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SHAPE

The Stockholm Study on Health Effects of Air Pollution and their Economic Consequences

Part I:

NO_2 and Particulate Matter in Stockholm

—

Concentrations and Population Exposure

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By

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SHAPE

The Stockholm Study on Health Effects of Air Pollution and their Economic Consequences

Part I:

NO_2 and Particulate Matter in Stockholm Concentrations and Population Exposure

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Abstract

This report presents air quality dispersion calculations of the ambient levels of NO_2 and particulate matter in the Stockholm region. The calculations are based on an Information and Air Quality Assessment System that is administered by the Regional Association for Air Quality Management in the counties of Stockholm and Uppsala. Detailed information about air pollutant sources is provided once a year from the members of the Association, which consists of about 30 municipalities and regional administrations in the counties.

Information on the emissions of NO_x and particulate matter is used to calculate the dispersion of these compounds for five different areas with a resolution ranging from 100 m to 2000 m. Road traffic emissions are calculated from detailed information on traffic volumes, vehicle compositions and driving conditions. The database also includes emissions from burning of different fuels in district and residential heating, emissions from ships and a special model for re-suspension of particulate matter from road dust. The re-suspension model considers the influence of meteorological conditions, the wetness of the road surface and effect of the traffic on the re-suspension rate.

Some of the information in the emission database has been validated by comparison with independent data. Excellent agreement is found between gasoline, diesel and fuel oil volumes delivered to the region according to official statistics and calculated consumption (combustion) of these fuels based on information in the emission database (e.g. traffic volumes and vehicle gasoline consumption rate, litre/vehicle-kilometre). The most uncertain emission data for particulate matter and NO_2 are wood combustion and off-road vehicles.

A Gaussian Air Quality Dispersion model is used to calculate the temporal and spatial distribution of NO_2 and particulate matter in the region. In order to estimate population exposures daytime and night-time concentrations are combined with daytime and night-time population data. Mean values as well as extreme (98-percentile) values have been calculated. Comparison of calculated concentrations with measurements in the city of Stockholm shows a very good agreement.

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Preface

Air pollution in urban areas in Sweden continues to pose a threat to an ecologically sustainable society and to human health in particular. Vehicle emissions are one of the major sources of pollution in Swedish cities. In its endeavour to improve urban air quality, the Swedish National Road Administration (Vägverket), together with the Swedish Foundation for Health Care Sciences and Allergy Research (Vårdalstiftelsen) have been a co-financier of SHAPE (The Stockholm Study on Health Effects of Air Pollution and their Economic Consequences). Although Stockholm was selected as the example of an urban area with air pollution problems, it will later be possible to apply the methodology to other urban areas.

The project, which was initiated by the National Institute of Public Health, is expected to provide knowledge on exposure to air pollution and its impact on health, including the socio-economic costs involved. It is also expected to help create a better basis for investment analyses and initiate new planning strategies within the road transport system.

SHAPE is a project undertaken in co-operation between the following public authorities: the National Institute of Public Health, the Swedish Environmental Protection Agency and the Swedish National Road Administration as well as researchers at the Environmental and Health Protection Administration of the city of Stockholm, the Environmental Medicine Department at the Stockholm County Council and the Department of Applied Environmental Science at Göteborg University.

The primary aims of the project are to:

- increase the knowledge about emission, dispersion and concentrations of nitrogen dioxide and fine particles
- estimate the exposure of these air pollutants to the general public
- quantify the major health effects of nitrogen dioxide and fine particles, as well as the specific role of traffic emissions of these pollutants
- estimate the socio-economic cost of health impacts from nitrogen oxides and fine particles in Stockholm and to develop methods for economic valuation of such emissions.

This is the first of three reports within *the Stockholm study on health effects of air pollution and their economic consequences*. The following two reports are supposed to be published during the spring of 1999.

The National Institute of Public Health, the Swedish Environmental Protection Agency and the Swedish National Road Administration would like to express their great appreciation to all those who, through their comments and viewpoints, have made a valuable contribution to the work on the project.

Contents

1. Introduction

This report presents dispersion calculations for the SHAPE project — *The Stockholm Study on Health effects of Air Pollution and their Economic Consequences*. For the air quality calculations an integrated system for air quality management have been used. The system is administered by the Air Quality Management Association of Stockholm and Uppsala and consists of databases and dispersion models for evaluation of past, present and future air quality. It is operated by personnel at the Environment and Health Protection Administration of Stockholm. A number of continuