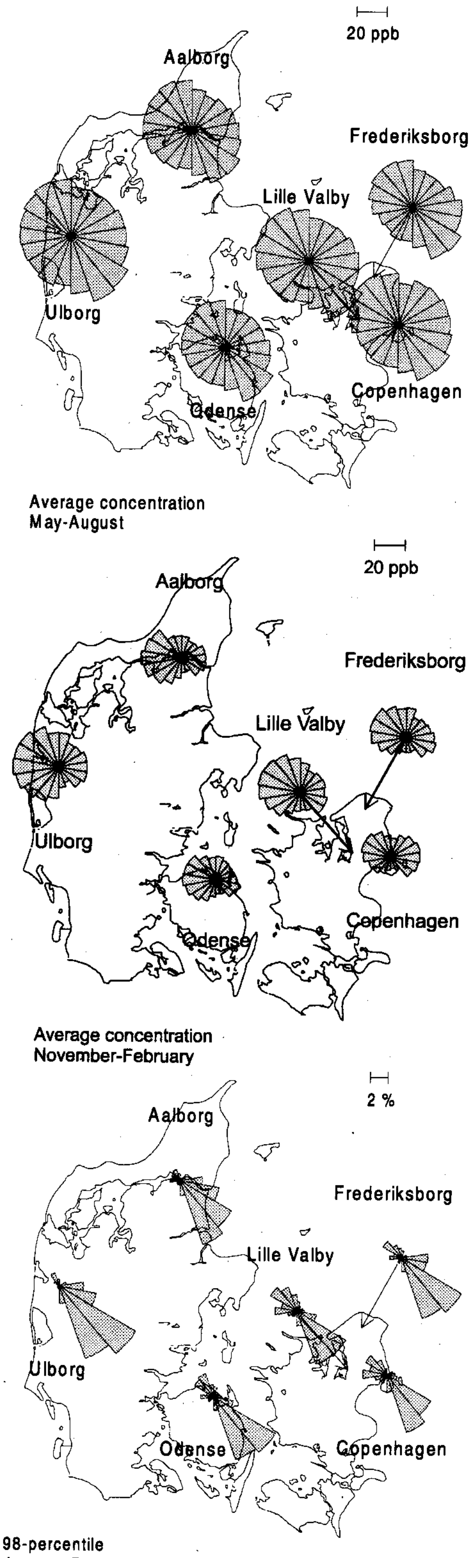
The annual means of the urban background stations are presented in Table 6.2. The levels seem to be the same in Copenhagen and Aalborg with slightly lower levels in Odense. The urban background levels are only slightly lower than the rural background levels (compare Tables 6.2 and 6.3) because of depletion of ozone due to local urban emissions of NO_x.

Table 6.2 Annual Means of O_3 (ppb) at Various Urban Background Stations

Station	Type	Period	1992	1993	1994	1995
Copenhagen (1259)	Roof top	4.93-12.95	-	(25.4)	25.5	22.7
Odense (9159)	Roof top	8.92-12.94	(16.4)	21.4	24.7	-
Aalborg (8159)	Roof top	12.92-12.94	(8.6)	24.6	25.4	-

* Years with limited observations are given in brackets.

Figure 6.1 Wind roses for hourly O_3 concentrations measured in 1993 and 1994



Upper and middle: The radii of the circle sections are proportional to the average concentrations for wind directions corresponding to the section

Lower: The radii of the circle sections are proportional to the number of hourly measurements in 1993 and 1994 above the 98-percentile relative to the number of hourly measurements in the specific sectors from *Granby et al. (1996)*

The geographic variation in O_3 levels at the various urban stations are minor. The differences between urban and rural O_3 levels are also minor (compare Tables 6.2 and 6.3). Therefore, the difference between different cities is expected to be insignificant.

For Copenhagen and other cities O_3 levels are calculated taking into account the observed NO_x levels at the city centre of Copenhagen and the rural levels of O_3 and NO_x, see Appendix E. NO_x levels are predicted in smaller cities as outlined in Appendix C. NO_x levels at an address are also influenced by the distance to the city centre, see Appendix D.

Trends

The longest Danish O_3 record for urban background concentrations started in August 1992 and no trends can be derived for such a short time series.

In the absent of measurements, the trends in urban background levels is assumed to follow the same trends as the rural background concentrations, see the following section 6.2.

Temporal variation

Appendix E gives a comparison between observed and calculated levels for the monthly variation, the weekly variation and the annual diurnal variation.

Urban background stations have almost the same monthly variation as the rural stations, and only slightly lower levels.

6.2 Rural Background

Geographic variation

The annual means of the rural background stations are presented in Table 6.3.

Table 6.3 Annual Means of O_3 (ppb) at Various Rural Background Stations

Station	Type	Period	85	86	87	88	89	90	91	92	93	94	95	Mean**
Lille Valby (2090)	Rural	06.91-12.94	-	-	-	-	-	-	(23.7)	26.4	25.9	28.2	25.7	26.6
Frederiks-borg (2002)	Forest	07.88-12.94	-	-	-	(23.1)	26.7	21.1	20.2	24.1	22.1	24.5	-	23.1
Ulborg (7001)*	Forest	09.85-05.91	(25.8)	28.4	26.3	28.7	27.0	24.0	(25.3)	-	-	-	-	26.9
Ulborg (7060)*	Forest	05.91-09.94	-	-	-	-	-	-	(32.0)	31.1	29.7	(33.3)	-	30.4

*The station became elevated in 1991 and renamed to Ulborg (7060). The general increase in levels since 1991 is probably due to less deposition of O_3 in the elevated height of the forest. Years with limited observations are given in brackets. ** Only for full annual records.

The levels are slightly higher in Ulborg compared to Lille Valby and Frederiksborg since Ulborg is not influenced by O_3 depletion due to NO_x emissions from the Copenhagen area.

In *Granby et al.* (1997), a comparison between Lille Valby and Frederiksborg based on linear regression analysis on data from 1993 and 1994 shows a strong correlation with $r^2 = 0.89$. The levels and distribution are similar at the two stations, and they are similarly influenced by NO_x emissions from Copenhagen. Levels are slightly lower at the forest station of Frederiksborg compared to the rural Lille Valby station probably due to a higher dry deposition of O_3 on forest compared to agricultural land.

A similar comparison between Lille Valby and Ulborg reveals less pronounced correlation although the correlation still is good with r^2 = 0.70 according to *Granby et al.* (1997).

Lille Valby is selected to represent rural background concentrations of O_3 in the Greater Copenhagen Area. The average concentration 26.9 ppb during 1994-95 is used. Frederiksborg is selected to represent the rest of Sealand and the rest of the country with the annual mean 24.5 ppb in 1994 as a reference. Ulborg has not been selected to represent the rest of the country since it is located close to the coast.

Trends

Ulborg has the longest Danish O_3 time serie starting in 1986, see Figure 6.2. The observations reveal no clear trends. Analysis of shorter records of Lille Valby and Frederiksborg show no clear trends either.

A long background O_3 record from 1976 measured at Zugspitze (2,962 m) in the southern part of Germany shows that the increase in O_3 levels seems to cease in the mid 80'ties, see also Figure 6.2. Based on these observations, Danish O_3 levels are assumed to be constant from 1986.

The longest background O_3 record in Europe is from Arkona in Germany located on the island Rügen in the Baltic Sea. Annual means have increased from 15 ppb to 24 ppb from 1956 to 1983 with an annual increase of 0.35 ppb, see Figure 6.2.

Arkona is assumed to represent the Danish O_3 trends from 1960 to 1985 since it is located very close to Denmark. Estimated developments in O_3 levels in Denmark are shown in Figure 6.3. The average annual level at Lille Valby from 1994 - 95 and the annual mean from 1994 at Frederiksborg are assumed to be representative for the period 1986 - 95. Extrapolations for the period 1960 - 85 are based on an annual increase of 0.35 ppb as for Arkona.

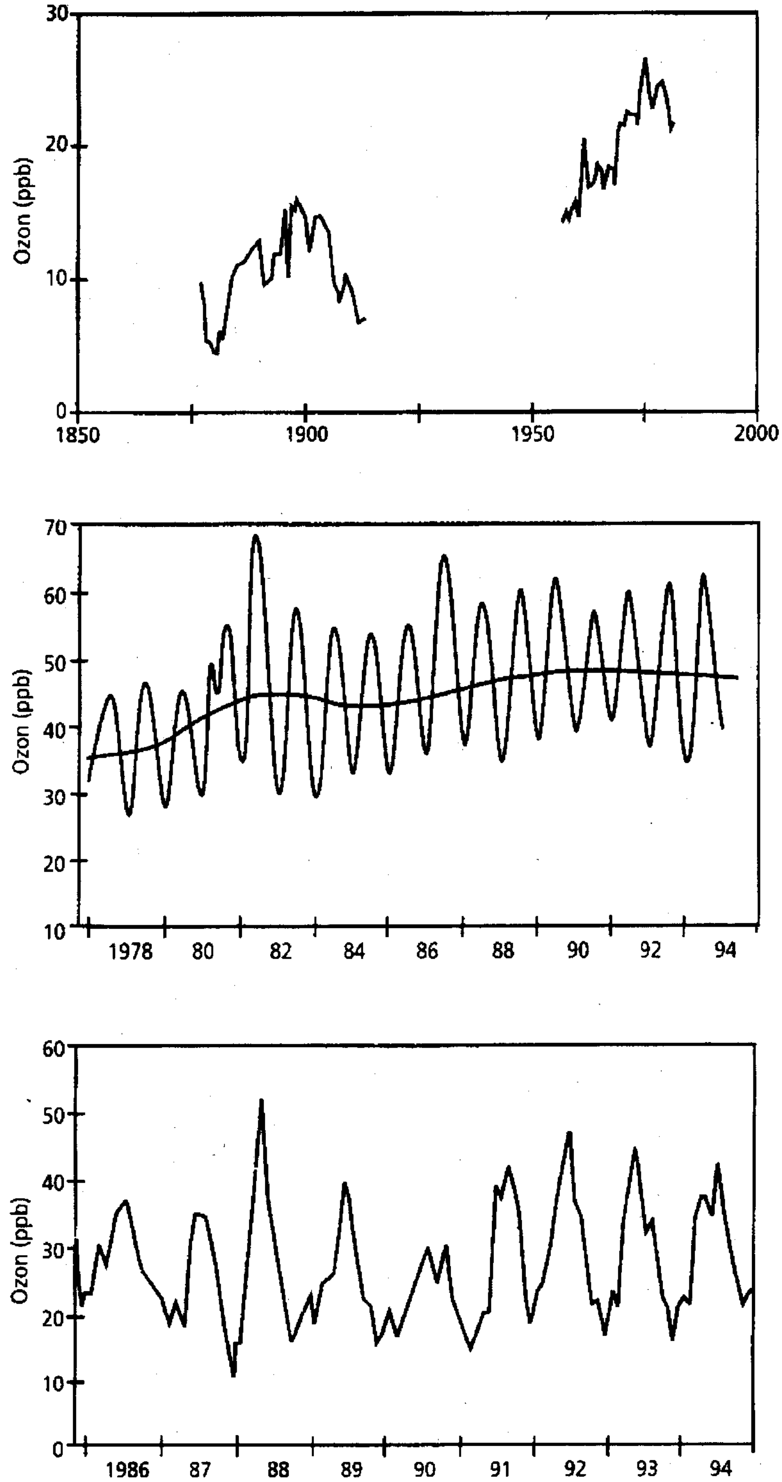
Figure 6.2

Upper: the longest background O_3 record in Europe from Arkona in Germany located on the island Rügen in the Baltic Sea

Middle: a background O_3 record from 1976 measured at Zugspitze (2962 m) in the southern part of Germany

Lower: the longest O_3 record starting in 1986 in Denmark from Ulborg.

The figure is from *Fenger (1995).*



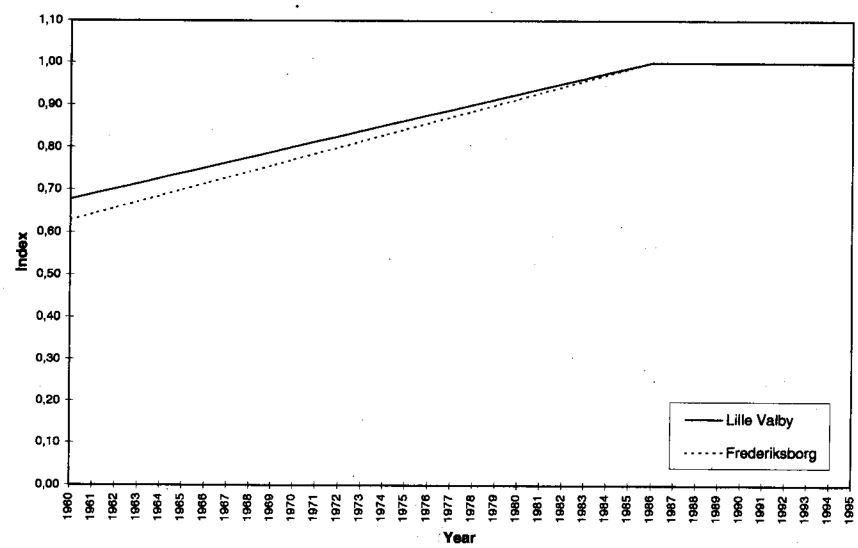


Figure 6.3 Estimated developments in O_3 concentrations for Lille Valby and Frederiksborg during 1960-95

The increase in O_3 levels during this century is caused by the general increase in emission of NO_x and VOCs on a regional scale. The stagnation in O_3 levels during the 80'ties and 90'ties correspond to a more or less constant emission of NO_x and VOCs during the same period.

Monthly variation

The monthly variations from year to year are by and large similar for Lille Valby, Frederiksborg and Ulborg, compare Figures 6.4, 6.5 and 6.6. Generally, the highest monthly mean concentrations are found in May, June or July. An important meteorological factor that influences the seasonal variation of O_3 is solar radiation which is highest during summer.

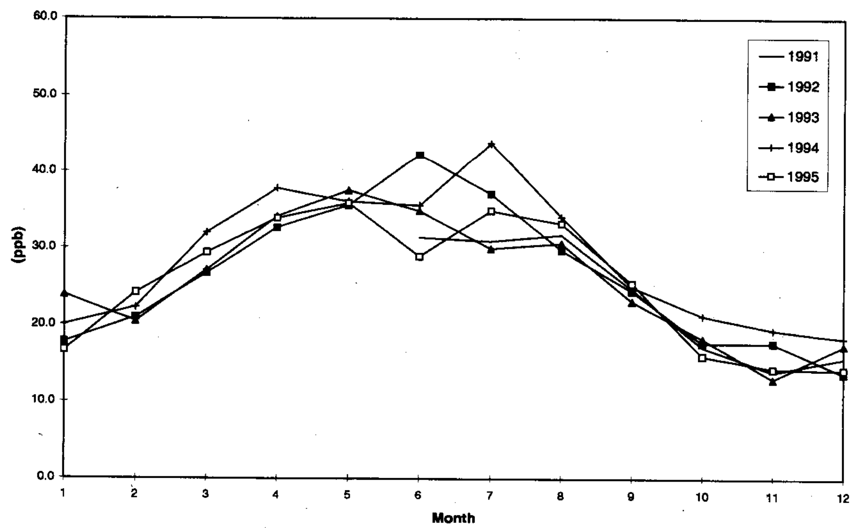


Figure 6.4 Monthly variation in O_3 during 1991-95 at Lille Valby (2090)

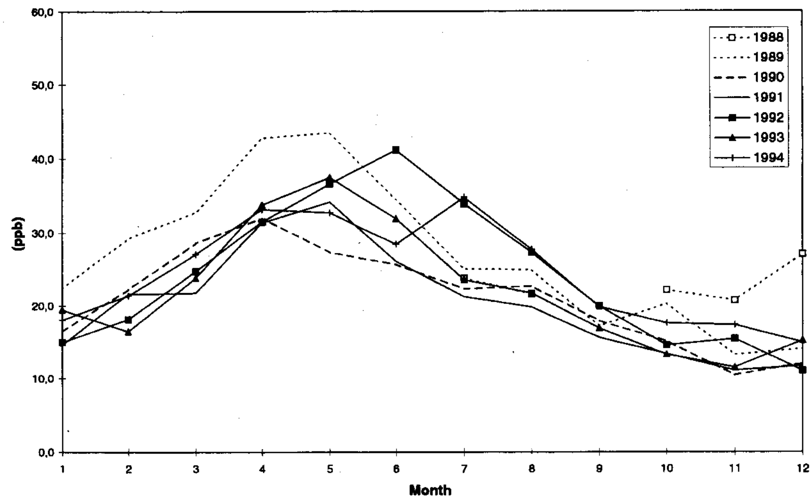


Figure 6.5 Monthly variation in O_3 during 1988-94 at Frederiksborg (2002)

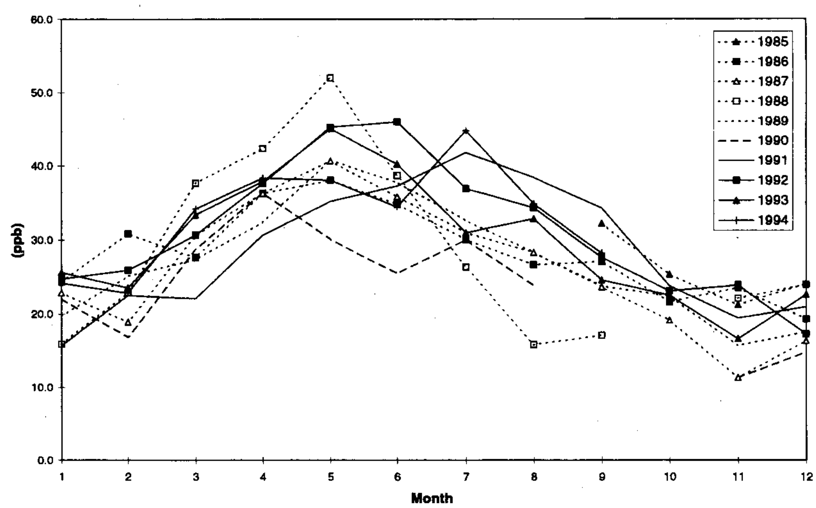


Figure 6.6 Monthly variation in O_3 during 1985-94 at Ulborg (7001 and 7060)

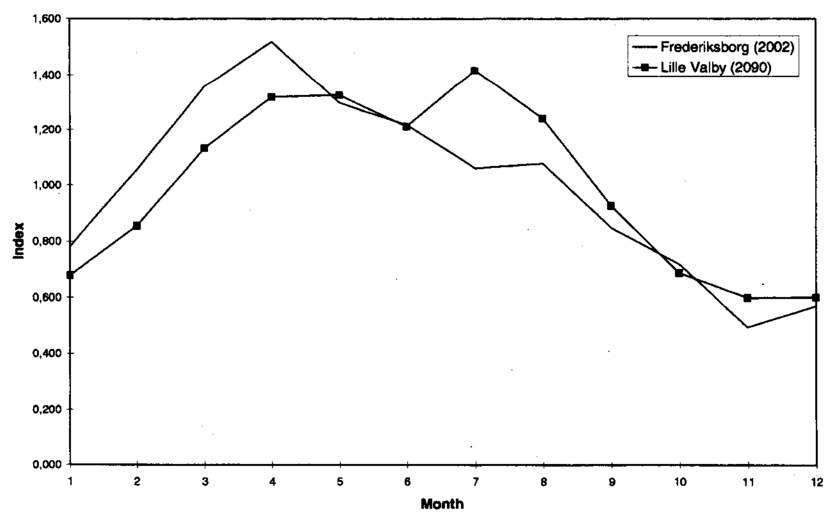


Figure 6.7 Monthly O_3 variation used to represent the Greater Copenhagen Area (Lille Valby 1994-95), the rest of Sealand and the rest of the country (Frederiksborg 1990)

To obtain an average monthly profile the time serie of Lille Valby from 1994 - 95 will be used to represent the Greater Copenhagen Area, and the record for Frederiksborg from 1990 is used to represent the rest of Sealand and the rest of the country, see Figure 6.7.

Diurnal variation

The annual diurnal variations from year to year are similar for the stations: Lille Valby, Frederiksborg and Ulborg although the levels differ. Consequently, the diurnal variation is assumed to be the same for the various years. As an example the annual diurnal variation is given for Frederiksborg (2002) in Figure 6.8 where 1990 seems to represent an average diurnal variation.

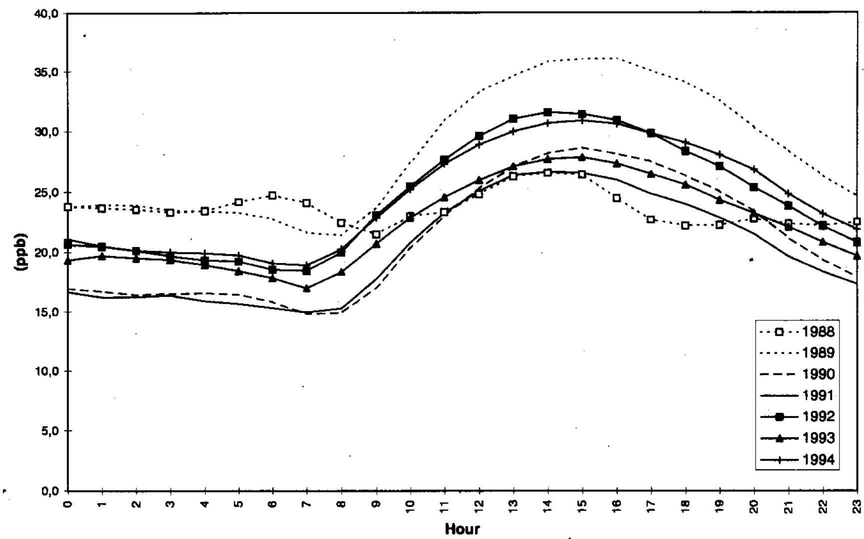


Figure 6.8 The annual diurnal variation of O_3 during 1988 - 1994 at Frederiksborg (2002)

Ozone is formed during the day due to photochemical dissociation of NO_2 caused by solar radiation. Local photochemical O_3 formation due to VOCs is of less importance in Denmark. Although the solar radiation is highest in the middle of the day O_3 concentrations reach a maximum in the afternoon. This is due to O_3 entrainment from the free troposphere into the mixing layer heights when it is deep in the afternoon. It is also due to dilution of NO_x because of a typically higher mixing layer height and higher wind speeds in the afternoon leaving less NO for depletion of O_3. During the night the O_3 levels are low and constant since the O_3 is depleted by the available NO and there is no production by photodissociation of NO_2.

Monthly variation in diurnal variation

There is a monthly diurnal variation, see Figure 6.9. The diurnal variation is weak during winter with generally low levels and strong during summer with generally high levels. The weak diurnal variation during winter is due to low solar radiation and a shallow mixing layer with minor entrainment of O_3 from the troposphere.

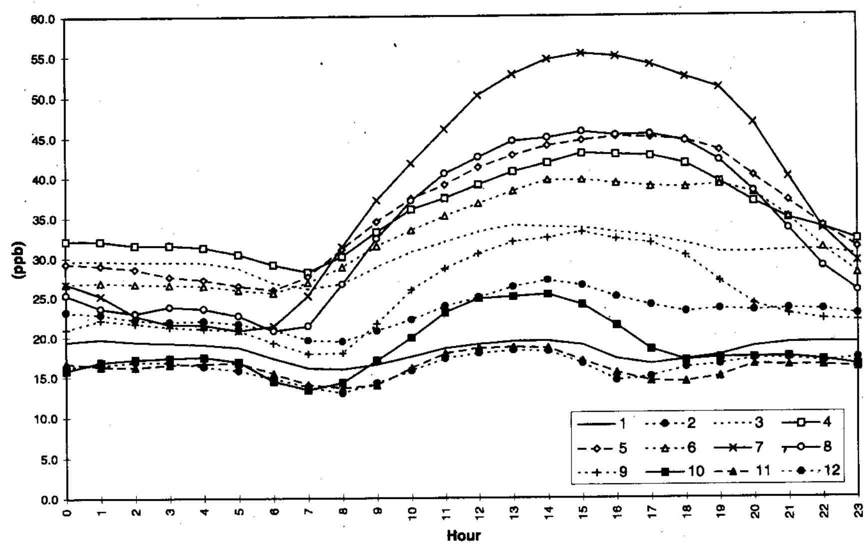


Figure 6.9 The monthly diurnal variation of O_3 in ppb at Lilly Valby (2090) as an average during 1994-95 used to represent the Greater Copenhagen Area

The average diurnal variation of Lille Valby for the period 1994 - 95 is used as a standard diurnal variation for the rural background concentrations in the Greater Copenhagen Area and the monthly diurnal variation at Frederiksborg during 1990 is used for the rest of Sealand and the rest of the country, see Figures 6.10 and 6.11.

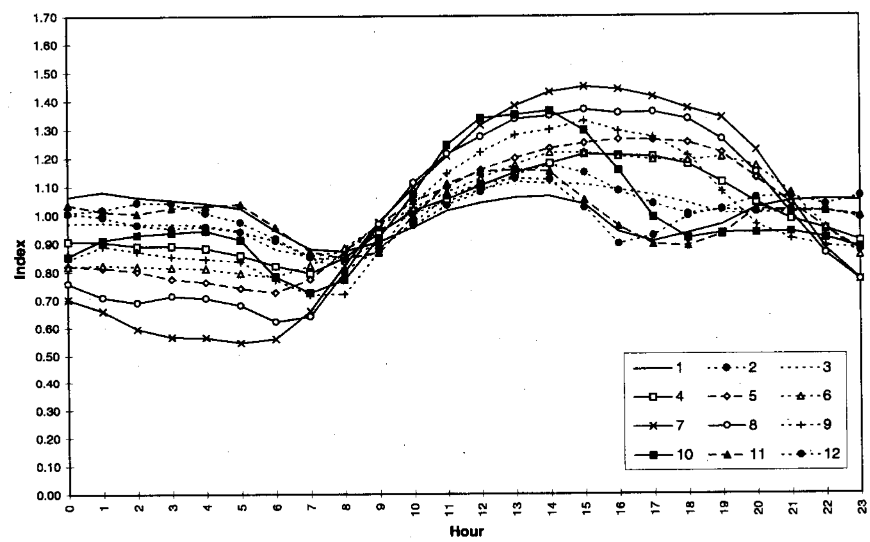


Figure 6.10 The monthly diurnal variation of O_3 as an index at Lilly Valby (2090) as an average during 1994-95 used to represent the Greater Copenhagen Area

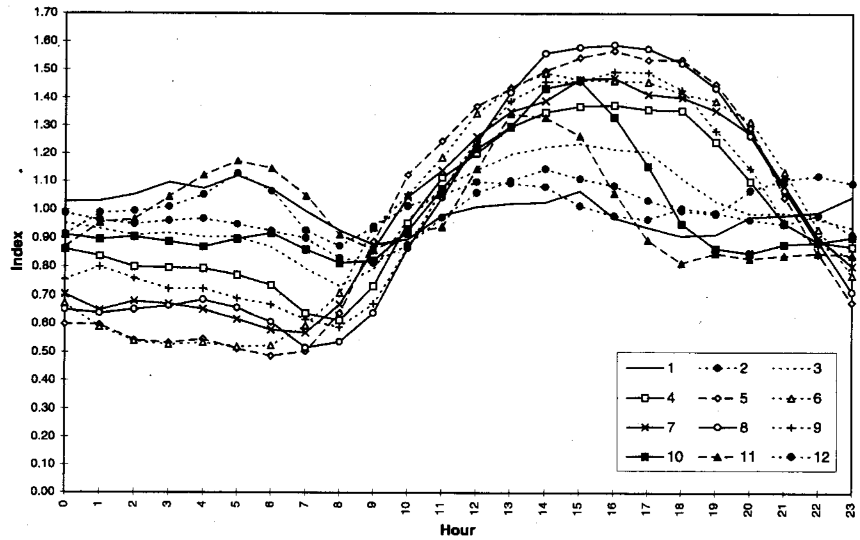


Figure 6.11 The monthly diurnal variation of O_3 as an index at Frederiksborg (2002) during 1990 used to represent the rest of Sealand and the rest of the country

7 Nitrogen Oxides and Nitrogen Dioxide

In this chapter, the annual means, trends and temporal variation of NO_x and NO_2 are derived.

Table 7.1 Regions represented by: Measurements (M) and Calculations (C)

Areas:	Areas:	Station	NO_x	NO_2	O_3
Urban	Copenhagen	Copenhagen (1259)	M	C	C
	Other cities	Fraction of Copenhagen	C	C	C
Rural	Greater Copenhagen Area	Lille Valby (2090)	C	M	M
	Rest of Sealand	Frederiksborg (2002[b]/2082[c]), Lille Valby (2090[d])	C	M	M
	Rest of the country	Frederiksborg (2002[b]/2082[c]), Lille Valby (2090[d])	C	M[a]	M

a Tange (6083) for annual mean of NO_2
b Used for O_3
c Used for monthly variation of NO_2
d Used for monthly diurnal variation of NO_2

7.1 Urban Background

Geographic variation

The annual means monitored at the urban background stations are presented in Table 7.2. An analysis of the concentrations levels during the same time periods shows that NO_x levels are about 4 per cent lower in Odense and about 28 per cent lower in Aalborg as compared to Copenhagen. For NO_2 it is about 16 per cent and 30 per cent. That is, the concentrations decrease with the number of inhabitants.

Table 7.2 Annual Means of NO_x and NO_2 (ppb) at Various Urban Background Stations

Station	Type	Period	1993		1994		1995	
			NO_x	NO_2	NO_x	NO_2	NO_x	NO_2
Copenhagen (1259)	Roof top	10.93-12.95	(22.8)	(12.1)	18.6	13.7	20.6	15.2
Odense (9159)	Roof top	5-8.93, 10-12.94	(15.4)	(9.5)	(20.8)	(12.7)	(16.1)	-
Aalborg (8159)	Roof top	5-9.94,6-8.95	-	-	(12.5)	(8.7)	(11.9)	-

* Years with limited observations are given in brackets

The average concentration during 1994-95 for Copenhagen (1259) is 19.6 ppb and this level is used as a reference to calculate NO_x levels in other cities.

For Copenhagen and other cities the NO_x levels are calculated with reference to the NO_x levels at the city centre of Copenhagen and the observed rural background levels of O_3 and NO_2. The NO_x level at the city centre of Copenhagen is predicted to 23.3 ppb (1994-95) based on the Copenhagen monitor station (1259), see Appendix D for details.

NO_x levels are predicted in smaller cities as outlined in Appendix C. NO_x levels at an address are also influenced by the distance to the city centre, see Appendix D for details.

Trends

The longest record of NO_2 and NO_x is for Copenhagen and it started in October 1993. The record is too short to suggest a trend.

In 1994 the annual mean of NO_2 is about 14 ppb in Copenhagen.

Campaigns

A few campaign measurements have been carried out in backyards at ground level adjacent to main urban streets in Copenhagen, Lyngby (north of Copenhagen) and Aalborg. Analyses show that the backyards were only to a minor extent influenced by the street and therefore "true" urban backgrounds (Rokkjær, 1986). Levels in Copenhagen in 1985-86 were in the range of 20-26 ppb, and one short campaign showed about 14 ppb. A comparison between campaign measurements for Copenhagen during May to June, 1986 and Aalborg May to July, 1986 shows that levels were about 50 % higher in Copenhagen.

The general picture is a notable decrease in NO_2 levels from 1985-86 to 1994 from about 20-26 ppb to 14 ppb in Copenhagen.

National inventories

During the same period national NO_x emissions have only decreased 10 % (Fenhann, Kilde 1994). However, national inventories are likely not to represent the development in urban emissions. The emission from traffic may have decreased more in cities due to a combination of more or less constant traffic loads (Bendtsen et al., 1994) and stringent emission standards whereas traffic has increased on a national scale. It is also likely that the decrease is partly due to a decline in emissions from other sources (heating, power plants) as SO_2 concentrations in Copenhagen have dropped by more than 50 % during the same period (Fenger, Kilde 1994).

Estimation of trends in urban NO_x emissions

As indicated above the development in national NO_x emission showed that trends are not likely to represent the development in urban background concentrations when compared to the few campaign measurements which have been performed.

Therefore, the development in NO_x emissions in central Copenhagen has been estimated and applied as an indicator for the trend in urban background concentrations of NO_x based on the actual development in traffic and emission factors. See Appendix F for details.

The estimated trend in NO_x emissions in central parts of Copenhagen is used to represent trends in urban background concentrations, see Figure 7.1.

The above mentioned campaign showed that NO_2 levels from 1985-86 to 1994 dropped from about 20-26 ppb to 14 ppb in Copenhagen which is equivalent to a decrease of 30-45 per cent. Estimated traffic NO_x emissions decreased about 26 per cent during the same period.

The estimated trend in emissions seems to give a fairly good representation of the trend in the concentrations.

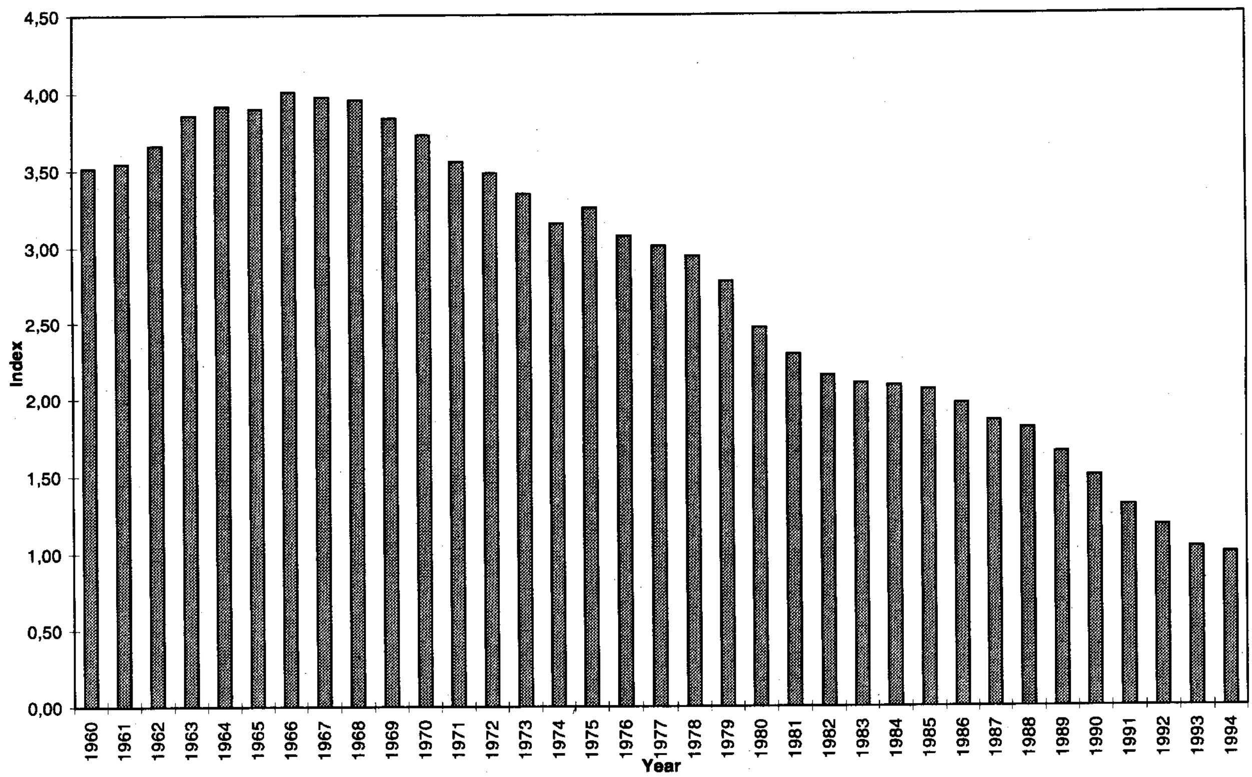


Figure 7.1 Estimated trends in NO_x emissions in central parts of Copenhagen used to represent urban background concentrations

Monthly variation

The monthly variation of NO_x is shown in Figures 7.2-7.4 for Copenhagen (1259), Odense (9159) and Aalborg (8159), respectively. For Copenhagen there seems to be a distinct seasonal variation with high levels during winter and low levels during summer. Odense and Aalborg show the same variation although observations are limited. The high levels during winter may be explained by higher emissions due to heating but also due to a more shallow mixing layer. During summer the mixing layer is typically deeper and traffic emissions are also lower at least during the holiday period of July.

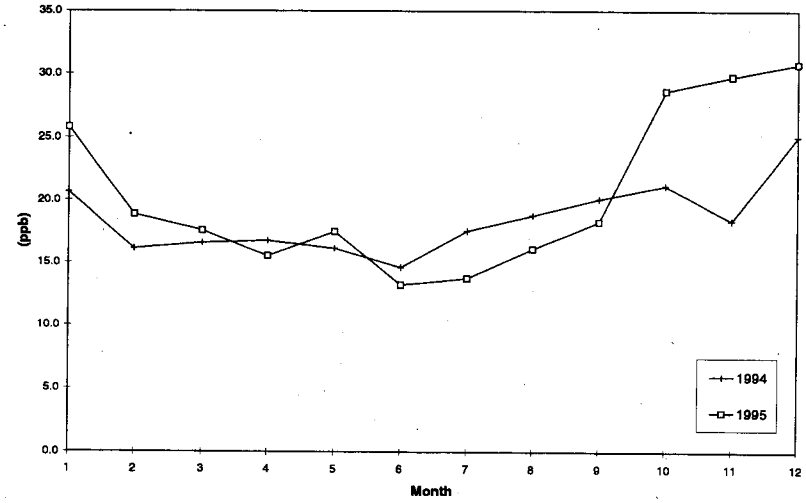


Figure 7.2 Monthly variation in NO_x during 1994-95 at Copenhagen (1259)

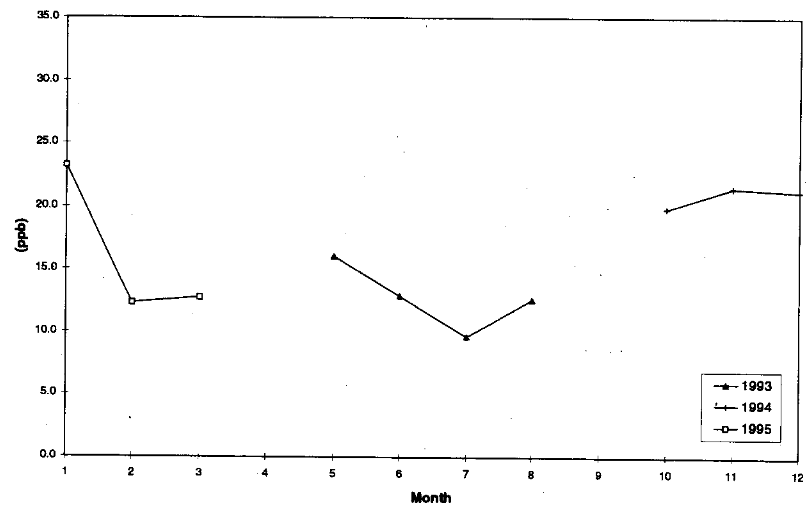


Figure 7.3 Monthly variation in NO_x during 1993-95 at Odense (9159)

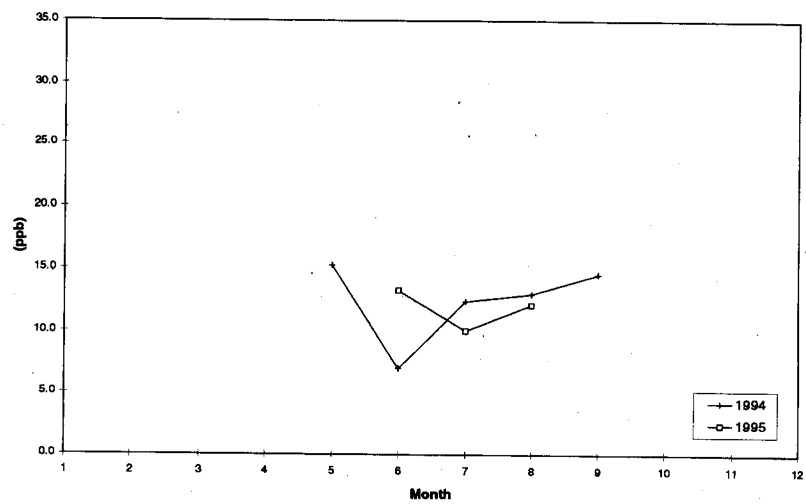


Figure 7.4 Monthly variation in NO_x during 1994-95 at Aalborg (8159)

The monthly variation of Copenhagen (1259) as an average of observations during 1994-95 is used to represent the urban background, see Figure 7.5 and 7.6.

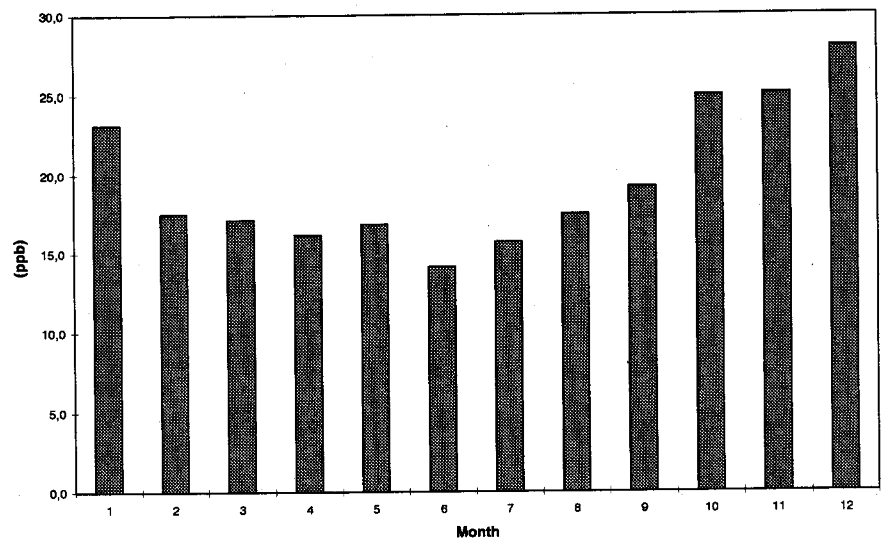


Figure 7.5 Monthly variation in NO_x in ppb as an average of 1994-95 at Copenhagen (1259) used to represent urban background concentrations

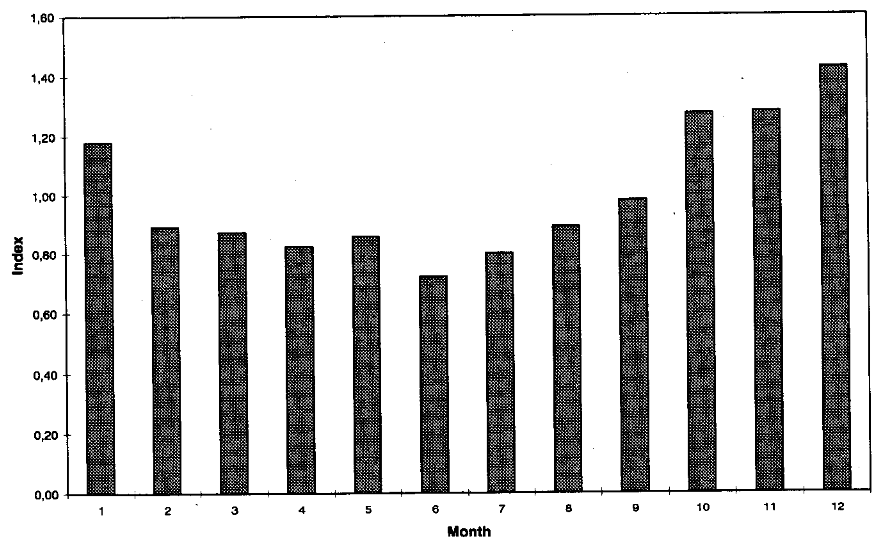


Figure 7.6 Monthly variation in NO_x as an index as an average of 1994-95 at Copenhagen (1259) used to represent urban background concentrations

Weekly variation

As shown in Figure 7.7 there is a weekly variation in the urban background concentrations. The levels during the week-end are lower than during the working days probably due to the fact that there is less traffic during the week-end (Jensen 1997). The difference between the various workings days is larger for NO_x than for CO (compare with Figure 5.5). This difference is not due to traffic as traffic loads are more or less the same during working days. To limit the number of calculations the minor weekly variation has not been taken into account.

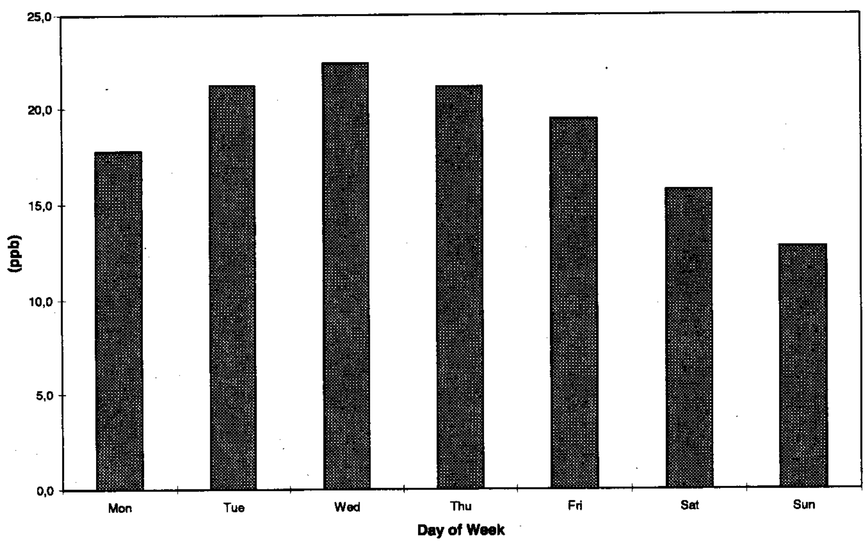


Figure 7.7 Weekly variation in NO_x during 1994 at Copenhagen (1259)

Diurnal variation

The diurnal variations of NO_x at Copenhagen (1259) during 1994 and 1995 is shown in Figure 7.8. The diurnal variation is similar for the two years and it is assumed that the diurnal variation is similar from year to year. The variation in traffic emissions with high emissions during morning and afternoon rush hours influences the diurnal variation of the background concentrations of NO_x. The build up of the boundary layer during the day dilutes the NO_x concentrations causing relatively low concentrations during the afternoon compared to the morning.

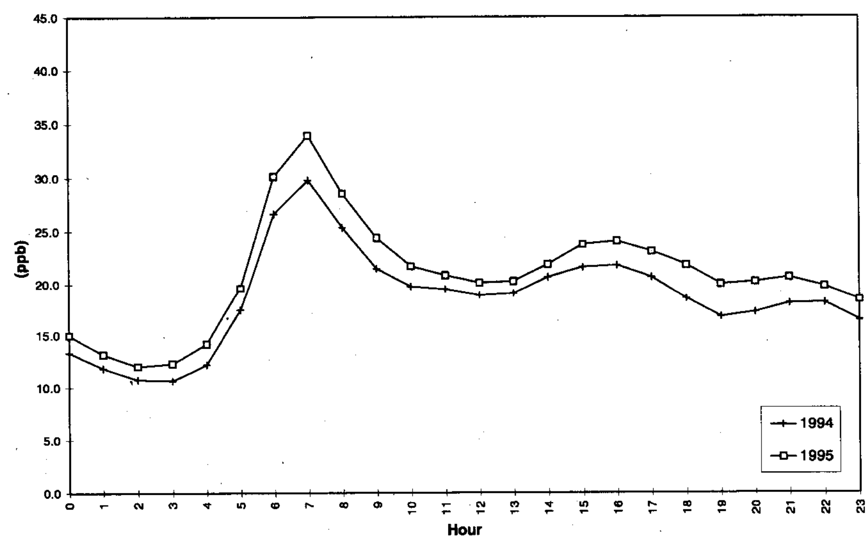


Figure 7.8 Annual diurnal NO_x variation during 1994 and 1995 at Copenhagen (1259)

Monthly variation in the diurnal variation

The monthly diurnal variation for Copenhagen (1259) for NO_x as an average of 1994 and 1995 is given in Figure 7.9. The diurnal variation is strong during winter with generally higher levels and weak during summer with generally lower levels.

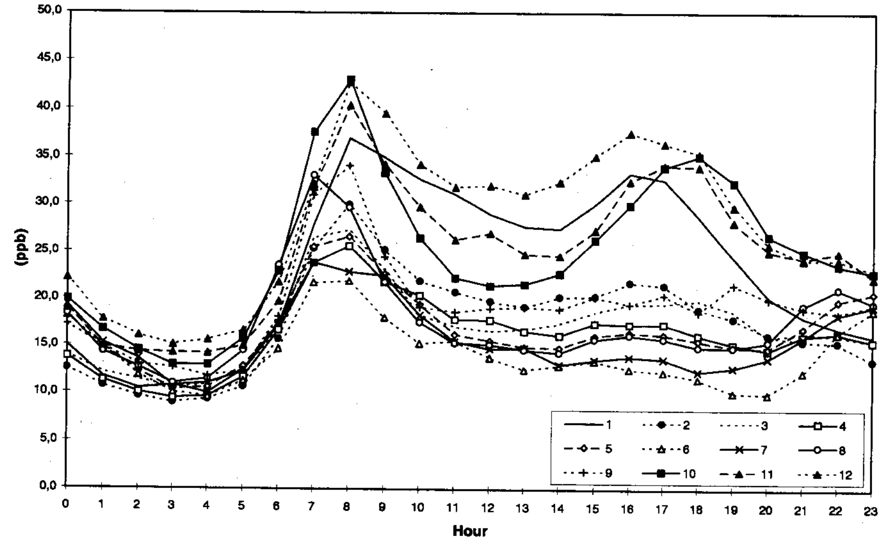


Figure 7.9 Monthly diurnal NO_x variation as an average of 1994 and 1995 at Copenhagen (1259)

The monthly diurnal variation for Copenhagen (1259) for NO_x as an index is given in Figure 7.10 and used to represent the monthly diurnal variation for urban background concentrations.

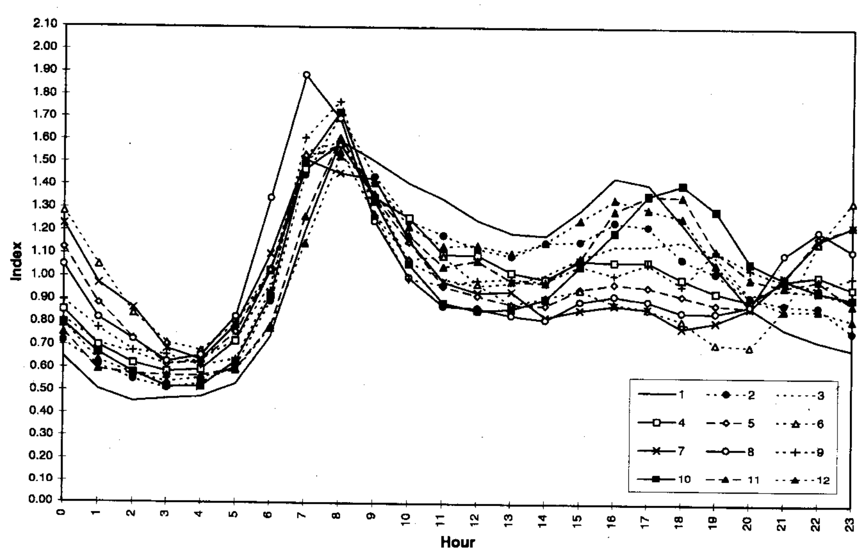


Figure 7.10 Monthly diurnal variation in NO_x as an average of 1994 and 1995 at Copenhagen (1259) as an index used to represent the monthly diurnal variation for urban background concentrations

7.2 Rural Background

In this section, the annual means, trends and temporal variation of NO_2 are derived for the rural background. NO_x for the rural background is calculated based on observed NO_2 and O_3 levels as outlined in Appendix B.

Geographic variation

Annual means of NO_2 at the rural background stations are given in Table 7.3.

Table 7.3 Annual Means of NO_2 (ppb) at Various Rural Background Stations

Station	Type	Period	1989	1990	1991	1992	1993	1994	1995	Mean**
Lille Valby (2090)	Rural	6.91-12.95	-	-	(7.8)	7.8	8.0	6.6	7.2	7.4
Frederiksborg (2082)	Forest	6.89-9.91	6.9	6.6	(5.9)	-	-	-	-	6.2
Ulborg (7081)	Forest	10.89-6.94	3.9	2.3	2.6	2.0	2.3	2.0	-	2.6
Tange (6083)	Forest	2.90-6.91	-	3.6	4.3	-	-	-	-	3.9
Keldsnor (9085)	Coast	2.90-3.91	-	4.6	7.6	-	-	-	-	5.3
Anholt (6081)	Coast	11.89-12.91,9.93-12.94	4.6	3.6	4.3	-	3.6	3.3	-	3.6

* Years with limited observations are given in brackets. ** Only for full annual records.

NO_2 levels at Lille Valby are a little higher than at Frederiksborg and somewhat higher than at the other stations. Levels are likely to be higher at Lille Valby and Frederiksborg because they are influenced by emissions from the nearby city of Roskilde and from the Copenhagen area. Levels are slightly higher at Lille Valby compared to Frederiksborg probably because of a closer location to urban emission sources and a slightly lower NO_2 deposition on agricultural land (Lille Valby) compared to forest (Frederiksborg). NO_x levels are only available for Lille Valby and NO_2 constitutes about 70 % of NO_x indicating that Lille Valby is influenced by local NO_x emissions since virtually all NO_x is expected to be in the form of NO_2 in remote rural areas.

Lille Valby is assumed to represent the rural NO_2 background levels in the Greater Copenhagen area with the average annual level during 1994 and 1995 of 6.8 ppb, Frederiksborg represents the rest of Sealand with the annual level of 6.6 ppb in 1990, and Tange represents the rest of the country with the annual level of 4.4 ppb (1.3.90-1.3.91). Ulborg has not been chosen to represent the rest of the country because it is located too close to the coast. Further, the coastal stations of Keldsnor and Anholt are not likely to represent rural conditions.

Trends

The longest record of rural NO_2 exists for Ulborg from 1989 to 1994. Linear regression shows a decrease of about 30 %. The other stations have either shorter time series or show a more diverse picture with decreases or increases.

At Rörvik and Vavihill in the southern part of Sweden for the period 1982 to 1988 decreases of 10 and 50 % have been observed (Lövblad et al. 1989).

For Danish sources national inventories of NO_x emissions began to be carried out in 1972. The emissions increased by about 25 % from the 70'ties to the middle of the 80'ties and have decreased by about 10 % from the middle of the 80'ties until 1993 (Fenhann & Kilde 1994).

The general picture of a decrease in NO_2 concentrations during the late eighties and the beginning of the nineties is supported by the decrease in the Danish NO_x emission during the same period.

Monitoring of concentrations have only been carried out in recent years, therefore, the development in estimated national road traffic

NO_x emissions have been taken as an indicator for the trends in NO_2 concentrations in rural areas to obtain a consistent trend since 1960, see Figure 7.11. Details are given in Appendix G. This trend shows an increase until the middle of the 80'ties followed by a small decrease. The trend follows the same development as the national NO_x inventory by *Fenhann & Kilde* (1994) but the pronounced decrease indicated by the few longer NO_2 records is only reflected as a minor decrease.

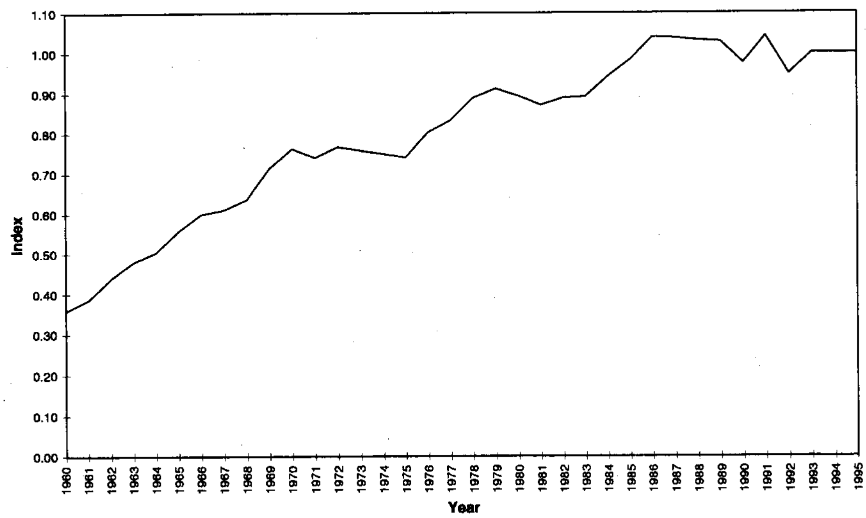


Figure 7.11 Trends in national traffic NO_x emissions as indicator for the development in rural NO_2 background concentrations

Monthly variation

The monthly variation from year to year is shown in Figures 7.12 and 7.13 for Lille Valby and Frederiksborg, respectively. There is a similar monthly variation from year to year for both stations. There is a minimum during summer and a maximum during winter. NO_2 levels are high during winter because the photodissociation of NO_2 is low due to low solar radiation and the levels are low during summer due to high depletion of NO_2 by photodissociation. Furthermore, NO_x emission are highest during winter compared to summer due to heating during winter, and the mixing layer is generally more shallow during winter compared to summer. Additionally, conversion of NO_2 to HNO_3 is also more rapid in summer.

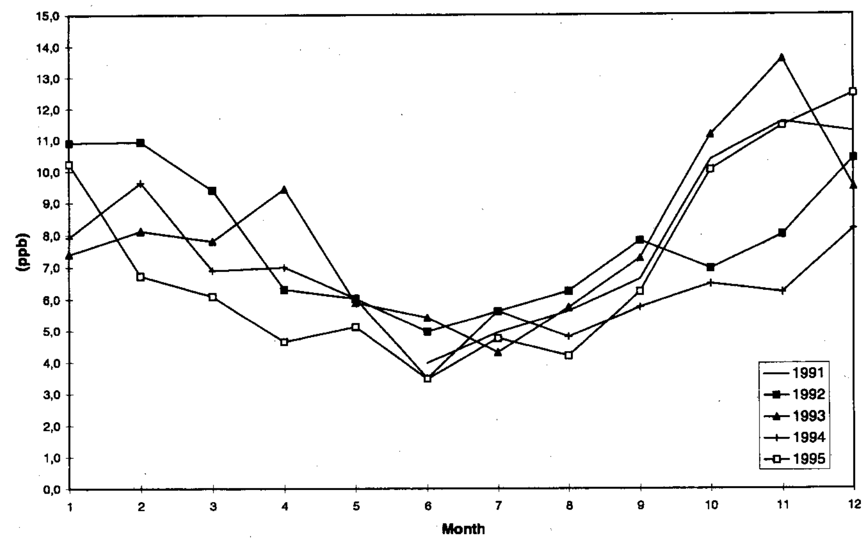


Figure 7.12 The monthly variation of NO_2 during 1991-95 at Lille Valby (2090)

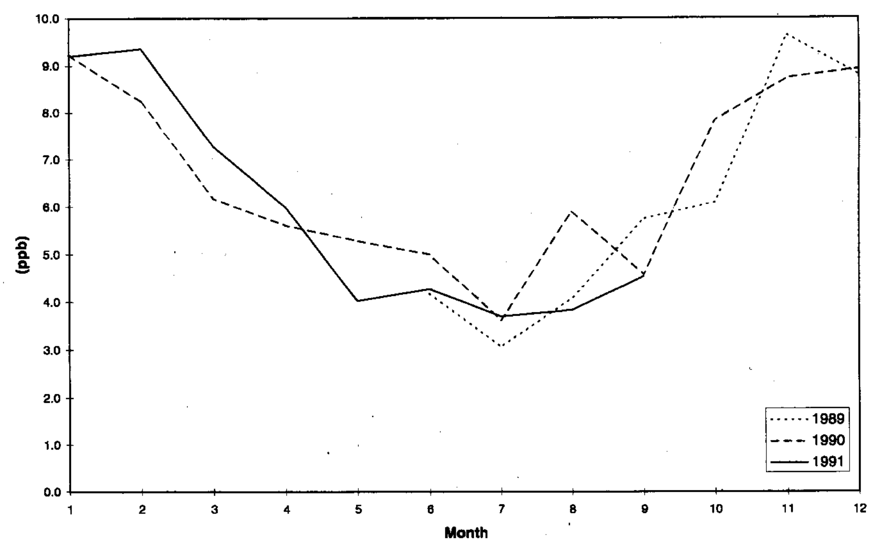


Figure 7.13 The monthly variation of NO_2 during 1989-91 at Frederiksborg (2082)

As an average during 1994-95, the monthly variation of NO_2 for Lille Valby is used to represent the rural background concentrations of the Greater Copenhagen area, Frederiksborg during 1990 will represent the rest of Sealand and the rest of the country, see Figure 7.14.

Weekly variation

The weekly variation has also been analysed and it varies from station to station with no clear picture other than the lowest levels are found during the week-end. Therefore, no weekly variation is assumed.

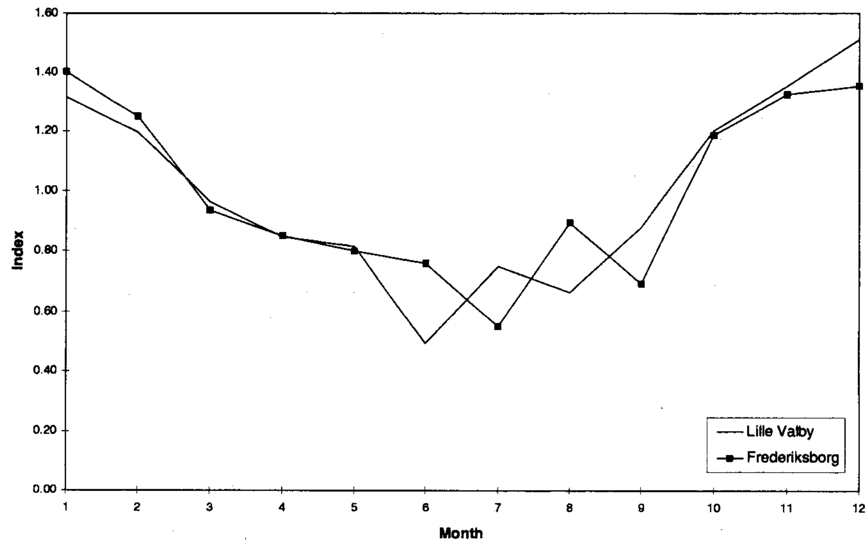


Figure 7.14 The monthly variation of NO_2 as an average during 1994-95 for Lille Valby represents the rural background concentrations of the Greater Copenhagen area, Frederiksborg during 1990 represents the rest of Sealand and the rest of the country

Diurnal variation

The diurnal variation is only available for Lille Valby (NO_x and NO_2) and for Ulborg (NO_2).

The NO_2 data from Ulborg covers a few months of the year and no clear picture of the diurnal variation from year to year can be derived. The data cover July of 1992, February and May of 1993 and June and July of 1994. The levels are more or less constant and very low. The low levels indicate that Ulborg is dominated by long-range transport of NO_2 and local NO_x emissions sources only play a minor role. It is not possible to generate a monthly diurnal variation for a year based on the limited data.

Lille Valby shows a distinct diurnal variation with high levels in the morning and in the evening, see Figure 7.15. The variation shows that Lille Valby is influenced by local NO_x emissions from the nearby city of Roskilde and from the Copenhagen area. Since the diurnal variation is similar from year to year although levels slightly varies the diurnal variation is assumed to be similar for all years.

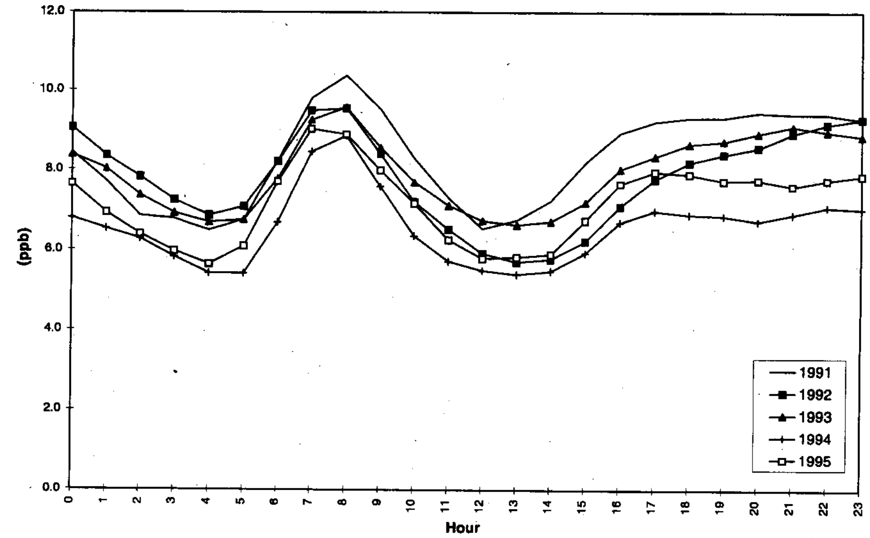


Figure 7.15 Annual diurnal NO_2 variation at Lille Valby (2090) during 1991-95

The diurnal variation of NO_2 is not available for Frederiksborg. However, there is a strong correlation for ozone between Frederiksborg and Lille Valby indicating that they are both influenced by emissions from the Greater Copenhagen area. Therefore, it is assumed that Frederiksborg has the same diurnal variation as Lille Valby.

Monthly variation in diurnal variation

Lille Valby has a distinct monthly diurnal variation of NO_2 with the highest levels during winter and lowest during summer, see Figure 7.16.

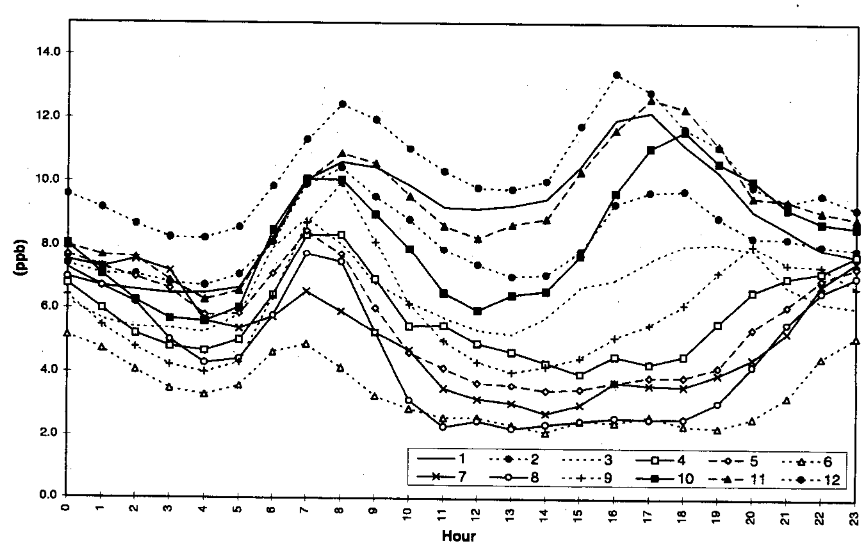


Figure 7.16 Monthly diurnal NO_2 variation for Lille Valby (2090) as average during 1994-95

Lille Valby is assumed to represent the diurnal variation in the various rural backgrounds regions. An average diurnal variation during 1994-95 has been generated as an index, see Figure 7.17.

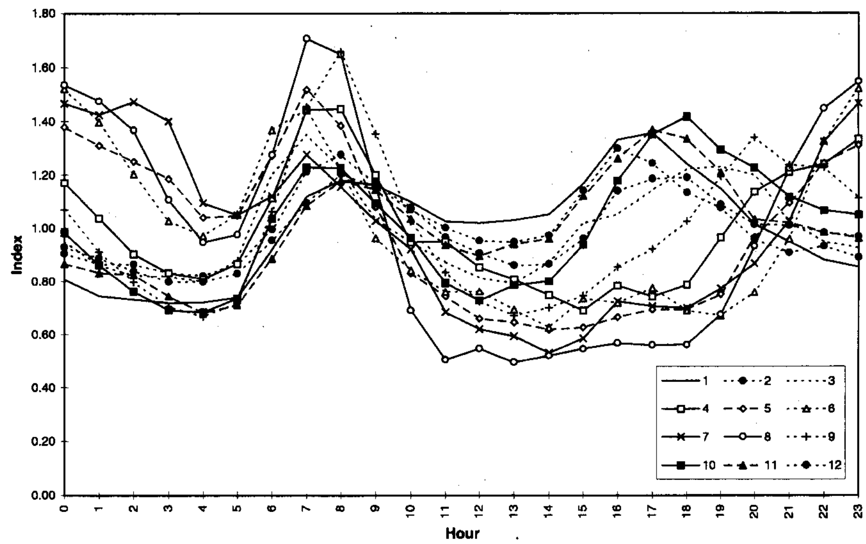


Figure 7.17 Monthly diurnal NO_2 variation for Lille Valby (2090) as average during 1994-95 used to represent the various rural background regions

8 Discussion and Conclusion

In this chapter a discussion of the method developed to estimate rural and urban background concentration levels at any location in Denmark from 1960-95 will be given. The discussion is focused on an evaluation of the assumptions, and the potential and limitations of the method.

8.1 Geographic Representativeness

Division of the country

The country was divided into three rural regions according to zip codes: the Greater Copenhagen Area, the rest of Sealand and the rest of the country. The Division of the country was based on an analysis of the annual mean and the annual diurnal variation of available monitor stations.

Proxy of rural CO

No Danish monitoring data was available for CO in rural areas, therefore, the ratio between urban and rural concentrations in the Netherlands was used as a proxy to derive rural background concentrations with reference to measured concentrations in Copenhagen. It has not been possible to test these assumptions.

Remote rural areas

The analysis was limited by the fact that the diurnal variation is only available for a few stations. Therefore, the stations selected to represent the country are located on Sealand. This is likely to introduce a bias as the rural areas on Sealand are clearly influenced by the emissions from the large cities. For remote rural areas on islands and rural areas far from larger cities the diurnal variation will be less influenced by city emissions.

Urban background

There were only minor differences in the annual diurnal variation between the three large cities that have monitor stations (Copenhagen, Odense and Aalborg). Therefore Copenhagen was selected to represent urban areas and a method was set up to predict levels in smaller cities.

Smaller cities

It is likely that the diurnal variation in smaller cities is slightly different from larger cities because of differences in the diurnal variation in traffic emissions which is the dominant source of air pollution in cities. In larger cities with many attractions traffic levels during the evening is probably higher than in smaller cities.

Concentration levels decline from the city centre to the outskirts of a city. Cities with no centre will have more evenly distributed concentrations levels. Further, for very small cities with a minor emission density background concentrations are excepted to be more or less constant over the city. We have arbitrarily chosen to separate rural and urban areas at 2.000 inhabitants.

8.2 Reference Year

A reference year has been established as an annual mean typically for either 1994 or 1995 or as an average of both years.

Averaging over more years

However, the meteorology differs from year to year and it may influence the annual mean by up to about 10 per cent. An average over several years makes the reference less dependent on a specific year. Therefore, the annual mean has been established as an average of two years for NO_x, NO_2, and O_3 to smoothen out annual differences. In the case that data are available it is a good idea to make an average of even more years, say 3-4 years provided that the development in emissions is minor during the period.

8.3 Trends

In the absence of monitoring data trends in NO_x and CO concentrations have been established based on the development in estimated traffic emissions since 1960. It is reasonable to assume that trends in annual concentration levels follow the trends in annual emissions although differences between years in meteorology will slightly modify the link between emissions and concentration levels.

Urban trends

For the urban background the development in emissions has been based on the development in emission factors and in traffic loads in the central part of Copenhagen because traffic loads in cities have had a different trend compared to national traffic performance. In Copenhagen, traffic levels increased fast during the 50'ties and become saturated in the beginning of the 60'ties and traffic levels have been more or less constant during 1960-1995. For smaller cities the saturation may have taken place at a later stage compared to Copenhagen.

Rural trends

For the rural background the development in emissions has also been based on the development in emission factors but applying the development in national traffic performance. Although traffic is a less dominant source in rural areas the development in national traffic has been used to obtain a consistent trend as the national inventories of all sources first started out in 1972.

The trend in O_3 is expected to be reliable since it is based on monitor data.

8.4 Temporal Variation

Weekly variation

The temporal variation has been established as a monthly variation and a monthly diurnal variation. The weekly variation with lower levels during week-ends has not been taken into account since the difference is not very substantial. However, the differences are not negligible and they may have been included. This would require a separate diurnal variation for working days and week-ends.

Impact of meteorology

The temporal variation has been based on the reference year and the temporal variation has been assumed to be the same for every year since 1960. However, the meteorology differs from year to year, therefore, the applied monthly variation and the monthly diurnal variation are specific for the reference year and it would slightly differ if another reference year was chosen.

Measured urban NO_x

In the urban background a monthly variation and a monthly diurnal variation of NO_x have been established based on observations. NO_2 and O_3 are calculated based on meteorology for every hour of the year. This implies that NO_x levels are constant for each hour during one day and night during a month whereas NO_2 and O_3 differs depending on the meteorology. Obviously, all hours from say 0-1 during a month will not experience the same NO_x levels. However, this method has been applied since it is simple and since the objective of the method has been to determine long-term exposure. The method will work for annual and monthly means but large differences will arise between observed and predicted levels if a comparison is made on an hourly basis. Similar problems are encountered in the rural background where NO_x levels are predicted based on a monthly variation and a monthly diurnal variation of observed NO_2 and O_3. This is a limitation of the method that need to be improved to be able to estimate reliable time-series during shorter time periods for prediction of short-term exposure.

8.5 Meteorology

The meteorology measured at Copenhagen Air Port (Kastrup) during 1995 is used for calculations of NO_x, NO_2, and O_3 for every year since 1960. This meteorological station is used to obtain a complete record as some periods were missing for Copenhagen (1259) and Lille Valby (2090).

Within the region of Denmark the meteorology of different stations is not expected to differ to such a degree that it would influence the monthly variation and the monthly diurnal variation of concentrations. However, the meteorology over cities may differ slightly from the rural setting due to differences in roughness.

The meteorology differs from year to year and the predicted hourly concentrations would be different if actual meteorology could have been applied for all years. However, it would have been more resource demanding to work with the actual meteorology for all years.

8.6 Photo-chemistry and Air Exchange

Rural background

In the rural background NO_x levels are predicted based on observed levels of NO_2 and O_3. These calculations are based on the assumption that there is a steady state between NO_x, NO_2 and O_3 and mass conservation of NO_x. A photochemical steady state will take place within a few minutes, therefore, the transport time of emissions from larger locale sources should be more than a few minutes to allow the steady state to take place. Consequently, the transport distance should be more than about 600 m for an average wind speed of 5 m/s. On the other hand, if there are strong local sources placed close to the considered location the steady state conditions will not be fulfilled.

Urban background

In the urban background NO_2 and O_3 levels are calculated from observed urban NO_x levels and observed rural NO_2 and O_3 levels assuming photochemical steady state. The method takes into account the exchange with the direct vehicle emissions in the streets and the exchange between the urban and rural air. The exchange is defined as the transport time over the city (the effective transport length over the city divided by the wind speed).

The effective transport distance has been set to the city diameter defined as the densely built up area of the city. For Copenhagen it is estimated to 4,000 m based on examination of a map and some uncertainty on this figure can be expected.

Further, the effective wind speed has been set to half of the wind speed at roof level based on model fitting. This reduction factor may be dynamicly depending on meteorological conditions.

Another assumption is that the direct NO_2 emission from vehicles constitutes 5 per cent of NO_x. However, it may be slightly different.

The formation of NO_2 and O_3 due to photochemical reactions involving VOC is believed to be of minor importance under Danish meteorological conditions as observations can be explained by taking into account only reactions between NO, NO_2 and O_3.

8.7 Validation

Rural background

In the rural setting calculation of NO_x was validated against observed NO_x at Lille Valby (2090). The relationship between observed and modelled concentrations was fairly good (r^2 is 0.65) and the model only underestimates the observed mean levels by about 10 per cent. The model also predicted the temporal variation (monthly variation, weekly and annual diurnal variation) fairly good.

Urban background

The model to predict NO_2 and O_3 levels in the urban background based on observed NO_2 and O_3 levels in the rural background at Lille Valby (2090) was validated against observations for Copenhagen (1259). The relationship between observed and modelled NO_2 and O_3 is good (r^2 are 0.91 and 0.89, respectively). However, the model slightly underestimates the observed O_3 annual levels with up to 16 per cent which may be due to some photochemical processes involving hydrocarbons in forming NO_2 and O_3. The model also predicts the temporal variation very well although O_3 concentrations are slightly underestimated.

Smaller cities

To predict the urban background concentration of CO and NO_x in the centre of a given city the annual mean at the city centre of Copenhagen is used as a reference and this concentration is scaled down to represent the urban background concentration of smaller cities. The method estimates urban background concentrations for area sources of known emission density based on the assumption that the concentrations are evenly distributed and that the dispersion is linearly depending on the dispersion distance (city diameter).

The method has been validated for Odense (145,000 inhabitants) and Aalborg (115,000 inhabitants). The formula predicts the same levels for the two cities. Observed annual means of NO_x are only overestimated with 13 per cent and 3 per cent for Odense and Aalborg, respectively. Observed NO_x levels are slightly lower in Odense compared to Aalborg. This is probably due to a smaller emission density since the city centre of Odense is characterised by relatively large green areas and relatively little traffic compared to Aalborg.

Dependence on distance to city centre

NO_2 measurements in Copenhagen show that concentrations are depending on the distance to the city centre. Therefore, an empirical expression was used to take into account the distance from an address location to the city centre. Although the expression was derived based on NO_2 measurements it is also assumed to be valid for NO_x and CO.

It has not been possible to test these assumptions against measurements. However, the expression produces reasonable results as the predicted concentration levels decrease from the city centre to the outskirts of the city and eventually becomes close to the rural background.

Some cities in the Greater Copenhagen Area may be characterised as sub-urban cities with no centre or a centre where the emission density is not much higher than in other places. The expression does not represent these areas very well.

Trends

The trends in CO and NO_x concentration levels are assumed to follow the development in emissions.

A campaign showed that NO_2 levels from 1985-86 to 1994 dropped from about 20-26 ppb to 14 ppb in Copenhagen which is equivalent to a decrease of 30-45 per cent. Estimated traffic NO_x emissions decreased about 26 per cent during the same period. The estimated trend in emissions seems to give a fairly good representation of the trend in the concentrations. However, it is difficult to assess the trend based on a single campaign and there will be a large uncertainty on the estimated development in concentrations.

8.8 Impact of Background Concentrations on Street Concentrations

The OSPM calculates the concentration levels in the street as a contribution from traffic emissions in the street and a contribution from the background concentrations.

Street concentrations

It is obvious, that the more traffic in a street the less important is the contribution from the background. In a busy street the contribution from the background may be 10-20 per cent for CO and 50-60 per cent for NO_2. In streets with very low traffic the background contribution will dominate. The uncertainty on the hour by hour predicted concentrations by the background model is expected to be higher than for the predicted concentration contribution from direct traffic emissions. Since the street concentrations are the sum of the contribution from the background and the direct contribution from traffic, it implies that the uncertainty on the street concentrations will be higher for streets with little traffic compared to busy streets. Since most rural areas will be characterised by low traffic levels the uncertainty on predicted street concentrations will generally be higher in rural areas compared to urban areas.

Test

In Vignati et al. (1997) a test of the modelled background concentrations was carried out for a busy street in Copenhagen (Jagtvej). The test included NO_x and NO_2 from 1994 with measured traffic levels and street configuration. The relationship between measured and calculated street concentrations was good (r^2=0.84). However, the model underestimates the highest concentrations because the background model is based on average profiles of monthly and monthly diurnal variations that will not account for extreme situations. For monthly means the differences for NO_x and NO_2 between the model and measurements were less than 10 and 15 per cent, respectively. For annual means the differences for NO_x and NO_2 were less than 1 and 3 per cent, respectively.

If a similar test would be possible to carry out for a street with low traffic in a rural area the results would most likely have shown much larger difference between calculations and measurements.

8.9 Application of Background Model in Exposure Assessment

The background model has been used as an input to the Childhood Cancer Project which is a large-scale epidemiological study. The study also uses other crude inputs like street configuration data based on information from a questionnaire and standardised traffic profiles.

Front-door concentrations

Part of this project has been to evaluate the ability of the OSPM to predict the concentration levels of NO_2 and benzene at the front-door of the children participating in the project. 103 street locations in Copenhagen and 101 locations in rural areas outside Copenhagen were evaluated in *Raaschou-Nielsen et al.* (1998).

Even with these crude input assumptions, the OSPM model system reproduced well the difference in NO_2 between Copenhagen and rural areas, and the differences between various locations in Copenhagen especially for long averaging times. The association between predicted and observed NO_2 levels declined with shorter averaging time: half year, one month and one week. For Copenhagen r^2 was 0.55, 0.38 and 0.34, respectively.

The predictions were generally better for urban areas compared to rural areas. In rural areas, the prediction power of the model was very low when compared to one week observations of NO_2 and benzene due to the largely empirical way background concentrations are estimated that will smooth out differences between weeks within a month.

The evaluation shows that the background model can be applied in epidemiological studies which consider long-term exposure on at least a monthly basis with the present design of the background model. Long-term exposure assessment in epidemiological studies is mainly suitable for studies of chronic effects e.g. cancers.

Personal exposure

The relation between front-door concentrations and personal exposures to NO_2 and benzene has also been evaluated in the Childhood Cancer Project (Raaschou-Nielsen et al. 1997a, b).

These studies show that the front-door NO_2 concentration is a fairly good indicator of personal exposure especially in urban areas but also in rural areas. Apart from front-door concentrations the personal exposure was also influenced by bedroom concentrations, time spent outdoors, gas appliances used at home, passive smoking and burning candles.

Front-door benzene concentrations was a less good indicator for personal exposure for urban areas when compared to NO_2 and misclassification would occur if applied for rural areas. Personal exposure of children was also influenced by riding in cars, exposure to gasoline vapours like motor cross, moped driving and refuelling of cars.

9 Future Research Needs

The discussion of future research needs has been divided into improvements of the presented empirical background model and an outline for using an existing regional background tranport model (ACDEP) with an improved emission inventory of higher resolution.

9.1 Improvements of Existing Background Model

The described background model is basically an empirical model based on a few monitoring stations which have been used to derive standardised temporal variation profiles in the form of a monthly variation and a monthly diurnal variation. In the case of NO_x in rural areas and NO_2 and O_3 in urban areas these were calculated assuming a steady state between NO, NO_2 and O_3 and using meteorological data from 1995.

Averaging over more years

One year or an average of two years of monitoring data have been used to generate a reference year and temporal variation profiles. Therefore, the reference year and the profiles are depending on the actual meteorology of the specific years. Since the data collection started for the present project a longer record is now available for several compounds and it is now possible to generate an average using more years say 3-4 years. This would smooth out the impact of the year to year meteorological variations on the reference year and temporal profiles and provide a better average estimate of concentrations levels over longer time periods as needed in the context of long-term exposure assessment. Furthermore, the most typical meteorological year of the years used for averaging concentration levels should be used or alternatively a standard meteorological year typical for many years should be applied. These improvements are expected to have some influence on the reference year as annual means may vary up to about 10 per cent from year to year. The influence on the monthly variation would be even higher.

Temporal breakdown

To some extent week-ends had lower concentrations of CO and NO_2 compared to working days. Therefore, the temporal breakdown could have been supplemented with separate monthly diurnal variations for working days and week-ends. However, a such refinement is only expected to have minor influence on the annual and monthly means used for long-term exposure assessment.

Concentration gradient over a city

The concentration gradient over a city was determined empirically based on data from Copenhagen to take into account the assumed decline in concentrations from the centre of a city to the outskirts. However, a study could be dedicated to determine the concentration gradient for different city sizes and types.

Smaller cities

The background concentrations in smaller cities were estimated using a simple formula with reference to Copenhagen, and it showed good results when tested for Odense and Aalborg. However, a study of the background concentration in smaller cities would provide new information. Such a study could be combined with a study of the concentration gradient over different types of cities.

CO in rural areas

CO concentrations in rural areas were estimated as a fraction of Copenhagen and the ratio between urban and rural backgrounds are determined based on Dutch monitoring data. A one year CO campaign at a rural background station could provide new information about annual means and temporal variation.

Trends

The trends in CO and NO_x concentration levels were assumed to follow the development in emissions due to lack of long monitor records.

For urban areas trends were based on developments in emissions factors and traffic in central part of Copenhagen. A study that looked into the presumable different traffic developments in different types of cities could be carried out. For rural areas trends followed estimated national traffic emissions although traffic is not a major source in rural areas. However, an estimate of all sources could be carried out although difficulties in data collection probably would arise for the years prior to 1972 as the national inventory started this year. Although, different trends may be identified it would be difficult to assess their validity because trends in emissions can not be compared with measurements.

Calculation of NO_x in urban areas

In urban areas O_3 and NO_2 were calculated based on measured urban background concentrations of NO_x and measured rural background concentrations of O_3 and NO_2. This standard monthly diurnal variation profile of NO_x is independent of meteorology which is a limitation in the present methodology.

Instead of being based on measurements, NO_x could have been calculated applying a more refined version of the area source dispersion formula used to down scale concentration levels in Copenhagen to smaller cities (See Appendix C). In *Hertel and Berkowicz* (1990) the more refined version is described which also includes actual meteorology, the emission density of the city and the extension of the city. The model has shown reasonable results when compared to measurements in *Hertel and Berkowicz* (1990) and model tests with recent and more complete emission data given in *Bendtsen and Reiff* (1996) may show even better results. This approach could be validated against measurements in Copenhagen, Odense and Aalborg.

If NO_x were calculated with actual meteorology then the variation in NO_x would be improved and therefore also the calculation of O_3 and NO_2 in urban areas. This approach is expected to improve estimated levels for shorter averaging times than the annual and monthly mean, thereby, improving the background model for application in short-term exposure assessment.

9.2 Application of the ACDEP-Model

The presented background model is based on empirical data derived from observations at the monitoring stations. However, it is also possible to determine background concentrations using air pollution transport models like the Atmospheric Chemistry and Deposition Model (ACDEP) described in *Hertel et al.* (1995). The ACDEP-model is a trajectory model. It has 10 vertical grids and the horizontal grids are divided into 150x150 km covering Europe. The model makes use of emissions and meteorological data from EMEP (Co-operative Programme for the Monitoring and Evaluation of Long Range Air Pollutants in Europe). For Denmark a finer grid resolution of 15x15 km for the emission inventory has been produced based on a top-down approach for geographic distribution of the national emission inventory.

Presently, the ACDEP-model calculates six hourly averages of various pollutants but it could be modified to calculate hourly concentrations of NO_x, NO_2, O_3 and CO for urban and rural backgrounds.

The reason why this approach has not been applied in the present study is the requirement of a very fine emission inventory grid. However, with the development in national digital maps and databases it should be possible to generate an emission inventory with a high geographic resolution of say 1x1 km close to the receptor point using GIS.

The emission inventory would make use of the digital land-use maps presently under development under the Land-Use Information System (AIS) in NERI. Detailed maps for land-use in rural areas are already available and new maps under development will include detailed information about land-use in urban areas e.g. industrial zones, residential areas, business districts etc. Emission factors for various types of land-use could be generated based on case studies. The temporal variation of emissions would also have to be defined. A GIS would make it possible to apply any resolution of a grid to generate the required emission inventory.

Since traffic is a major source it could be treated separately using the "VejNetDK" developed by the National Survey and Cadastre and the Danish Road Directorate. It combines a digital road network (1:200,000) and data from the Danish Road Database (VIS) that includes traffic data from state, county and main municipal roads.

Calculations with the ACDEP-model using the high resolution emission inventory could be validated against measurements at both rural and urban monitoring stations.

For application in the Childhood Cancer Project the country would still have to be divided into larger geographic areas to limit the number of calculations because calculations by the ACDEP-model are quite computer time demanding.

The ACDEP-model was not applied in the Childhood Cancer Project because it would have been time demanding to generate the emission inventory.

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Danish Summary - Dansk resumé

Baggrundskoncentrationer for brug i OSPM-modellen

Faglig rapport fra DMU, Nr. 234, 1998

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Baggrundsmodellen

Der er udviklet en baggrundskoncentrations model som anvendes i OSPM - Operational Street Pollution Model - i forbindelse med modellering af langtidseksponering i "Børnecancer Projektet". Børnecancer Projektet er en omfattende epidemiologisk undersøgelse af danske børn, som omhandler sammenhængen mellem udviklingen af cancer og eksponering med trafik luftforurening i barndommen (Raaschou-Nielsen et al. 1996). De børn, som er med i undersøgelsen, er spredt ud over hele Danmark.

Baggrundsmodellen er en semi-empirisk metode baseret på målte by- og landbaggrundskoncentrationer af NO_2, NO_x, O_3 og CO fra få moniteringsstationer, der repræsenterer forskellige geografiske områder i Danmark.

Den historiske udvikling i koncentrationsniveauerne er estimeret ud fra trafikemissioner som et indeks, og et basisår med årsmiddel niveauet fra et specifikt år. Udviklingen i ozon er alene baseret på målinger. Den tidslige variation er beskrevet ved et indeks, hvor sæsonvariationen fremkommer ved månedlige faktorer, og døgnvariationen ved faktorer måned for måned. På denne måde kan koncentrationsniveauet estimeres på time basis i perioden 1960-1995.

Niveauet af NO_x er beregnet i landområder baseret på målinger af koncentrationprofiler af NO_2 og O_3, idet der er taget hensyn til simpel fotokemi mellem NO, O_3 og NO_2. NO_2 og O_3 er beregnet i byområder baseret på målte koncentrationsprofiler af NO_x i byområder og målte koncentrationsprofiler af NO_2 og O_3 i landområder. En validering af antagelsen om simpel fotokemi som den dominerende faktor viste gode resultater. CO koncentrationerne i landområder er beregnet som en fraktion af niveauet i København, og forholdet mellem by- og landbaggrund blev bestemt ud fra hollandske moniterings data.

For at bestemme koncentrationsniveauet i mindre byer, hvor moniterings data ikke er til rådighed, blev et simplificeret spredningsudtryk brugt til at etablere en reduktions faktor med reference til antallet af indbyggere i en by for at nedskalere de observerede niveauer i København til mindre byer. Denne metode er valideret for Odense og Ålborg med gode resultater. Inden for et byområde er der brugt en simpel empirisk formel til at beregne reduktionen i koncentrationsniveauet fra centrum til periferien.

Baggrundskoncentrationens indflydelse på gadekoncentrationen

OSPM-modellen beregner koncentrationsniveauet i gaden som et bidrag fra trafikemissioner i gaden og et bidrag fra baggrundskoncentrationen. Dette medfører, at baggrundskoncentrationens betydning for gadekoncentrationen vil være højere for gader med lidt trafik i forhold til trafikerede gader. Da de fleste landområder er karakteriseret ved lidt trafik, vil baggrundskoncentrationens indflydelse på gadekoncentrationen generelt være højere i landområder end i byområder.

En kontrol af den modellerede baggrundskoncentration er gennemført for en stærk trafikeret gade i København (Vignati et al. (1997)). Sammenhængen mellem de observerede og de beregnede gadekoncentrationer er gode ($r^2 = 0,84$). Modellen undervurderer dog de højeste timekoncentrationer, fordi baggrundsmodellen er baseret på gennemsnitsprofiler af månedsmiddel og månedlige døgnvariationer, som ikke medregner ekstreme forhold. For månedsmiddel af NO_x og NO_2 er forskellen mellem modellen og målingerne henholdsvis mindre end 10% og 15%. Forskellen for NO_x og NO_2 på årsmiddelværdien er henholdsvis mindre end 1% og 3%. Usikkerheden ved modellen stiger ved kortere midlingstid (årsmiddel til månedsmiddel) og usikkerheden vil yderligere stige ved endnu kortere midlingstid (uger, døgn og timer).

Anvendelse til eksponeringvurdering

Baggrundsmodellen er blevet brugt som en del af et modelsystem i Børnecancer Projektet. Dette projekt har også brugt andre grove input parametre såsom gadekonfigurations data indsamlet ved hjælp af et spørgeskema, samt standardiseret trafikprofiler. En vurdering af det OSPM baserede modelsystems evne til at forudsige målte NO_2 og benzenkoncentrationsniveauerne ved hoveddøren hos udvalgte børn i København og i landområde udenfor København, viste at baggrundsmodellen kan anvendes i epidemiologiske undersøgelser som Børnecancer Projektet, som tager langtidseksponering på mindst måneds basis i betragtning (Raaschou-Nielsen et al. (1998)).

Sammenhængen mellem koncentrationen ved hoveddøren og personlig eksponering af NO_2 og benzen er også blevet vurderet i Børnecancer Projektet (Raaschou-Nielsen et al. 1997a). Disse undersøgelser viser, at koncentrationen af NO_2 ved hoveddøren er en rimelig god indikator for personlig eksponering specielt i byområder, men også i landområder. Udover koncentrationerne ved hoveddøren blev den personlige eksponering også påvirket af koncentrationen i soveværelset, ophold udendørs, brug af gaskomfur, passiv rygning og stearinlys. Benzenkoncentrationen ved hoveddøren var en mindre god indikator for personlig eksponering i byområder, når man sammenligner med NO_2 og misklassifikation kan forekomme, når den anvendes i landområder. Personlig eksponering af børn blev også påvirket af bilkørsel, eksponering med udstødning fra motocross, knallertkørsel og påfyldning af benzin på tankstation.

Fremtidige forsknings behov

Mulige forbedringer af den præsenterede semi-emperiske baggrundsmodel er diskuteret. Disse forbedringer vil formentlig kunne forbedre estimeringen af års- og månedsmiddelværdier i forbindelse med vurdering af langtidseksponeringen.

Eftersom modellen delvis er baseret på standard koncentrationsprofiler vil den udglatte de ekstreme variationer i niveauerne. Denne begrænsning skal udbedres for at være i stand til at estimere pålidelige tidsserier for kortere tidsperioder for at kunne forudsige korttidseksponering med større nøjagtighed.

En skitse til en anden metode er også blevet diskuteret, den er baseret på anvendelse af en eksisterende regional baggrunds transportmodel (ACDEP) men med en højere opløsning i emissionsopgørelsen. Denne metode forventes, at kunne bestemme baggrundskoncentrationsniveauerne for kortere tidsperioder med større nøjagtighed. Metoden egner sig derfor i højere grad til bestemmelse af de højeste forekommende niveauer, hvilket vil forbedre OSPM-modellens anvendelighed til vurdering af korttidseksponering - især under forhold, hvor baggrundsbidraget har væsentlig indflydelse på gadekoncentrationerne.

Appendix A

Division of the Country into Geographic Regions

The country has been divided into urban and rural areas to characterise background concentrations. Urban areas are subdivided into Copenhagen (defined by zip codes) and other cities (defined by No. of inhabitants). Zip codes and No. of inhabitants are derived from the questionnaire of the Childhood Cancer Project. Rural areas are subdivided by zip codes into rural areas within the Greater Copenhagen Area, the rest of Sealand, and the rest of the country. A rural area is defined by information in the Questionnaire.

Table A.1 Definition of Geographic Regions by Zip Codes

Region:	Indicator station:	Zip Codes:
Copenhagen (within the motorway ring)	Copenhagen (1259)	1000-2000 and 2100, 2200, 2300, 2400, 2450, 2500, 2610, 2650, 2700, 2720, 2770, 2820, 2860, 2900, 2920
Greater Copenhagen Area (excl. CHP)	Lille Valby (2090)	2600, 2605, 2620, 2625, 2630, 2635, 2640, 2660, 2665, 2670, 2680, 2690, 2730, 2740, 2750, 2765, 2800, 2830, 2840, 2850, 2880, 2930, 2942, 2950, 4000
The rest of Sealand	Frederiksborg (2002/2082)	4001 - 4999
The rest of the country (Jutland, Funen, Bornholm):	Tange (6083) and Frederiksborg (2002/2082)	3700,3720,3730,3740,3751,3760,3770,3782, 3790,5000 - 9999

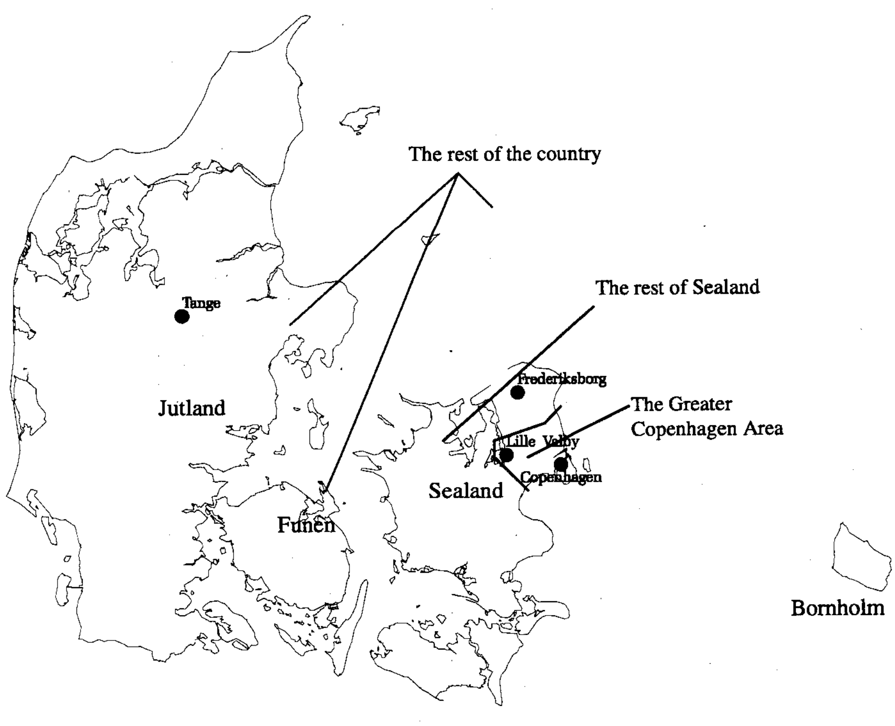


Figure A.1 Division of the country into four regions: Copenhagen, the Greater Copenhagen Area, the rest of Sealand, and the rest of the country defined by their zip code, and the locations of indicator monitor stations

Appendix B

Calculation Procedure for Estimation of Rural Background Concentrations of NO_x and CO

Introduction

This appendix describes how NO_x has been calculated in rural areas based on observed rural background concentrations of NO_2 and O_3 concentrations. Rural CO concentrations have simply been estimated as half of observed urban concentrations as outlined in section 5.2.

Table B.1 shows the monitor stations that have been used to represent the different rural areas. The rural areas are defined in Appendix A.

Table B.1 Monitor Stations Used to Represent Annual Means and Temporal Variation for Different Rural Areas: Measurements (M) and calculations (C)

Areas:	Areas:	Station	CO	NO_x	NO_2	O_3
Rural	Greater Copenhagen Area	Lille Valby (2090)	C	C	M	M
	Rest of Sealand	Frederiksborg (2002/2082)	C	C	M	M
	Rest of the country	Frederiksborg (2002/2082)	C	C	M*	M

* Tange (6083) has been used for the annual mean of NO_2

Methodology

The production and loss of NO, NO_2 and O_3 are covered by the below reactions:

$$NO + O_3 \rightarrow NO_2 + O_2 \quad (1)$$
$$NO_2 + hv \rightarrow O_3 + NO \quad (2)$$

To reduce the number of independent variables and to reduce the influence of local emissions a steady state between NO, NO_2 and O_3 as well as mass conservation of NO_x have been assumed in the calculation of NO_x. These assumptions are expressed below:

$$- k\,[NO]\,[O_3] + J\,[NO_2] = 0 \quad (3)$$
$$[NO_x] = [NO] + [NO_2] \quad (4)$$

From expression (3) and (4) the below expression can be derived:

$$[NO_x] = R^*[NO_2] \,/\, [O_3] + [NO_2] \quad (5)$$

where:

$R = J/k$

R: photochemical steady state constant

J: photodissociation rate

k: chemical reaction rate

if q<1 then J = 0 else J = (0.8E-3) * exp(-10/q) + (7.4E-6) * q

k = (5.38E-2) * exp(-1430/temp)

temp: temperature (Kelvin) at Copenhagen Airport.

q: solar radiation (w/m^2) calculated from cloud cover measured at Copenhagen Airport.

Expression (5) is used to calculate NO_x concentrations in rural areas hour by hour based on the observed monthly and diurnal variation profiles of NO_2 and O_3 rural background concentrations.

Validation of assumptions

The assumption - a steady state between NO, NO_2 and O_3 in rural areas has been tested. For this purpose, hourly observations of NO_2 and O_3 are used.

The observed NO_x concentrations during 1994-95 at the rural background station Lille Valby (2090) were compared to the calculated concentrations given by (5). Table B.2 and Figure B.4 reveal that the model based on equation (5) generally reproduces the observed concentrations of NO_x fairly well especially for the majority of lower concentrations. High concentrations are more difficult to predict as they may be influenced by local emission sources from the nearby city of Roskilde which has about 50.000 inhabitants and the nearby streets of Frederiksborg. Linear regression analysis shows that the r^2 is 0.65 and the model only underestimates the observed mean levels by about 10 per cent. This also indicates that under Danish meteorological conditions with relatively high wind speeds, low temperatures and little sun light the reactions between NO_2, NO and O_3 are the main photochemical processes taking place and other photochemical processes such as reactions involving hydrocarbons are of minor importance. Figures B.1-3 show that the model predicts the temporal variation fairly well although NO_x concentrations are slightly underestimated. The modelled NO_x concentrations are underestimated during night and evening hours probably because both NO_2 and O_3 are generally low during this period, and the ratio $[NO_2]/[O_3]$ in equation (5) is not well determined.

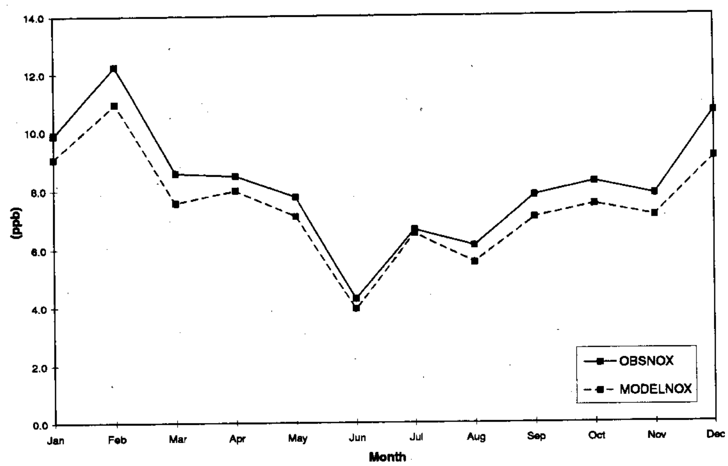


Figure B.1 The monthly variation in observed and modelled concentrations of NO_x at the rural background station Lille Valby (2090) during 1994

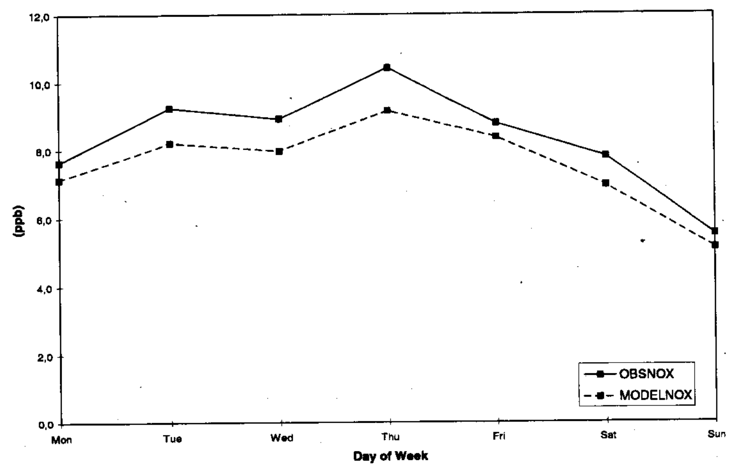


Figure B.2 The weekly variation in observed and modelled concentrations of NO_x at the rural background station Lille Valby (2090) during 1994

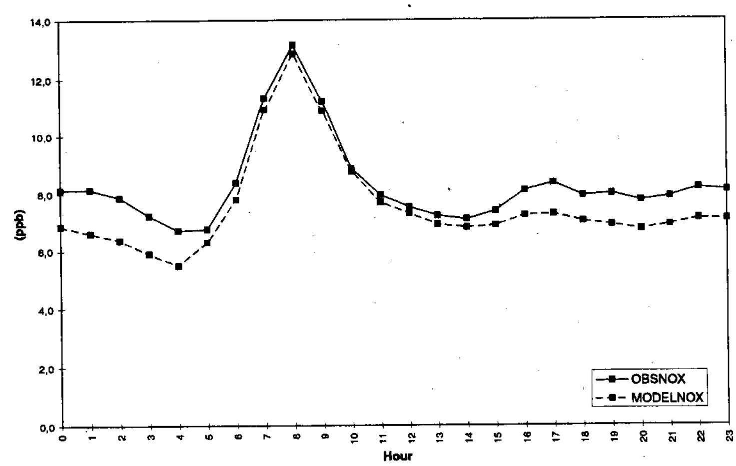


Figure B.3 The diurnal variation in observed and modelled concentrations of NO_x at the rural background station Lille Valby (2090) during 1994

Table B.2 Comparison of Observed Versus Modelled Rural Background Concentrations at Lille Valby (2090) in ppb

Compound	Year	Min.	Max.	Std. Deviation	Mean
NO_x (Obs.)	1994	0.50	138.1	9.7	8.3
NO_x (Mod.)	1994	0.50	103.1	8.0	7.5
NO_x (Obs.)	1995	0.50	245.9	14.3	9.8
NO_x (Mod.)	1995	0.50	212.7	10.6	8.7

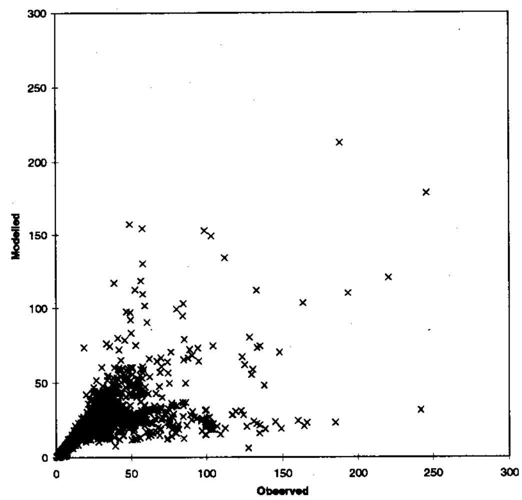


Figure B.4 Observed NO_x concentrations (ppb) during 1994-95 at the rural background station Lille Valby (2090) compared to calculated concentrations

Appendix C

Down-scaling of Urban Background Concentrations of NO_x and CO to a Given City Size

Introduction

Since the monitor programmes only cover the largest cities in Denmark it was necessary to develop an extrapolation method to estimate the urban background concentrations in smaller cities. To predict the urban background concentration of CO and NO_x in the centre of a given city the annual mean at the city centre of Copenhagen is used as a reference and this concentration is scaled down to represent the urban background concentration of the city in question.

Table C.1 shows the monitor station that has been used to represent the urban areas. See Appendix A for definition of the different urban areas.

Table C.1 Monitor Station used to Represent Urban Background Concentrations: Measurements (M) or calculations (C)

Areas:	Areas:	Station	CO	NO_x
Urban	Copenhagen	Copenhagen (1259)	M	M
	Other cities	Fraction of Copenhagen	C	C

Methodology

The method reported in *Hertel and Berkowicz* (1990) was applied. Estimated concentration levels have been normalised with respect to the levels in Copenhagen. The method estimates urban background concentrations for area sources of known emission density based on the assumption that the emissions are evenly distributed and that the dispersion is linearly depending on the dispersion distance (city diameter). The emission density is derived from *Bendtsen and Reiff* (1996) and the city diameter has been obtained from maps for selected cities. Details are given below.

In *Hertel and Berkowicz* (1990) a simplified area source dispersion formula is used to calculate urban background concentrations based on the emission density and the extension of the city:

$$C = \sqrt{\frac{2}{\pi}} * \frac{Q}{\sigma_w} * \ln\left[\frac{\sigma_w * d + h_0 * u}{h_0 * u}\right] \qquad (1)$$

where:

- C: concentration
- Q: emission density
- d: transport distance (city diameter)
- h_0: initial dispersion height (average building height)
- u: wind speed
- σ_w: vertical dispersion turbulence

Based on the assumption that the vertical dispersion turbulence is about 10 per cent of the wind speed ($\sigma_w/u = 0.1$) expression (1) can be reduced to expression (2) where the urban concentrations are only dependent on the emission density, the city diameter and the initial dispersion height:

$$C \approx Q * \ln\left[\frac{0.1 * d}{h_0} + 1\right] \tag{2}$$

In Table C.2 a reduction factor as an index with Copenhagen as reference is given based on expression (2). Calculations have been carried out for a number of cities. The city diameter is obtained from maps and the emission density is derived from *Bendtsen and Reiff* (1996) in which the emission densities of parts of Copenhagen were mapped. The emission density that best matched the characteristics of the selected cities was used.

Table C.2 Urban Background NO_x and CO Concentrations and City Area Characteristics

City	Inhabitants	d City diameter	h_0 Initial dispersion height	Q-NO_x Emission density	Q-CO Emission density	City Area Characteristics	$Factor_{NOx}$	$Factor_{CO}$
		(meter)	(meter)	(kg/km^2)	(kg/km^2)		(Index)	(Index)
København	550,000	4,000	20	125	960	Central Copenhagen, multi-storey buildings and heavy traffic	1.00	1.00
Århus	200,000	1,500	20	104	910	Frederiksberg, multi-storey houses and dense traffic	0.58	0.67
Aalborg	115,000	1,500	20	104	910	do	0.58	0.67
Vejle	53,000	1,000	10	67	500	Brøndbyøster, semi-dense built-up areas and some traffic	0.42	0.41
Roskilde	40,000	700	10	67	500	do	0.37	0.36
Køge	31,000	500	10	67	500	do	0.32	0.31
Holbæk	22,000	400	10	67	500	do	0.28	0.28
Struer	11,500	300	10	30	240	Rødovre, low density built-up area with little traffic	0.11	0.11
Billund	5,000	1,000	6	5	55	Ordrup, open residential area with little traffic	0.04	0.05
Gedser	1,000	500	6	5	55	do	0.03	0.04

In Figure C.1 the reduction factors are shown for CO and NO_x for any given city size.

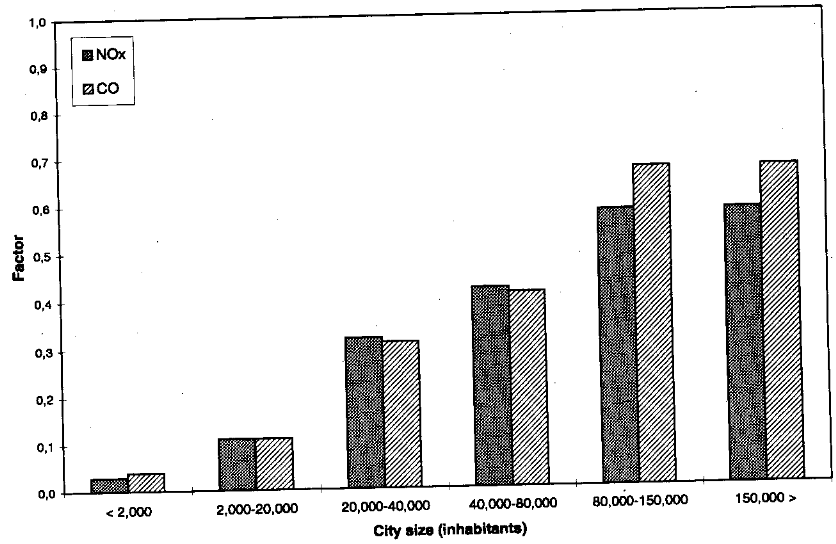


Figure C.1 The factors depending on city size used to scale down background concentrations of CO and NO_x with reference to concentrations in Copenhagen (1259)

The annual urban background concentration in a city is calculated as:

$$[NO_x]_{urban} = [NO_x]_{rural} + Factor*([NO_x]_{CHP,centre} - [NO_x]_{rural}) \quad (3)$$

where:

$[NO_x]_{urban}$	the urban background concentration in a city defined by the number of inhabitants
$[NO_x]_{rural}$	the rural background concentration
$[NO_x]_{CHP,centre}$	the urban background concentration levels in the city centre of Copenhagen, see Appendix D.
Factor	the reduction factor for NO_x depending on the number of inhabitants in the city given in Figure C.1.

Similar calculations are carried out for CO where $[NO_x]$ in formula (3) is substituted with [CO].

Validation

Formula (3) to predict urban background concentrations in smaller cities has been tested for Odense (145,000 inhabitants) and Aalborg (115,000 inhabitants). The formula reproduces the observed levels very well. The formula only overestimates the observed levels with 13 per cent and 3 per cent for Odense and Aalborg, respectively, see Table C.3. The reason why NO_x levels are slightly lower in Odense compared to Aalborg is probably due to a smaller emission density since the city centre of Odense is characterised by relative large green areas and relatively little traffic. Both urban background stations are located in the centre of the cities and, therefore, the observed levels have not been adjusted to take into account the distance between the city centre and the actual location of the monitor station as was the case for Copenhagen. It has not been possible to test the formula for other smaller cities due to lack of data from such sites. However, Figure C.1 shows that the down scale factor depending on the city size produces reasonable results as the factor decreases with decreasing city size and for small cities with less than 2,000 inhabitants it almost becomes zero indicating that the rural background level has been reached.

Table C.3 Comparison of Predicted and Observed Annual Levels (ppb) in Odense and Aalborg Using the Down Scale Model

Location:	$[NO_x]_{urban}$
Model Odense (9159), 1-12, 1994	15.1***
Obs. Odense (9159), 1-12, 1994*	13.2
Difference in per cent	13.0
Model Aalborg (8159), 1-12, 1994	15.1***
Obs. Aalborg (8159), 1-12, 1994**	14.7
Difference in per cent	3.0

* The observed mean at Odense (9159) during Oct-Dec 1994 was 17.5 ppb. The annual mean has been
estimated assuming that Odense (9159) has the same seasonal variation as Copenhagen (1259).
** The observed mean at Aalborg (8159) during May-Sept 1994 was 12.5 ppb. The annual mean has been
estimated assuming that Aalborg (8159) has the same seasonal variation as Copenhagen (1259).
*** The preticted NO_x concentration is given by equation (3) with $[NO_x]_{rural}$ equals 4.4 ppb, $[NO_x]_{CHP, centre}$ equals 22.9 ppb, and Factor $_{NOx}$ equals 0.58.

Appendix D

Urban Background Concentrations at a Given Distance From the City Centre

Introduction

Measurements carried out as part of the Childhood Cancer Project (Raaschou-Nielsen et al. (1997)) indicate that the urban NO_2 background concentrations are higher the closer an address is located to the city centre. This observation is taken into account in the calculation of the urban background concentration at an address using the distance from the address to the city centre given by the Questionnaire.

Methodology

On average the concentrations fell with about 50 per cent from about 500 m to 3,500 m from the city centre. An exponential fit to the data, and a further assumption that the city extension plays a role, led to the following expression:

$$[C]_{urban} = [C]_{centre} * \exp(-1.6 * (d / S)) + [C]_{rural} \quad (1)$$

where

$[C]_{urban}$ is the urban background concentration at the distance d from the city centre of a city with the city size parameter S

$[C]_{centre}$ is the concentration at the city centre and $[C]_{rural}$ is the concentration in the rural background. Details are given in *Raaschou-Nielsen et al.* (1997)

Although the above expression was derived based on NO_2 measurements it is assumed to be valid for NO_x and CO.

Figure D.1 shows the reduction factor "$\exp(-1.6 * (d / S))$" and its dependence of the distance to the city centre and of the city size parameter. The further the address is located from the city centre the smaller is the reduction factor, and the smaller the city is the smaller is the reduction factor.

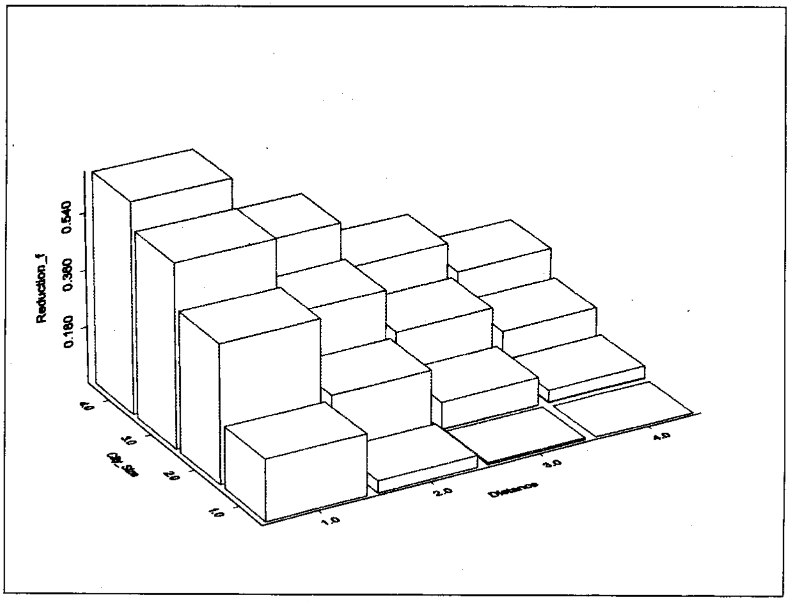


Figure D.1 The reduction factor "$\exp(-1.6*(d / S))$" and its dependence on the distance to the city centre and on the city size parameter

Annual Means of CO and NO_x at the City Centre of Copenhagen

Data from the monitoring station of Copenhagen (1259) which is located at a distance of 2 km from the city centre was used to calculate annual means of NO_x and CO at the city centre of Copenhagen, see Table D.1. The annual means of the city centre of Copenhagen are used as a reference down scaled to represent smaller cities.

Table D.1 Estimated Annual Means in the City Centre of Copenhagen

Location:	Distance (d) from city centre	City size parameter (S)	Annual means		
			NO_x (ppb)	NO_x (ppb)	CO (ppm)
			1994	1994-95	1994
City centre of Copenhagen	0	4	22.9	23.3	0.36
Copenhagen monitor station (1259)	2	4	18.6	19.6	0.31
Rural background	-	-	8.3	9.1	0.15

The city size parameter has been derived from maps for a number of cities to establish a simple relation between the city size parameter and the number of inhabitants of a city, see also Appendix C. The latter is given by the Questionnaire.

Table D.2 City Size Parameter Dependingon No. of Inhabitants in a City

Inhabitants	City size parameter (S)
- 2,000	100
2,000 - 20,000	300
20,000 - 40,000	500
40,000 - 80,000	1,000
80,000 - 50,000	1,500
150,000 -	1,500

Appendix E

Calculation Procedure for Urban Background Concentrations of NO_2 and O_3

Introduction

NO_2 and O_3 are calculated to reduce the number of independent variables and to make the relationship between NO_2, NO and O_3 consistent to reduce the influence of local emission sources.

Table E.1 shows the monitor station that has been used to represent the urban areas. See Appendix A for definition of the different urban areas.

Table E.1 Urban background represented by: Measurements (M) or calculations (C)

Areas:	Areas:	Station	NO_x	NO_2	O_3
Urban	Copenhagen	Copenhagen (1259)	M	C	C
	Other cities	Fraction of Copenhagen	C	C	C

Methodology

The chemistry of nitrogen oxides in Danish urban areas can roughly be described by the below reactions:

$$NO + O_3 \rightarrow NO_2 + O_2 \quad (1)$$

$$NO_2 + hv \rightarrow O_3 + NO \quad (2)$$

In order to take into account the exchange of air between the urban background and the rural background the rate of change of NO, NO_2 and O_3 can be expressed by the following expressions as given in *Hertel and Berkowicz* (1989b) and *Hertel et al.* (1997):

$$\frac{d[NO]}{dt} = -k[NO][O_3] + J[NO_2] + \frac{[NO]_v}{\tau} + \frac{[NO]_{rural} - [NO]}{\tau} \quad (3)$$

$$\frac{d[NO_2]}{dt} = k[NO][O_3] - J[NO_2] + \frac{[NO_2]_v}{\tau} + \frac{[NO_2]_{rural} - [NO_2]}{\tau} \quad (4)$$

$$\frac{d[O_3]}{dt} = -k[NO][O_3] + J[NO_2] + \frac{[O_3]_{rural} - [O_3]}{\tau} \quad (5)$$

The first two terms in equations (3)-(5) describe the loss and production due to chemical transformation given by (1) and (2). The third term in equation (3) and (4) describes the direct emission from vehicles in the streets and τ is the residence time defined as D_{city}/u where D_{city} is the effective transport length over the city centre (city diameter) and u is the wind speed above roof level. The last term is the exchange rate between the urban and rural air masses for the different compounds where the index "rural" indicates the rural background.

Assuming that a photochemical steady state is achieved in the urban background between NO, NO_2 and O_3 equations (1)-(3) become zero and assuming the following mass conservation equations:

$$[NO_2] = [NO_2]_v + [NO_2]_{rural} + [O_3]_{rural} - [O_3] \quad (6)$$

$$[NO_x] = [NO_x]_v + [NO_x]_{rural} \quad (7)$$

$$[NO_x] = [NO] + [NO_2] \quad (8)$$

then, equations (3)-(8) may be stated as:

$$[NO_2] = 0.5*\left(b - sqrt\left(b*b - 4*\left([NO_x]*NO_2\_a + NO_2\_n*D\right)\right)\right) \quad (9)$$

$$[O_3] = NO_2\_a - [NO_2] \quad (10)$$

where:
if q < 1 then J = 0
else J= (0.8E-3) * exp(-10/q) + (7.4E-6) * q
k = (5.38E-2) * exp(-1430/temp)
R = J/k (photochemical steady state constant)
$[NO_2]_v$=0.05 * ($[NO_x]$ - $[NO_x]_{rural}$)
$NO_2\_n = [NO_2]_v + [NO_2]_{rural}$
$NO_2\_a = NO_2\_n + [O_3]_{rural}$
b = $[NO_x]$ + R + D + NO_2_a
f = 0.5 (f is a reduction factor for wind speed which is model fitted to give the effective transport time over the city)
D = f * u/(D_{city} * k)
D_{city} is city diameter which is 4,000 m for Copenhagen
u: wind speed (m/s)
temp: temperature (Kelvin)
q: solar radiation (w/m^2)
v: index v stands for vehicle and the direct emission of NO_2 is assumed to be 5 per cent.

Equations (9) and (10) are used to calculate the urban background concentration of NO_2 and O_3 based on measurements of urban background concentrations of NO_x together with measurements of rural background concentration of NO_2 and O_3 and predicted rural background concentrations of NO_x. These calculations take place hour by hour for the years in question. Meteorological data from the Copenhagen Airport are used for calculations in the background model because the data record was complete.

Validation of Assumptions

The photochemical steady state assumption that equation (9) and (10) are built upon has been tested for the urban background station Copenhagen (1259) using Lille Valby (2090) as rural background station. Hour by hour measurements have been used to test the assumptions not the standardised profiles. Meteorological data from the Copenhagen University building: the H.C. Ørsted Institute, were used for the test.

Table E.2 and Figures E.1-2 show that the equations (9) and (10) generally reproduce the observed concentrations of NO_2 and O_3 very well. Linear regression analysis of the relationship between observed and modelled NO_2 and O_3 shows that the r^2 is 0.91 and 0.89, respectively. This indicates that under Danish conditions with relatively high wind speeds, low temperatures and little sun light the reactions between NO_2, NO and O_3 are the main photochemical processes taking place and other photochemical processes such as reactions involving hydrocarbons are of minor importance. However, the model slightly underestimates the observed annual levels with up to 16 per cent which may be due to some photochemical processes involving hydrocarbons in forming NO_2 and O_3 or that the steady-state assumption is not fulfilled.

Figure E.2 indicates that the model especially underestimates O_3 concentrations. This may be due to special meteorological conditions with very high wind speeds in which the retention time for the formation of O_3 in photochemical processes is limited. Figures E.3-5 show that the model also predicts the temporal variation very well although O_3 concentrations are slightly underestimated.

Test of the Urban Background Model

In Vignati et al. (1997) a test of the urban background model was performed. At that time the model was generated based on data from 1994. The monthly and monthly diurnal variation were compared to measurements from 1995 to test if variations were pronounced from year to year. Comparison of the two years showed that the levels were slightly different but that the temporal variation was in good agreement. However, to smooth out differences between the years, an average of 1994 and 1995 was generated for the model calculations.

Table E.2 Comparison of Observed Versus Modelled Urban Background Concentrations (ppb)

Compound	Year	Min.	Max.	Std. Deviation	Mean
NO_2 (Obs.)	1994	0.50	60.3	8.2	13.8
NO_2 (Mod.)	1994	0.50	51.3	7.4	13.8
O_3 (Obs.)	1994	0.50	89.0	14.1	25.5
O_3 (Mod.)	1994	0.00	91.2	13.9	21.4
NO_2 (Obs.)	1995	0.50	60.4	8.8	15.1
NO_2 (Mod.)	1995	0.50	46.3	7.6	14.2
O_3 (Obs.)	1995	0.50	98.7	13.9	22.7
O_3 (Mod.)	1995	0.00	94.0	13.5	19.0

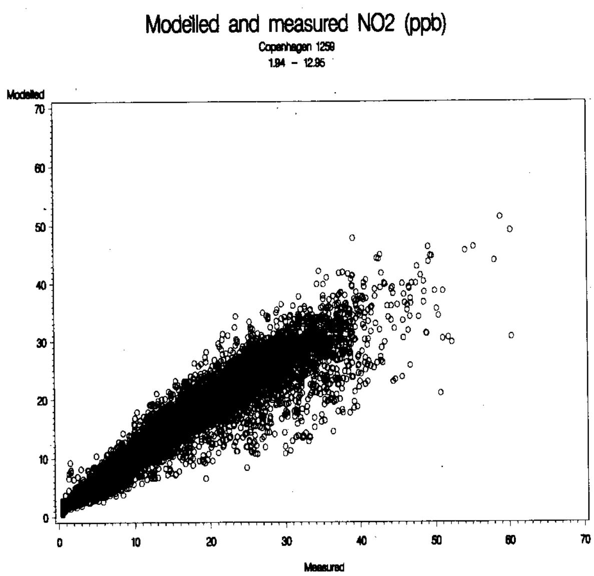


Figure E.1 Comparison of modelled and measured urban background concentrations of NO_2

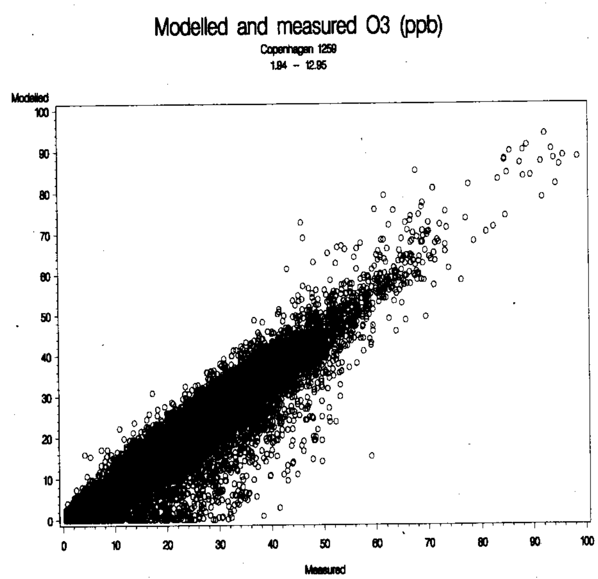


Figure E.2 Comparison of modelled and measured urban background concentrations of O_3

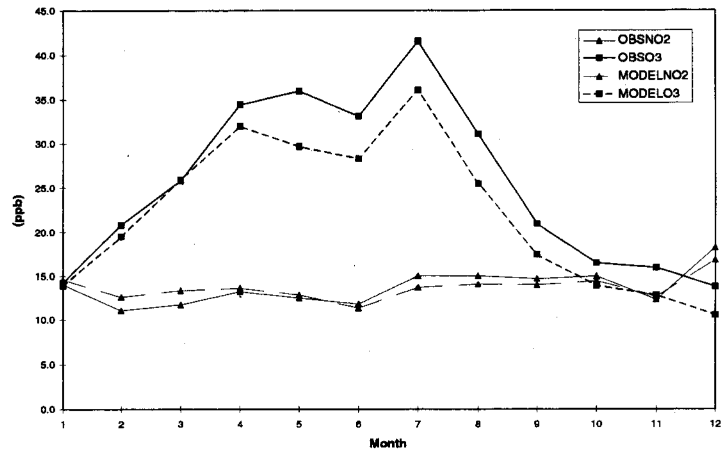


Figure E.3 The monthly variation in observed and modelled concentrations of NO_2 and O_3 at the urban background monitor station (1259) during 1994

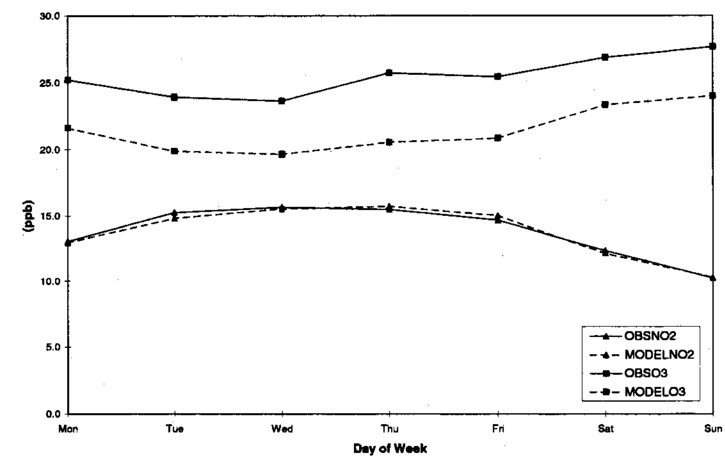


Figure E.4 The weekly variation in observed and modelled concentrations of NO_2 and O_3 at the urban background monitor station (1259) during 1994

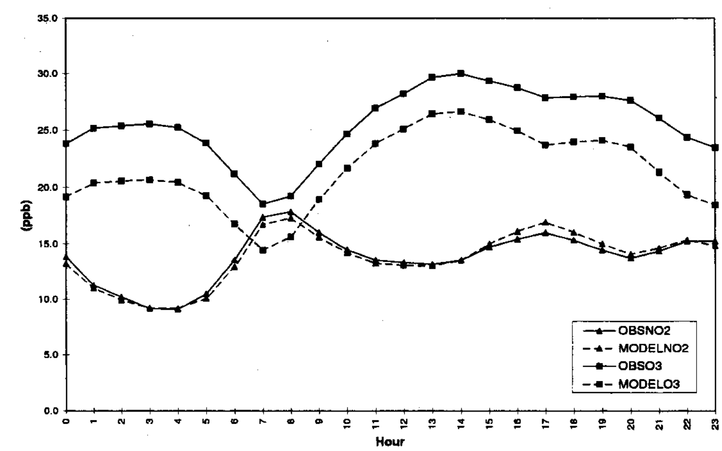


Figure E.5 The annual diurnal variation in observed and modelled concentrations of NO_2 and O_3 at the urban background monitor station (1259) during 1994

Appendix F

Trends in Urban CO and NO_x Traffic Emissions

Introduction

The Childhood Cancer Project requires an estimation of CO and NO_x background concentrations during 1960-1995. However, continuous monitoring of CO and NO_x has only been carried out for a limited period which does not allow for trend analysis. An assessment of the development in national CO and NO_x emission showed that trends in national emissions were not likely to represent the development in urban background concentrations when compared to the few campaign measurements which have been performed. Furthermore, studies by the Danish Road Directorate indicates that traffic has increased much more on regional roads than on urban roads in downtown areas.

Methodology and Data

The development in CO and NO_x emissions in central Copenhagen has been estimated and applied as an indicator for the trend in urban background concentrations of CO and NO_x based on the actual development in traffic and emission factors.

Traffic performance and traffic composition are based on data from the Municipality of Copenhagen. As indicators for the development traffic counts carried out at the arterial roads to the central parts of Copenhagen are used ("Søsnittet" and "Havnebroerne"), see Table F.1.

The basic CO and NO_x emission factors for the reference year 1993 have been established based on a study from the Danish Road Directorate (Jensen 1992). The development in emission factors is based on data from the Laboratory for Energetics, Technical University of Denmark (Sorenson, private communication), see Table F.2 and F.3.

The latest available data during the period of data collection originates from 1994, therefore, data for 1995 has been assumed to be similar to 1994.

Table F.1 Development in Traffic Performance and Composition in Central Copenhagen during 1960-1994

	Traffic Performance		Traffic Composition (per cent)					
Year	(1000 km)	(index)	Passenger cars (without catalyst)	Passenger cars (catalyst)	Vans	Trucks	Bus	All
1960	268	0.91	73.4	0.0	13.8	11.9	0.9	100
1961	276	0.94	73.8	0.0	13.7	11.5	1.0	100
1962	297	1.01	72.2	0.0	13.6	13.1	1.1	100
1963	312	1.06	74.6	0.0	13.4	10.8	1.2	100
1964	322	1.10	75.4	0.0	12.9	10.4	1.3	100
1965	327	1.11	75.8	0.0	13.0	9.8	1.4	100
1966	343	1.17	76.2	0.0	13.1	9.2	1.5	100
1967	347	1.18	76.7	0.0	13.1	8.6	1.6	100
1968	354	1.20	77.1	0.0	13.2	8.0	1.7	100
1969	350	1.19	77.5	0.0	13.3	7.4	1.8	100
1970	352	1.20	77.4	0.0	12.5	7.8	2.3	100
1971	340	1.16	79.3	0.0	10.9	7.5	2.3	100
1972	338	1.15	81.2	0.0	9.3	7.2	2.3	100
1973	330	1.12	83.0	0.0	7.9	6.8	2.3	100
1974	316	1.07	84.9	0.0	6.2	6.5	2.4	100
1975	332	1.13	86.5	0.0	5.7	5.5	2.3	100
1976	324	1.10	86.5	0.0	5.6	5.5	2.4	100
1977	330	1.12	86.6	0.0	5.4	5.5	2.5	100
1978	333	1.13	86.6	0.0	5.2	5.6	2.6	100
1979	326	1.11	86.7	0.0	5.1	5.6	2.6	100
1980	305	1.04	85.8	0.0	5.7	5.8	2.7	100
1981	295	1.00	85.6	0.0	6.0	5.8	2.6	100
1982	289	0.98	85.5	0.0	6.1	5.8	2.6	100
1983	295	1.00	85.4	0.0	6.1	5.9	2.6	100
1984	307	1.04	85.2	0.0	6.3	5.9	2.6	100
1985	318	1.08	85.0	0.0	6.5	5.9	2.6	100
1986	323	1.10	84.2	0.0	7.5	5.9	2.4	100
1987	319	1.09	84.6	0.0	7.6	5.4	2.4	100
1988	325	1.11	85.0	0.0	7.7	4.9	2.4	100
1989	316	1.07	85.4	0.0	7.8	4.4	2.4	100
1990	309	1.05	83.7	1.7	7.7	4.5	2.4	100
1991	300	1.02	78.2	5.9	9.1	4.4	2.4	100
1992	297	1.01	74.8	10.2	8.0	4.6	2.4	100
1993	290	0.99	70.2	14.4	8.8	4.1	2.5	100
1994	294	1.00	63.8	21.3	8.4	4.2	2.4	100

Note: The fraction of petrol powered cars equipped with catalyst from 1990, 1991, 1992, 1993 and 1994 is 2, 7, 12, 17 and 25 per cent, respectively

Table F.2 Development in CO Emission Factors (as index) during 1960-1994 with Base Year of 1993 for Central Copenhagen

Year	Passenger cars (without catalyst)	Passenger cars (catalyst)	Vans	Trucks	Bus
1960	3.68	-	3.68	4.00	4.00
1961	3.59	-	3.59	3.87	3.87
1962	3.51	-	3.51	3.70	3.70
1963	3.43	-	3.43	3.57	3.57
1964	3.35	-	3.35	3.43	3.43
1965	3.27	-	3.27	3.30	3.30
1966	3.19	-	3.19	3.17	3.17
1967	3.11	-	3.11	3.03	3.03
1968	3.02	-	3.02	2.87	2.87
1969	2.95	-	2.95	2.73	2.73
1970	2.86	-	2.86	2.57	2.57
1971	2.78	-	2.78	2.43	2.43
1972	2.70	-	2.70	2.30	2.30
1973	2.62	-	2.62	2.13	2.13
1974	2.54	-	2.54	2.00	2.00
1975	2.46	-	2.46	1.83	1.83
1976	2.38	-	2.38	1.70	1.70
1977	2.29	-	2.29	1.53	1.53
1978	2.22	-	2.22	1.40	1.40
1979	2.14	-	2.14	1.30	1.30
1980	2.05	-	2.05	1.23	1.23
1981	1.97	-	1.97	1.17	1.17
1982	1.89	-	1.89	1.10	1.10
1983	1.81	-	1.81	1.07	1.07
1984	1.73	-	1.73	1.07	1.07
1985	1.65	-	1.65	1.07	1.07
1986	1.56	-	1.56	1.07	1.07
1987	1.48	-	1.48	1.07	1.07
1988	1.41	-	1.41	1.03	1.03
1989	1.32	-	1.32	1.03	1.03
1990	1.24	1.00	1.24	1.03	1.03
1991	1.16	1.00	1.16	1.03	1.03
1992	1.08	1.00	1.08	1.00	1.00
1993	1.00	1.00	1.00	1.00	1.00
1994	1.00	1.00	1.00	1.00	1.00

Basic CO emission factors (g/km) at 40 km/h in 1993

Passenger cars (without catalyst)	Passenger cars (catalyst)	Vans	Trucks	Bus
26.9	11.0*	10.0	2.3	2.3

The emission factor for catalyst cars is too high since it represents an average catalyst car that is assumed to have a mileage of 107,000 km, and deterioration factors that correspond to this mileage. Since catalyst cars were introduced in 1990 the average catalyst car on the road has less mileage.

Table F.3 Development in NO_x Emission Factors (as index) from 1960 to 1994 with Base Year of 1993 for Central Copenhagen

Year	Passenger cars (not catalyst)	Passenger cars (catalyst)	Vans	Trucks	Bus
1960	0.70	-	1.00	0.41	0.41
1961	0.70	-	1.00	0.43	0.43
1962	0.70	-	1.00	0.45	0.45
1963	0.70	-	1.00	0.47	0.47
1964	0.70	-	1.00	0.50	0.50
1965	0.70	-	1.00	0.51	0.51
1966	0.75	-	1.00	0.53	0.53
1967	0.75	-	1.00	0.54	0.54
1968	0.75	-	1.00	0.56	0.56
1969	0.75	-	1.00	0.58	0.58
1970	0.75	-	1.00	0.59	0.59
1971	0.75	-	1.00	0.60	0.60
1972	0.75	-	1.00	0.62	0.62
1973	0.75	-	1.00	0.64	0.64
1974	0.75	-	1.00	0.65	0.65
1975	0.80	-	1.00	0.66	0.66
1976	0.85	-	1.00	0.68	0.68
1977	0.90	-	1.00	0.69	0.69
1978	0.90	-	1.00	0.71	0.71
1979	0.95	-	1.00	0.73	0.73
1980	1.00	-	1.00	0.75	0.75
1981	1.00	-	1.00	0.77	0.77
1982	1.00	-	1.00	0.79	0.79
1983	1.00	-	1.00	0.80	0.80
1984	1.00	-	1.00	0.83	0.83
1985	1.00	-	1.00	0.84	0.84
1986	1.00	-	1.00	0.86	0.86
1987	1.00	-	1.00	0.88	0.88
1988	1.00	-	1.00	0.90	0.90
1989	1.00	-	1.00	0.91	0.91
1990	1.00	1.00	1.00	0.93	0.93
1991	1.00	1.00	1.00	0.95	0.95
1992	1.00	1.00	1.00	0.98	0.98
1993	1.00	1.00	1.00	1.00	1.00
1994	1.00	1.00	1.00	1.00	1.00

Note: Since limited data is available of the development in emissions factors for vans they are assumed to be constant. The development in emission factors for buses has been assumed to be the same as for trucks.

Basic NO_x emission factors (g/km) at 40 km/h in 1993

Passenger cars (not catalyst)	Passenger cars (catalyst)	Vans	Trucks	Bus
2.1	0.5	1.7	10.7	10.7

Results

The development in traffic, CO and NO_x emissions for the central parts of Copenhagen is shown in Figure F.1. The trends in CO and NO_x emissions are used as indicator for the trends in CO and NO_x urban background concentrations from 1960-1994.

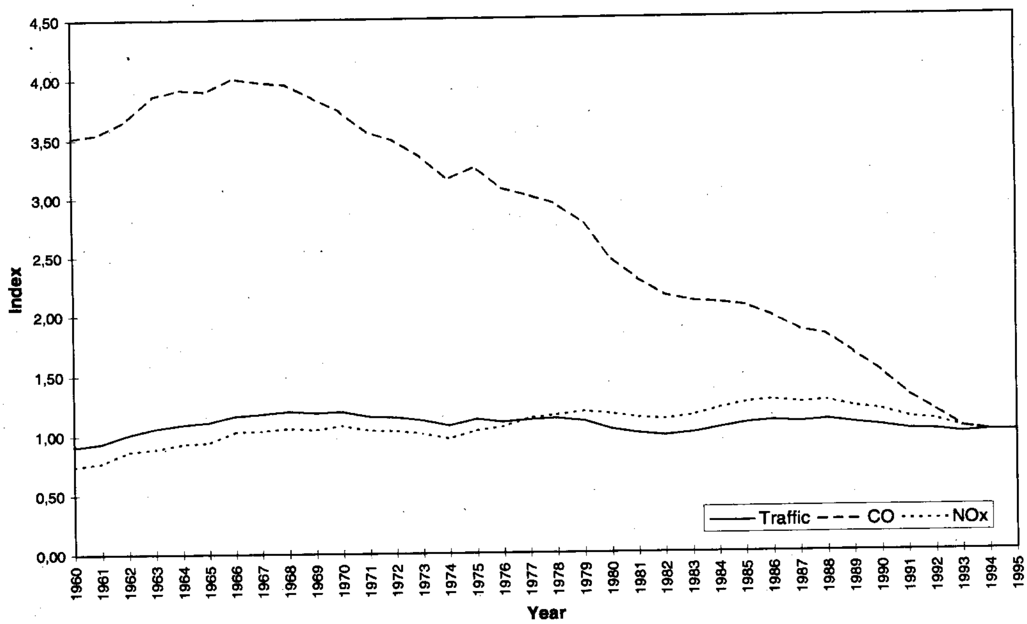


Figure F.1 Development in CO emission, NO_x emission and traffic levels from 1960 to 1995 in central parts of Copenhagen with 1994 as reference year

Appendix G

Trends in Rural CO and NO_x Emissions

Introduction

The Childhood Cancer Project requires an estimate of the development in CO and NO_x concentrations in rural areas since 1960. Monitoring of concentrations have only been carried out in recent years, therefore, the development in national road emissions have been taken as an indicator for the trends in concentrations in rural areas. Road emissions account for about 70 and 35 per cent of national emissions of CO and NO_x emissions, respectively (Fenhann, Kilde 1994).

The first Danish national CO and NO_x emission inventory began in 1972 (Fenhann, Kilde 1994). According to this reference CO emissions are more or less constant during 1972-94. However, the inventory has been established based on the energy consumption and applying constant CO emission factors (CO/energy) during 1972-89 and revised emissions factors during 1990-94 (Sorenson, private communication 1996a). The assumption that CO emission factors should have been constant for 1972-89 is fare too simplistic. Therefore, the development in CO and NO_x emissions has been calculated based on the actual development in national traffic performance and emission factors (g/km).

Methodology

The national traffic performance and traffic composition are based on data from the Danish Road Directorate and Statistics Denmark. (Danmarks Statistik 1995, 1996, Vejdirektoratet 1971, 1991).

The basic CO and NO_x emission factors for the reference year 1993 have been established based on a study from the Danish Road Directorate (Vejdirektoratet 1992). The development in emission factors is based on data from the Technical University of Denmark (Sorenson, private communication 1996b).

Data

The national traffic performance has been split into urban and rural traffic, and the average travel speeds on urban and rural roads have been assumed to be 40 km/h and 70 km/h, respectively.

Table G.1 Percentage of National Traffic Performance Classified as Urban and Rural Traffic

	Urban (40 km/h)	Rural (70 km/h)
Light vehicles	40	60
Heavy vehicles	50	50

The emission factors for the reference year 1993 are given below for 40 km/h and 70 km/h. The development in emission factors are the same as in Appendix F.

Table G.2 Basic CO Emission Factors (g/km) in 1993

Travel speed	Passenger cars (without catalyst)	Passenger cars (catalyst)	Vans	Trucks	Bus
40 km/h	26.9	11.0	10.0	2.3	2.3
70 km/h	17.0	3.0	7.0	1.5	1.0

Table G.3 Basic NO_x Emission Factors (g/km) in 1993

Travel speed	Passenger cars (without catalyst)	Passenger cars (catalyst)	Vans	Trucks	Bus
40 km/h	2.1	0.50	1.7	10.7	10.7
70 km/h	2.2	0.35	1.5	8.1	8.1

Results

The developments in national traffic performance, estimated national traffic emissions of CO and NO_x are shown in Figure G.1. The trends in CO and NO_x emissions are used as indicator for the trends in the rural background concentrations of CO and NO_x during 1960-1994.

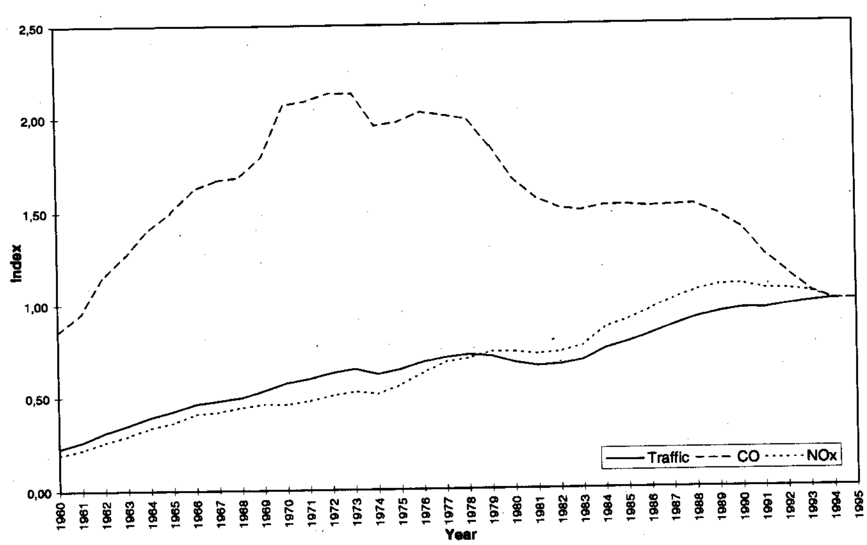


Figure G.1 Development in estimated national traffic CO and NO_x emission from 1960 to 1995 as an index with 1994 as reference year

Table G.4 Development in National Traffic Performance (billion km) and Traffic Composition (per cent)

Year	Total billion km	Passenger cars (not catalyst)	Passenger cars (catalyst)	Vans<3t	Trucks>3t	Bus
1960	9.10	62.3	0.0	17.5	17.5	2.2
1961	10.40	62.0	0.0	19.0	16.3	1.9
1962	12.30	65.9	0.0	18.2	14.3	1.6
1963	13.80	66.5	0.0	18.2	13.9	1.4
1964	15.50	67.1	0.0	18.1	13.5	1.3
1965	16.80	68.3	0.0	17.1	13.2	1.2
1966	18.30	69.9	0.0	16.0	12.5	1.1
1967	18.90	72.3	0.0	14.6	11.5	1.1
1968	19.50	73.0	0.0	14.4	11.3	1.5
1969	21.20	73.1	0.0	15.5	9.0	1.4
1970	22.60	85.4	0.0	8.3	5.0	1.3
1971	23.40	85.9	0.0	8.0	4.9	1.3
1972	24.60	85.8	0.0	8.0	5.0	1.2
1973	25.40	85.4	0.0	8.2	5.2	1.2
1974	24.30	84.4	0.0	8.7	5.7	1.2
1975	25.30	84.6	0.0	8.5	5.7	1.2
1976	26.80	84.7	0.0	8.4	5.7	1.1
1977	27.70	84.5	0.0	8.3	5.8	1.4
1978	28.30	84.5	0.0	8.3	5.9	1.4
1979	27.90	81.7	0.0	9.7	7.1	1.4
1980	26.50	81.1	0.0	9.9	7.4	1.5
1981	25.80	81.4	0.0	9.7	7.4	1.6
1982	26.00	81.5	0.0	9.5	7.4	1.5
1983	26.90	81.4	0.0	9.3	7.4	1.9
1984	28.30	81.6	0.0	11.2	9.1	1.8
1985	29.70	81.5	0.0	11.4	9.0	1.7
1986	31.50	80.6	0.0	11.7	9.1	1.6
1987	33.30	80.5	0.0	12.0	8.8	1.5
1988	34.80	81.0	0.0	12.2	8.5	1.4
1989	35.90	81.1	0.0	12.2	8.3	1.4
1990	36.60	79.8	1.6	12.1	7.9	1.4
1991	37.50	73.6	5.5	12.1	7.6	1.3
1992	38.40	69.6	9.5	12.0	7.4	1.3
1993	38.90	65.7	13.5	12.5	7.1	1.3
1994	39.40	59.2	19.7	12.9	6.9	1.3

Appendix H

Input Files

In table H.1 the input files are listed. The regions and variables they cover are given and the monitor stations from which the data is generated is also stated.

A copy of the input files are attached given the annual mean and indices for trends, monthly variation and monthly diurnal variation.

Table H.1 Geographic Regions, Variables and Monitor Stations Associated with Input Files

Region:	Input Filename	Variables:				Indicator Monitor Stations
		O_3	NO_x	NO_2	CO	
Urban:						
	CO1259				$[CO]_{CHP,centre}$	Copenhagen (1259)
	NOx1259		$[NO_x]_{CHP,centre}$			Copenhagen (1259)
Rural:						
All rural areas					$[CO]_{rural}$	0.5 of Copenhagen (1259)
Greater Copenhagen Area	NO22090			$[NO_2]_{rural}$		Lille Valby (2090)
Greater Copenhagen Area	O32090	$[O_3]_{rural}$				Lille Valby (2090)
Rest of Sealand	NO22082			$[NO_2]_{rural}$		Frederiksborg (2082), Lille Valby (2090)
Rest of Sealand	O32002	$[O_3]_{rural}$				Frederiksborg (2002)
Rest of Country	NO26083			$[NO_2]_{rural}$		Tange (6083), Frederiksborg (2082), Lille Valby (2090)
Rest of Country	O32002	$[O_3]_{rural}$				Frederiksborg (2002)

CO, Urban, Copenhagen 1259
Annual mean 1994 (ppm)
0.31

Trends	Index
1960	3.514
1961	3.545
1962	3.659
1963	3.857
1964	3.916
1965	3.900
1966	4.009
1967	3.975
1968	3.956
1969	3.838
1970	3.726
1971	3.552
1972	3.481
1973	3.345
1974	3.150
1975	3.253
1976	3.069
1977	3.008
1978	2.939
1979	2.775
1980	2.469
1981	2.292
1982	2.153
1983	2.102
1984	2.089
1985	2.061
1986	1.970
1987	1.855
1988	1.809
1989	1.654
1990	1.500
1991	1.309
1992	1.178
1993	1.036
1994	1.000
1995	1.000

Seasonal variation

YY	MM	Index
1995	1	1.456
1995	2	1.123
1995	3	1.061
1995	4	0.856
1995	5	0.864
1995	6	0.665
1995	7	0.639
1995	8	0.698
1995	9	0.855
1995	10	1.225
1995	11	1.223
1995	12	1.335
		12

Monthly diurnal variation

	Hour																									
YY	MM	0	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	All
1995	1	0.758	0.657	0.628	0.624	0.621	0.640	0.766	1.166	1.564	1.258	1.145	1.083	1.019	1.015	1.066	1.252	1.418	1.328	1.237	1.126	1.008	0.928	0.862	0.830	24.00
1995	2	0.881	0.788	0.717	0.694	0.704	0.721	0.881	1.197	1.402	1.141	1.042	1.056	1.030	1.009	1.052	1.146	1.224	1.212	1.145	1.065	0.953	0.978	1.007	0.955	24.00
1995	3	0.843	0.750	0.716	0.693	0.697	0.740	0.910	1.340	1.472	1.163	0.970	0.937	0.942	0.949	1.037	1.166	1.219	1.171	1.173	1.137	1.004	0.997	1.000	0.978	24.00
1995	4	0.960	0.823	0.789	0.766	0.770	0.793	0.937	1.225	1.300	1.059	0.969	0.956	0.982	0.984	1.046	1.091	1.163	1.086	1.051	1.003	1.010	1.081	1.073	1.080	24.00
1995	5	1.201	0.974	0.808	0.757	0.770	0.793	0.989	1.302	1.418	1.058	0.932	0.856	0.842	0.848	0.906	0.995	1.060	0.991	0.958	0.974	0.985	1.111	1.186	1.286	24.00
1995	6	1.094	0.894	0.717	0.683	0.676	0.712	0.925	1.313	1.412	1.092	0.984	0.986	0.981	0.982	0.969	1.059	1.101	1.070	1.018	0.948	0.939	1.027	1.204	1.213	24.00
1995	7	1.163	0.920	0.863	0.802	0.767	0.769	0.901	1.091	1.095	0.976	0.918	0.925	0.951	0.935	1.005	1.172	1.124	1.042	0.992	1.008	1.019	1.139	1.189	1.232	24.00
1995	8	1.053	1.043	0.819	0.775	0.742	0.739	1.124	1.405	1.447	1.051	0.884	0.838	0.858	0.837	0.897	0.980	1.007	1.019	0.990	0.968	1.021	1.144	1.236	1.124	24.00
1995	9	0.997	0.834	0.721	0.646	0.648	0.671	0.912	1.415	1.699	1.208	0.897	0.873	0.865	0.890	0.941	1.106	1.144	1.117	1.080	1.087	1.103	1.086	0.981	1.079	24.00
1995	10	0.846	0.700	0.622	0.574	0.552	0.598	0.765	1.224	1.589	1.143	0.981	0.864	0.826	0.853	0.927	1.136	1.337	1.439	1.448	1.307	1.121	1.078	1.057	1.012	24.00
1995	11	0.848	0.693	0.667	0.654	0.654	0.670	0.774	1.238	1.676	1.300	1.034	0.884	0.875	0.843	0.898	1.180	1.378	1.358	1.227	1.161	1.027	0.981	1.018	0.967	24.00
1995	12	0.985	0.796	0.767	0.714	0.701	0.632	0.723	0.978	1.263	1.144	0.999	0.990	1.021	1.006	1.053	1.187	1.328	1.304	1.297	1.100	1.017	0.980	1.009	1.004	

NOx (observed), Urban, Copenhagen (1259)
Annual mean 1994+1995 (ppb)
19.6

Trends

	Index
1960	0.745
1961	0.775
1962	0.870
1963	0.892
1964	0.933
1965	0.944
1966	1.032
1967	1.039
1968	1.060
1969	1.047
1970	1.080
1971	1.040
1972	1.036
1973	1.010
1974	0.966
1975	1.035
1976	1.064
1977	1.135
1978	1.160
1979	1.188
1980	1.167
1981	1.134
1982	1.118
1983	1.148
1984	1.207
1985	1.254
1986	1.274
1987	1.249
1988	1.262
1989	1.213
1990	1.184
1991	1.120
1992	1.097
1993	1.033
1994	1.000
1995	1.000

Seasonal variation

YY	MM	
1994/95	1	1.180
1994/95	2	0.893
1994/95	3	0.873
1994/95	4	0.824
1994/95	5	0.858
1994/95	6	0.720
1994/95	7	0.801
1994/95	8	0.891
1994/95	9	0.980
1994/95	10	1.273
1994/95	11	1.279
1994/95	12	1.427
		12

Monthly diurnal variation

YY	MM	Hour 0	1	2	3	4	5	6	7	8	9	10	11
1994/95	1	0.649	0.506	0.452	0.465	0.474	0.531	0.740	1.201	1.596	1.508	1.409	1.340
1994/95	2	0.717	0.612	0.549	0.512	0.532	0.607	0.893	1.445	1.707	1.438	1.248	1.183
1994/95	3	0.817	0.709	0.665	0.617	0.635	0.725	0.969	1.545	1.592	1.349	1.069	0.994
1994/95	4	0.854	0.699	0.621	0.585	0.593	0.720	1.029	1.471	1.582	1.346	1.260	1.101
1994/95	5	1.124	0.884	0.730	0.616	0.628	0.759	1.035	1.514	1.578	1.343	1.154	0.963
1994/95	6	1.284	1.051	0.841	0.712	0.678	0.796	1.036	1.536	1.547	1.274	1.079	1.094
1994/95	7	1.229	0.971	0.864	0.686	0.636	0.789	1.101	1.514	1.453	1.426	1.162	0.983
1994/95	8	1.049	0.818	0.726	0.628	0.655	0.826	1.344	1.888	1.693	1.244	1.000	0.875
1994/95	9	0.895	0.775	0.676	0.659	0.611	0.640	0.940	1.610	1.771	1.271	0.995	0.968
1994/95	10	0.797	0.667	0.581	0.519	0.519	0.630	0.917	1.505	1.722	1.332	1.059	0.890
1994/95	11	0.754	0.594	0.572	0.567	0.566	0.597	0.785	1.265	1.609	1.361	1.184	1.047
1994/95	12	0.792	0.635	0.573	0.539	0.557	0.592	0.776	1.147	1.528	1.412	1.222	1.136

YY	MM	12	13	14	15	16	17	18	19	20	21	22	23	All
1994/95	1	1.249	1.194	1.184	1.296	1.436	1.406	1.238	1.051	0.874	0.782	0.728	0.691	24.00
1994/95	2	1.127	1.094	1.154	1.158	1.241	1.224	1.083	1.022	0.920	0.887	0.878	0.768	24.00
1994/95	3	0.966	0.978	1.014	1.087	1.139	1.140	1.161	1.091	0.930	0.957	0.943	0.906	24.00
1994/95	4	1.098	1.024	0.996	1.076	1.070	1.072	0.997	0.939	0.902	0.994	1.013	0.959	24.00
1994/95	5	0.922	0.884	0.879	0.953	0.976	0.962	0.923	0.886	0.875	1.005	1.179	1.228	24.00
1994/95	6	0.974	0.885	0.914	0.949	0.889	0.866	0.815	0.715	0.705	0.867	1.157	1.336	24.00
1994/95	7	0.935	0.940	0.828	0.861	0.881	0.867	0.783	0.810	0.872	1.010	1.169	1.229	24.00
1994/95	8	0.866	0.835	0.817	0.897	0.922	0.900	0.851	0.849	0.882	1.106	1.207	1.123	24.00
1994/95	9	0.989	0.996	0.985	1.053	1.013	1.063	0.969	1.120	1.041	0.988	0.964	1.009	24.00
1994/95	10	0.857	0.864	0.908	1.051	1.198	1.358	1.405	1.294	1.068	1.000	0.944	0.915	24.00
1994/95	11	1.074	0.986	0.979	1.087	1.292	1.358	1.353	1.122	1.000	0.967	0.995	0.888	24.00
1994/95	12	1.141	1.108	1.155	1.252	1.341	1.300	1.262	1.062	0.921	0.863	0.864	0.821	24.00

CO, Rural, Lille Valby 2090 (annual level half of 1259)
Annual mean 1994 (ppm)
0.15

Trends

Trends	Index
1960	0.854
1961	0.954
1962	1.153
1963	1.275
1964	1.407
1965	1.503
1966	1.621
1967	1.669
1968	1.683
1969	1.795
1970	2.067
1971	2.088
1972	2.129
1973	2.127
1974	1.954
1975	1.973
1976	2.023
1977	2.006
1978	1.985
1979	1.842
1980	1.666
1981	1.562
1982	1.511
1983	1.494
1984	1.521
1985	1.522
1986	1.513
1987	1.517
1988	1.521
1989	1.470
1990	1.394
1991	1.256
1992	1.154
1993	1.051
1994	1.000
1995	1.000

Seasonal variation

YY	MM	Index
1995	1	1.456
1995	2	1.123
1995	3	1.061
1995	4	0.856
1995	5	0.864
1995	6	0.665
1995	7	0.639
1995	8	0.698
1995	9	0.855
1995	10	1.225
1995	11	1.223
1995	12	1.335
		12

Monthly diurnal variation

YY	MM	Hour 0	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	All
1995	1	0.758	0.657	0.628	0.624	0.621	0.640	0.766	1.166	1.564	1.258	1.145	1.083	1.019	1.015	1.066	1.252	1.418	1.328	1.237	1.126	1.008	0.928	0.862	0.830	24.00
1995	2	0.881	0.788	0.717	0.694	0.704	0.721	0.881	1.197	1.402	1.141	1.042	1.056	1.030	1.009	1.052	1.146	1.224	1.212	1.145	1.065	0.953	0.978	1.007	0.955	24.00
1995	3	0.843	0.750	0.716	0.693	0.697	0.740	0.910	1.340	1.472	1.163	0.970	0.937	0.942	0.949	1.037	1.166	1.219	1.171	1.173	1.137	1.004	0.997	1.000	0.978	24.00
1995	4	0.960	0.823	0.789	0.766	0.770	0.793	0.937	1.225	1.300	1.059	0.969	0.956	0.982	0.984	1.046	1.091	1.163	1.086	1.051	1.003	1.010	1.081	1.073	1.080	24.00
1995	5	1.201	0.974	0.808	0.757	0.770	0.793	0.989	1.302	1.418	1.058	0.932	0.856	0.842	0.848	0.906	0.995	1.060	0.991	0.958	0.974	0.985	1.111	1.186	1.286	24.00
1995	6	1.094	0.894	0.717	0.683	0.676	0.712	0.925	1.313	1.412	1.092	0.984	0.986	0.981	0.982	0.969	1.059	1.101	1.070	1.018	0.948	0.939	1.027	1.204	1.213	24.00
1995	7	1.163	0.920	0.863	0.802	0.767	0.769	0.901	1.091	1.095	0.976	0.918	0.925	0.951	0.935	1.005	1.172	1.124	1.042	0.992	1.008	1.019	1.139	1.189	1.232	24.00
1995	8	1.053	1.043	0.819	0.775	0.742	0.739	1.124	1.405	1.447	1.051	0.884	0.838	0.858	0.837	0.897	0.980	1.007	1.019	0.990	0.966	1.021	1.144	1.236	1.124	24.00
1995	9	0.997	0.834	0.721	0.646	0.648	0.671	0.912	1.415	1.699	1.208	0.897	0.873	0.865	0.890	0.941	1.106	1.144	1.117	1.080	1.087	1.103	1.086	0.981	1.079	24.00
1995	10	0.846	0.700	0.622	0.574	0.552	0.598	0.765	1.224	1.589	1.143	0.981	0.864	0.826	0.853	0.927	1.136	1.337	1.439	1.448	1.307	1.121	1.078	1.057	1.012	24.00
1995	11	0.848	0.693	0.667	0.654	0.654	0.670	0.774	1.238	1.676	1.300	1.034	0.884	0.875	0.843	0.898	1.180	1.378	1.358	1.227	1.161	1.027	0.981	1.018	0.967	24.00
1995	12	0.985	0.796	0.767	0.714	0.701	0.632	0.723	0.978	1.263	1.144	0.999	0.990	1.021	1.006	1.053	1.187	1.328	1.304	1.297	1.100	1.017	0.980	1.009	1.004	24.00

NO2 (observed), Rural, Lill Valby
Annual mean 1994+1995 (ppb)
6.8

Trends index

1960	0.358
1961	0.386
1962	0.440
1963	0.480
1964	0.505
1965	0.559
1966	0.600
1967	0.612
1968	0.636
1969	0.715
1970	0.764
1971	0.742
1972	0.768
1973	0.759
1974	0.751
1975	0.742
1976	0.804
1977	0.833
1978	0.889
1979	0.912
1980	0.894
1981	0.871
1982	0.889
1983	0.893
1984	0.943
1985	0.982
1986	1.040
1987	1.037
1988	1.032
1989	1.029
1990	0.975
1991	1.043
1992	0.948
1993	1.000
1994	1.000
1995	1.000

Seasonal variation

YY	MM	Index
1994/95	1	1.318
1994/95	2	1.198
1994/95	3	0.963
1994/95	4	0.846
1994/95	5	0.817
1994/95	6	0.496
1994/95	7	0.750
1994/95	8	0.664
1994/95	9	0.879
1994/95	10	1.202
1994/95	11	1.353
1994/95	12	1.513
		12

Monthly diurnal variation

Hour

YY	MM	0	1	2	3	4	5	6	7	8	9	10	11
1994/95	1	0.807	0.745	0.731	0.718	0.721	0.739	0.918	1.119	1.180	1.162	1.099	1.024
1994/95	2	0.904	0.866	0.864	0.832	0.821	0.863	0.994	1.212	1.276	1.166	1.078	0.964
1994/95	3	0.949	0.861	0.822	0.819	0.799	0.884	1.206	1.350	1.224	1.034	0.945	0.872
1994/95	4	1.172	1.038	0.903	0.832	0.809	0.867	1.113	1.442	1.445	1.199	0.946	0.949
1994/95	5	1.380	1.311	1.251	1.186	1.040	1.047	1.272	1.517	1.384	1.080	0.830	0.743
1994/95	6	1.522	1.398	1.204	1.028	0.970	1.053	1.368	1.444	1.224	0.962	0.841	0.759
1994/95	7	1.467	1.425	1.473	1.401	1.096	1.049	1.121	1.277	1.156	1.027	0.919	0.684
1994/95	8	1.536	1.476	1.368	1.107	0.946	0.975	1.275	1.706	1.648	1.159	0.690	0.504
1994/95	9	1.070	0.911	0.798	0.705	0.667	0.717	1.063	1.453	1.658	1.354	1.024	0.834
1994/95	10	0.982	0.862	0.762	0.692	0.683	0.735	1.035	1.229	1.226	1.094	0.963	0.793
1994/95	11	0.867	0.832	0.825	0.744	0.680	0.710	0.886	1.085	1.179	1.145	1.034	0.935
1994/95	12	0.928	0.887	0.839	0.799	0.798	0.829	0.954	1.096	1.204	1.157	1.069	1.001

YY	MM	12	13	14	15	16	17	18	19	20	21	22	23	All
1994/95	1	1.018	1.029	1.052	1.167	1.331	1.356	1.240	1.148	1.013	0.947	0.880	0.853	24.00
1994/95	2	0.904	0.859	0.864	0.959	1.139	1.185	1.190	1.088	1.013	1.010	0.983	0.967	24.00
1994/95	3	0.812	0.792	0.872	1.017	1.052	1.144	1.222	1.230	1.198	1.023	0.947	0.924	24.00
1994/95	4	0.852	0.806	0.747	0.689	0.783	0.741	0.786	0.963	1.136	1.210	1.241	1.332	24.00
1994/95	5	0.659	0.645	0.617	0.625	0.664	0.693	0.693	0.749	0.965	1.095	1.244	1.310	24.00
1994/95	6	0.761	0.693	0.626	0.734	0.717	0.774	0.689	0.670	0.758	0.958	1.326	1.520	24.00
1994/95	7	0.619	0.593	0.530	0.584	0.723	0.705	0.698	0.771	0.867	1.028	1.323	1.463	24.00
1994/95	8	0.546	0.494	0.520	0.545	0.566	0.559	0.561	0.674	0.931	1.224	1.444	1.546	24.00
1994/95	9	0.723	0.670	0.700	0.745	0.853	0.920	1.025	1.194	1.339	1.237	1.227	1.113	24.00
1994/95	10	0.727	0.785	0.800	0.936	1.178	1.350	1.415	1.293	1.226	1.118	1.066	1.049	24.00
1994/95	11	0.894	0.939	0.959	1.120	1.261	1.367	1.334	1.208	1.032	1.020	0.984	0.961	24.00
1994/95	12	0.951	0.946	0.972	1.140	1.299	1.243	1.133	1.076	0.956	0.907	0.931	0.888	24.00

O32090

O3 (observed), Rural, Lille Valby
Annual mean 1994+1995 (ppb)
26.9

Trends

	Index
1960	0.677
1961	0.690
1962	0.702
1963	0.715
1964	0.727
1965	0.739
1966	0.752
1967	0.764
1968	0.777
1969	0.789
1970	0.801
1971	0.814
1972	0.826
1973	0.839
1974	0.851
1975	0.863
1976	0.876
1977	0.888
1978	0.901
1979	0.913
1980	0.926
1981	0.938
1982	0.950
1983	0.963
1984	0.975
1985	0.988
1986	1.000
1987	1.000
1988	1.000
1989	1.000
1990	1.000
1991	1.000
1992	1.000
1993	1.000
1994	1.000
1995	1.000

Seasonal variation

YY	MM	Index
1994/95	1	0.679
1994/95	2	0.856
1994/95	3	1.133
1994/95	4	1.319
1994/95	5	1.326
1994/95	6	1.212
1994/95	7	1.418
1994/95	8	1.241
1994/95	9	0.927
1994/95	10	0.688
1994/95	11	0.599
1994/95	12	0.601
		12

Monthly diurnal variation

YY	MM	Hour 0	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	All
1994/95	1	1.064	1.082	1.063	1.052	1.042	1.023	0.945	0.879	0.870	0.904	0.956	1.014	1.042	1.060	1.064	1.036	0.938	0.904	0.932	0.963	1.022	1.046	1.051	1.047	24.00
1994/95	2	1.006	0.993	0.964	0.952	0.959	0.941	0.908	0.850	0.843	0.901	0.962	1.033	1.080	1.138	1.175	1.145	1.082	1.036	1.001	1.016	1.007	1.015	1.010	0.985	24.00
1994/95	3	0.971	0.973	0.967	0.968	0.964	0.942	0.878	0.853	0.874	0.944	1.005	1.045	1.088	1.115	1.108	1.105	1.086	1.067	1.039	1.002	1.002	1.007	1.010	0.988	24.00
1994/95	4	0.906	0.905	0.890	0.890	0.882	0.857	0.820	0.794	0.848	0.935	1.015	1.054	1.102	1.147	1.179	1.211	1.207	1.202	1.175	1.111	1.040	0.981	0.947	0.903	24.00
1994/95	5	0.821	0.814	0.802	0.774	0.763	0.741	0.725	0.773	0.869	0.966	1.048	1.095	1.157	1.198	1.232	1.250	1.263	1.260	1.251	1.215	1.126	1.039	0.948	0.870	24.00
1994/95	6	0.819	0.824	0.819	0.815	0.811	0.794	0.782	0.822	0.882	0.963	1.023	1.078	1.127	1.174	1.217	1.217	1.204	1.192	1.189	1.200	1.168	1.074	0.952	0.853	24.00
1994/95	7	0.701	0.660	0.596	0.567	0.565	0.546	0.560	0.659	0.821	0.974	1.095	1.206	1.314	1.381	1.430	1.448	1.439	1.412	1.372	1.338	1.224	1.049	0.876	0.768	24.00
1994/95	8	0.758	0.708	0.690	0.713	0.705	0.680	0.622	0.640	0.797	0.967	1.111	1.212	1.273	1.334	1.346	1.367	1.355	1.358	1.333	1.261	1.145	1.002	0.859	0.766	24.00
1994/95	9	0.842	0.890	0.872	0.853	0.844	0.836	0.771	0.716	0.719	0.869	1.037	1.144	1.219	1.278	1.298	1.328	1.292	1.271	1.206	1.078	0.963	0.912	0.886	0.877	24.00
1994/95	10	0.854	0.911	0.929	0.937	0.943	0.911	0.782	0.724	0.771	0.918	1.073	1.244	1.338	1.349	1.364	1.294	1.154	0.990	0.915	0.933	0.934	0.937	0.915	0.880	24.00
1994/95	11	1.037	1.012	1.005	1.025	1.032	1.039	0.956	0.871	0.845	0.867	0.995	1.108	1.150	1.157	1.151	1.051	0.958	0.893	0.888	0.928	1.018	1.010	1.009	0.993	24.00
1994/95	12	1.007	1.019	1.044	1.042	1.007	0.975	0.915	0.852	0.802	0.880	0.972	1.065	1.107	1.126	1.122	1.024	0.895	0.923	0.993	1.017	1.058	1.061	1.032	1.061	24.00

NO2 (observed), Rural, Frederiksborg 2082 (chemistry database)

Annual mean 1993 (ppb)

6.6

Trends index

1960	0.358
1961	0.386
1962	0.440
1963	0.480
1964	0.505
1965	0.559
1966	0.600
1967	0.612
1968	0.636
1969	0.715
1970	0.764
1971	0.742
1972	0.768
1973	0.759
1974	0.751
1975	0.742
1976	0.804
1977	0.833
1978	0.889
1979	0.912
1980	0.894
1981	0.871
1982	0.889
1983	0.893
1984	0.943
1985	0.982
1986	1.040
1987	1.037
1988	1.032
1989	1.029
1990	0.975
1991	1.043
1992	0.948
1993	1.000
1994	1.000
1995	1.000

Seasonal variation

YY	MM	Index
1990	1	1.402
1990	2	1.249
1990	3	0.936
1990	4	0.850
1990	5	0.801
1990	6	0.758
1990	7	0.550
1990	8	0.894
1990	9	0.694
1990	10	1.187
1990	11	1.325
1990	12	1.354
		12

Monthly diurnal variation

		Hour												
YY	MM	0	1	2	3	4	5	6	7	8	9	10	11	12
1994/95	1	0.807	0.745	0.731	0.718	0.721	0.739	0.918	1.119	1.180	1.162	1.099	1.024	1.018
1994/95	2	0.904	0.866	0.864	0.832	0.821	0.863	0.994	1.212	1.276	1.166	1.078	0.964	0.904
1994/95	3	0.949	0.861	0.822	0.819	0.799	0.884	1.206	1.350	1.224	1.034	0.945	0.872	0.812
1994/95	4	1.172	1.038	0.903	0.832	0.809	0.867	1.113	1.442	1.445	1.199	0.946	0.949	0.852
1994/95	5	1.380	1.311	1.251	1.186	1.040	1.047	1.272	1.517	1.384	1.080	0.830	0.743	0.659
1994/95	6	1.522	1.398	1.204	1.028	0.970	1.053	1.368	1.444	1.224	0.962	0.841	0.759	0.761
1994/95	7	1.467	1.425	1.473	1.401	1.096	1.049	1.121	1.277	1.156	1.027	0.919	0.684	0.619
1994/95	8	1.536	1.476	1.368	1.107	0.946	0.975	1.275	1.706	1.648	1.159	0.690	0.504	0.546
1994/95	9	1.070	0.911	0.798	0.705	0.667	0.717	1.063	1.453	1.658	1.354	1.024	0.834	0.723
1994/95	10	0.982	0.862	0.762	0.692	0.683	0.735	1.035	1.229	1.226	1.094	0.963	0.793	0.727
1994/95	11	0.867	0.832	0.825	0.744	0.680	0.710	0.886	1.085	1.179	1.145	1.034	0.935	0.894
1994/95	12	0.928	0.887	0.839	0.799	0.798	0.829	0.954	1.096	1.204	1.157	1.069	1.001	0.951

YY	MM	13	14	15	16	17	18	19	20	21	22	23	All
1994/95	1	1.029	1.052	1.167	1.331	1.356	1.240	1.148	1.013	0.947	0.880	0.853	24.00
1994/95	2	0.859	0.864	0.959	1.139	1.185	1.190	1.088	1.013	1.010	0.983	0.967	24.00
1994/95	3	0.792	0.872	1.017	1.052	1.144	1.222	1.230	1.198	1.023	0.947	0.924	24.00
1994/95	4	0.806	0.747	0.689	0.783	0.741	0.786	0.963	1.136	1.210	1.241	1.332	24.00
1994/95	5	0.645	0.617	0.625	0.664	0.693	0.693	0.749	0.965	1.095	1.244	1.310	24.00
1994/95	6	0.693	0.626	0.734	0.717	0.774	0.689	0.670	0.758	0.958	1.326	1.520	24.00
1994/95	7	0.593	0.530	0.584	0.723	0.705	0.698	0.771	0.867	1.028	1.323	1.463	24.00
1994/95	8	0.494	0.520	0.545	0.566	0.559	0.561	0.674	0.931	1.224	1.444	1.546	24.00
1994/95	9	0.670	0.700	0.745	0.853	0.920	1.025	1.194	1.339	1.237	1.227	1.113	24.00
1994/95	10	0.785	0.800	0.936	1.178	1.350	1.415	1.293	1.226	1.118	1.066	1.049	24.00
1994/95	11	0.939	0.959	1.120	1.261	1.367	1.334	1.208	1.032	1.020	0.984	0.961	24.00
1994/95	12	0.946	0.972	1.140	1.299	1.243	1.133	1.076	0.956	0.907	0.931	0.888	24.00

O3 (observed), Rural, Frederiksborg 2002
Annual mean 1994 (ppb)
24.5

Trends

	Index
1960	0.629
1961	0.643
1962	0.657
1963	0.671
1964	0.686
1965	0.700
1966	0.714
1967	0.729
1968	0.743
1969	0.757
1970	0.771
1971	0.786
1972	0.800
1973	0.814
1974	0.829
1975	0.843
1976	0.857
1977	0.871
1978	0.886
1979	0.900
1980	0.914
1981	0.929
1982	0.943
1983	0.957
1984	0.971
1985	0.986
1986	1.000
1987	1.000
1988	1.000
1989	1.000
1990	1.000
1991	1.000
1992	1.000
1993	1.000
1994	1.000
1995	1.000

Seasonal variation

YY	MM	
1990	1	0.783
1990	2	1.056
1990	3	1.357
1990	4	1.519
1990	5	1.297
1990	6	1.218
1990	7	1.060
1990	8	1.077
1990	9	0.850
1990	10	0.719
1990	11	0.494
1990	12	0.571
		12

Monthly diurnal variation

YY	MM	Hour 0	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	All
1990	1	1.029	1.030	1.053	1.096	1.075	1.120	1.072	0.994	0.926	0.877	0.896	0.980	1.011	1.022	1.027	1.067	0.972	0.938	0.908	0.914	0.975	0.981	0.992	1.047	24.00
1990	2	0.987	0.963	0.949	0.961	0.968	0.949	0.925	0.899	0.830	0.812	0.876	0.972	1.060	1.102	1.145	1.110	1.085	1.034	0.998	0.986	1.069	1.108	1.122	1.094	24.00
1990	3	0.953	0.933	0.910	0.918	0.904	0.904	0.868	0.793	0.732	0.793	0.963	1.080	1.141	1.197	1.224	1.236	1.216	1.208	1.113	1.032	0.986	0.990	0.968	0.937	24.00
1990	4	0.861	0.836	0.799	0.796	0.793	0.771	0.735	0.636	0.612	0.732	0.953	1.115	1.202	1.295	1.349	1.371	1.375	1.359	1.356	1.243	1.102	0.953	0.887	0.870	24.00
1990	5	0.599	0.597	0.542	0.533	0.545	0.512	0.486	0.503	0.637	0.888	1.124	1.245	1.370	1.436	1.495	1.543	1.568	1.535	1.536	1.450	1.293	1.045	0.844	0.675	24.00
1990	6	0.673	0.588	0.540	0.526	0.533	0.519	0.523	0.593	0.709	0.937	1.052	1.186	1.344	1.438	1.489	1.465	1.462	1.458	1.417	1.390	1.316	1.136	0.933	0.771	24.00
1990	7	0.704	0.648	0.680	0.670	0.651	0.616	0.578	0.568	0.668	0.860	1.045	1.140	1.261	1.351	1.390	1.467	1.471	1.414	1.402	1.356	1.270	1.085	0.902	0.802	24.00
1990	8	0.650	0.638	0.650	0.663	0.684	0.656	0.606	0.515	0.536	0.638	0.864	1.043	1.241	1.420	1.558	1.580	1.588	1.575	1.524	1.436	1.265	1.070	0.888	0.711	24.00
1990	9	0.756	0.799	0.759	0.723	0.724	0.690	0.667	0.615	0.587	0.671	0.866	1.071	1.247	1.389	1.458	1.459	1.494	1.491	1.430	1.283	1.149	0.959	0.877	0.834	24.00
1990	10	0.911	0.896	0.905	0.888	0.869	0.897	0.917	0.859	0.812	0.820	0.925	1.075	1.218	1.297	1.434	1.462	1.332	1.155	0.951	0.865	0.847	0.878	0.885	0.904	24.00
1990	11	0.869	0.955	0.966	1.046	1.122	1.173	1.146	1.048	0.912	0.858	0.917	0.940	1.145	1.343	1.329	1.264	1.058	0.894	0.812	0.848	0.828	0.839	0.848	0.840	24.00
1990	12	0.913	0.988	0.995	1.010	1.052	1.128	1.065	0.927	0.872	0.940	1.012	1.053	1.099	1.094	1.082	1.015	0.979	0.967	1.007	0.991	0.966	0.950	0.982	0.913	24.00

NO26083

NO2 Tange

Annual mean (ppb) 1.3.90-1.3.91

4.2

Trends	Index
1960	0.358
1961	0.386
1962	0.440
1963	0.480
1964	0.505
1965	0.559
1966	0.600
1967	0.612
1968	0.636
1969	0.715
1970	0.764
1971	0.742
1972	0.768
1973	0.759
1974	0.751
1975	0.742
1976	0.804
1977	0.833
1978	0.889
1979	0.912
1980	0.894
1981	0.871
1982	0.889
1983	0.893
1984	0.943
1985	0.982
1986	1.040
1987	1.037
1988	1.032
1989	1.029
1990	0.975
1991	1.043
1992	0.948
1993	1.000
1994	1.000
1995	1.000

Seasonal variation

YY	MM	Index
1990	1	1.402
1990	2	1.249
1990	3	0.936
1990	4	0.850
1990	5	0.801
1990	6	0.758
1990	7	0.550
1990	8	0.894
1990	9	0.694
1990	10	1.187
1990	11	1.325
1990	12	1.354
		12

Monthly diurnal variation

		Hour																								
YY	MM	0	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	All
1994/95	1	0.807	0.745	0.731	0.718	0.721	0.739	0.918	1.119	1.180	1.162	1.099	1.024	1.018	1.029	1.052	1.167	1.331	1.356	1.240	1.148	1.013	0.947	0.880	0.853	24.00
1994/95	2	0.904	0.866	0.864	0.832	0.821	0.863	0.894	1.212	1.276	1.166	1.078	0.964	0.904	0.859	0.864	0.959	1.139	1.185	1.190	1.088	1.013	1.010	0.983	0.967	24.00
1994/95	3	0.949	0.861	0.822	0.819	0.799	0.884	1.206	1.350	1.224	1.034	0.945	0.872	0.812	0.792	0.872	1.017	1.052	1.144	1.222	1.230	1.198	1.023	0.947	0.924	24.00
1994/95	4	1.172	1.038	0.903	0.832	0.809	0.867	1.113	1.442	1.445	1.199	0.946	0.949	0.852	0.806	0.747	0.689	0.783	0.741	0.786	0.963	1.136	1.210	1.241	1.332	24.00
1994/95	5	1.380	1.311	1.251	1.186	1.040	1.047	1.272	1.517	1.384	1.080	0.830	0.743	0.659	0.645	0.617	0.625	0.664	0.693	0.693	0.749	0.965	1.095	1.244	1.310	24.00
1994/95	6	1.522	1.398	1.204	1.028	0.970	1.053	1.368	1.444	1.224	0.962	0.841	0.759	0.761	0.693	0.626	0.734	0.717	0.774	0.689	0.670	0.758	0.958	1.326	1.520	24.00
1994/95	7	1.467	1.425	1.473	1.401	1.096	1.049	1.121	1.277	1.156	1.027	0.919	0.684	0.619	0.593	0.530	0.584	0.723	0.705	0.698	0.771	0.867	1.028	1.323	1.463	24.00
1994/95	8	1.536	1.476	1.368	1.107	0.946	0.975	1.275	1.706	1.648	1.159	0.690	0.504	0.546	0.494	0.520	0.545	0.566	0.559	0.561	0.674	0.931	1.224	1.444	1.546	24.00
1994/95	9	1.070	0.911	0.798	0.705	0.667	0.717	1.063	1.453	1.658	1.354	1.024	0.834	0.723	0.670	0.700	0.745	0.853	0.920	1.025	1.194	1.339	1.237	1.227	1.113	24.00
1994/95	10	0.982	0.862	0.762	0.692	0.683	0.735	1.035	1.229	1.226	1.094	0.963	0.793	0.727	0.785	0.800	0.936	1.178	1.350	1.415	1.293	1.226	1.118	1.066	1.049	24.00
1994/95	11	0.867	0.832	0.825	0.744	0.680	0.710	0.886	1.085	1.179	1.145	1.034	0.935	0.894	0.939	0.959	1.120	1.261	1.367	1.334	1.208	1.032	1.020	0.984	0.961	24.00
1994/95	12	0.928	0.887	0.839	0.799	0.798	0.829	0.954	1.096	1.204	1.157	1.069	1.001	0.951	0.946	0.972	1.140	1.289	1.243	1.133	1.076	0.956	0.907	0.931	0.888	24.00

Appendix I

The Questionnaire

The address ______________________________

The time period ______________________________

(This part is filled out by the Danish Cancer Society)

The questions below refer to the middle of the time period considered.

If there have been major traffic changes during the period please indicate this in question No. 10.

For all questions you are requested to make the best judgement when precise information is not available.

1. Was the type of road mentioned on the front page , a

1) ___ *municipality street or private street*
2) ___ *county road, give county number ___, administrative road number ___, and kilometric position at address*
3) ___ *state road, give administrative number ___ and kilometric position at address ___.*

Note: If it is a county or state road and the municipality does not have traffic figures, go to question No. 4. If the municipality has traffic data, or these can be estimated, you are requested to answer question No. 2 and No. 3 for county and state roads.

2. How much traffic was there on the street at the address?

___ *vehicles/day (Average Daily Traffic)*

The Average Daily Traffic is the annual average for all 365 days for traffic in both directions. Please indicate if the figure is given in other terms than the Average Daily Traffic.

3. How much heavy traffic (buses and vans over 3500 kg) did the street carry?

The exact fraction of heavy traffic was ___ %

If the exact fraction is not known, you are requested to use your best judgement to choose one of the below options

1) ___ *Almost no heavy traffic (less than 1% of traffic)*
2) ___ *Small fraction (1-4% of traffic)*
3) ___ *Moderate fraction (5-8% of traffic)*
4) ___ *Large fraction (9% of traffic or more)*

4. What was the actual mean speed at the address?

If the actual speed cannot be estimated, use the speed limit as a basis for an estimate. Unsteady driving patterns due to e.g. traffic lights lowers the mean driving speed.

1) ___ *00-35 km/h*
2) ___ *35-45 km/h*
3) ___ *45-55 km/h*
4) ___ *above 65 km/*

5. Which of the below road surroundings suits best the address: (A-E) ___.

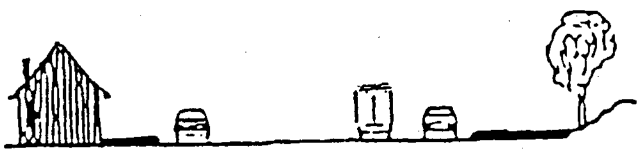
A A house in an open street. The house can be situated close to or far from the street. There may be other houses nearby.

What was the distance from the facade of the address house to the furthest road edge on the furthest driving lane?
About ___ m

B Low scattered houses. Choose the one which fits best.

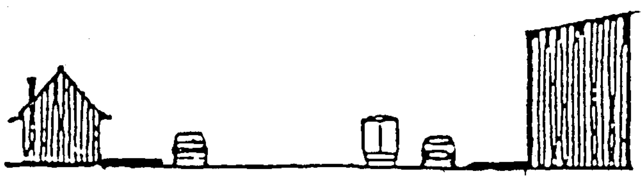
1) ___ *A row of low houses on one side of the street. Almost no houses on the other side.*
2) ___ *Villas on both sides with space (gardens) in between.*
3) ___ *Low houses with open front areas (parking lots, gardens etc.)*

What was the distance from the facade of the address house to the furthest road edge on the furthest driving lane?
Approximate distance ___ m
How high are the houses at the address side of the street?
___ *storeys*
How high are the houses on the other side of the street (if no houses write 0)?
___ *storeys*



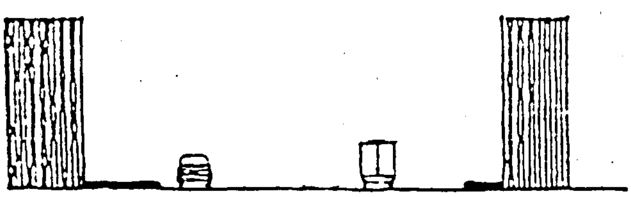
C Low houses on one side of the street and high houses on the other side of the street.

What was the distance between the house facades?
Approximately ___ m
How high are the houses on the other side of the street?
___ *storeys*



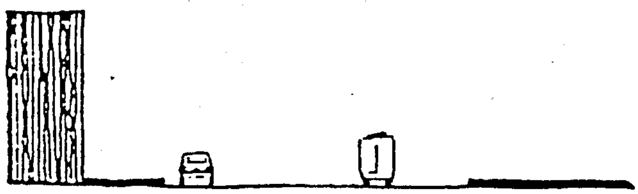
D High houses on both sides of the street

What was the distance between the house facades?
Approximately ___ m
How high are the houses on the address side of the street?
___ *storeys*
How high are the houses on the other side of the street?
___ *storeys*



E High houses on one side of the street. Almost no houses on the other side.

What was the distance from the address house facade to the furthest road edge on the furthest driving lane?
Approximately ___ m
How high is the address house?
___ storeys
Was the address side
1) ___ *On the side of the street with high houses*
2) ___ *A single house on the other side*



6. Was there within 50 m from the address a cross road, which had more traffic than the street at which the address is located?
1) ___ *Yes*
2) ___ *No*

If yes, how many vehicles were on this street? (If there were several cross roads, answer for the most trafficked)
1) ___ *0-2,000 vehicles/day*
2) ___ *2,000-5,000 vehicles/day*
3) ___ *5,000-10,000 vehicles/day*
4) ___ *10,000-15,000 vehicles/day*
5) ___ *more than 15,000 vehicles/day*

7. How was the traffic in the quarter around the address? (Within 300 m around the address)
1) ___ *Rural area or other, where almost no other streets than the one with the address*
2) ___ *Urban areas with residential or low speed streets, or other streets with low traffic intensity (only local traffic)*
3) ___ *Urban traffic, however without heavily trafficked streets (below 10,000 vehicles/day)*
4) ___ *Heavy urban traffic, where one or more of the streets were heavily trafficked (above 10,000 vehicles/day)*
5) ___ *Other*

8. How was the main part of the houses in the quarter around the address? (Within 300 m around the address)
1) ___ *No or almost no houses*
2) ___ *Low density built-up areas (e.g. villages, small towns and residential neighbourhoods)*
3) ___ *Semi-dense built-up areas (e.g. 2-3 storey buildings in central areas in middle-sized cities)*
4) ___ *Scattered multi-storey buildings (e.g. Gellerup-Parken in Aarhus)*
5) ___ *Dense built-up areas (e.g. 4-6 storey buildings with street in between, e.g. "Bro-kvartererne" in Copenhagen)*

9. How many inhabitants had the city in which the address was situated? (A city is a coherent built-up area. If almost all of the municipality is urban area, the municipality is regarded as one city. Copenhagen and Frederiksberg Municipalities are regarded as one city),

1) ___ *The address was located within a urban area (go to question No. 10)*
2) ___ *Less than 2,000 inhabitants (go to question No. 10)*
3) ___ *2,000-20,000 inhabitants (go to question 10)*
4) ___ *20,000-40,000 inhabitants*
5) ___ *40,000-80,000 inhabitants*
6) ___ *80,000-150,000 inhabitants*
7) ___ *More than 150,000 inhabitants*

If the city had more than 20,000 inhabitants:

What was the distance (direct line) from the address to the centre of the city? *Approximately* ___ *km.*

What was the distance (direct line) from the address to a larger area without built-up area, e.g. forest, field or water? *Approximately* ___ *km.*

A larger area has to be at least 1 x 1 km^2 e.g. Horsens Fjord, Jægersborg Dyrehave, Hareskov at Værløse, Vest-amager and Utterslev Mose at Copenhagen. The following are **not** large enough: Sports stadium, parks and smaller lakes (e.g. Damhussøen at Rødovre).

10. Are there special conditions, which you think should be mentioned?

If there have been major traffic changes during the period given on the front page, please indicate what has happened and when it happened

____

____

____

Thank you for your help.

11. This questionnaire has been filled out

by ______________________________ /

Institution______________________________

National Environmental Research Institute

The National Environmental Research Institute, NERI, is a research institute of the Ministry of Environment and Energy. In Danish, NERI is called *Danmarks Miljøundersøgelser (DMU).*
NERI's tasks are primarily to conduct research, collect data, and give advice on problems related to the environment and nature.

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Publications:
NERI publishes professional reports, technical instructions, and the annual report. A R&D projects' catalogue is available in an electronic version on the World Wide Web.
Included in the annual report is a list of the publications from the current year.

Faglige rapporter fra DMU/NERI Technical Reports

Nr. 203: Rådyr, mus og selvforyngelse af bøg ved naturnær skovdrift. Af Olesen, C.R., Andersen, A.H. & Hansen, T.S. 60 s., 80,00 kr.

Nr. 204: Spring Migration Strategies and Stopover Ecology of Pink-Footed Geese. Results of Field Work in Norway 1996. By Madsen, J. et al. 29 pp., 45,00 DKK.

Nr. 205: Effects of Experimental Spills of Crude and Diesel Oil on Arctic Vegetation. A Long-Term Study on High Arctic Terrestrial Plant Communities in Jameson Land, Central East Greenland. By Bay, C. 44 pp., 100,00 DKK.

Nr. 206: Pesticider i drikkevand 1. Præstationsprøvning. Af Spliid, N.H. & Nyeland, B.A. 273 pp., 80,00 kr.

Nr. 207: Integrated Environmental Assessment on Eutrophication. A Pilot Study. Af Iversen, T.M., Kjeldsen, K., Kristensen, P., de Haan, B., Oirschot, M. van, Parr, W. & Lack, T. 100 pp., 150,00 kr.

Nr. 208: Markskader forvoldt af gæs og svaner - en litteraturudredning. Af Madsen, J. & Laubek, B. 28 s., 45,00 kr.

Nr. 209: Effekt af Tunø Knob vindmøllepark på fuglelivet. Af Guillemette, M., Kyed Larsen, J. & Clausager, I. 31 s., 45,00 kr.

Nr. 210: Landovervågningsoplande. Vandmiljøplanens Overvågningsprogram 1996. Af Grant, R., Blicher-Mathiesen, G., Andersen, H.E., Laubel, A.R., Grevy Jensen, P. & Rasmussen, P. 141 s., 150,00 kr.

Nr. 211: Ferske vandområder - Søer. Vandmiljøplanens Overvågningsprogram 1996. Af Jensen, J.P., Søndergaard, M., Jeppesen, E., Lauridsen, T.L. & Sortkjær, L. 103 s., 125,00 kr.

Nr. 212: Atmosfærisk deposition af kvælstof. Vandmiljøplanens Overvågningsprogram 1996. Af Ellermann, T., Hertel, O., Kemp, K., Mancher, O.H. & Skov, H. 88 s., 100,00 kr.

Nr. 213: Marine områder - Fjorde, kyster og åbent hav. Vandmiljøplanens Overvågningsprogram 1996. Af Jensen, J.N. et al. 124 s., 125,00 kr.

Nr. 214: Ferske vandområder - Vandløb og kilder. Vandmiljøplanens Overvågningsprogram 1996. Af Windolf, J., Svendsen, L.M., Kronvang, B., Skriver, J., Olesen, N.B., Larsen, S.E., Baattrup-Pedersen, A., Iversen, H.L., Erfurt, J., Müller-Wohlfeil, D.-I. & Jensen, J.P. 109 s., 150,00 kr.

Nr. 215: Nitrogen Deposition to Danish Waters 1989 to 1995. Estimation of the Contribution from Danish Sources. By Hertel, O. & Frohn, L. 53 pp., 70,00 DKK.

Nr. 216: The Danish Air Quality Monitoring Programme. Annual Report for 1996. By Kemp, K., Palmgren, F. & Mancher, O.H. 61 pp., 80,00 DKK.

Nr. 217: Indhold af organiske opløsningsmidler og phthalater i legetøj. Analytisk-kemisk kontrol af kemiske stoffer og produkter. Af Rastogi, S.C., Worsøe, I.M., Køppen, B., Hansen, A.B. & Avnskjold, J. 34 s., 40,00 kr.

Nr. 218: Vandføringsevne i danske vandløb 1976-1995. Af Iversen, H.L. & Ovesen, N.B. 2. udg. 55 s., 50,00 kr.

Nr. 219: Kragefuglejagt i Danmark. Reguleringen af krage, husskade, skovskade, råge og allike i sæsonen 1990/91 og jagtudbyttet i perioden 1943-1993. Af Asferg, T. & Prang, A. 58 s., 80,00 kr.

Nr. 220: Interkalibrering af bundvegetationsundersøgelser. Af Middelboe, A.L., Krause-Jensen, D., Nielsen, K. & Sand-Jensen, K. 34 s., 100,00 kr.

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Nr. 221: Pollution of the Arctic Troposphere. Northeast Greenland 1990-1996. By Heidam, N.Z., Christensen, J., Wåhlin, P. & Skov, H. 58 pp., 80,00 DKK.

Nr. 222: Sustainable Agriculture and Nature Values - using Vejle County as a Study Area. By Hald, A.B. 93 pp., 100,00 DKK.

Nr. 223: Ændringer i bekæmpelsesmidlernes egenskaber fra 1981-1985 frem til 1996. Af Clausen, H. 61 s., 45,00 kr.

Nr. 224: Natur og Miljø 1997. Påvirkninger og tilstand. Red. Holten-Andersen, J., Christensen, N., Kristiansen, L.W., Kristensen, P. & Emborg, L. 288 s., 190.00 kr.

Nr. 225: Sources of Phthalates and Nonylphenoles in Municipal Waste Water. A Study in a Local Environment. By Vikelsøe, J., Thomsen, M. & Johansen, E. 50 pp., 45,00 kr.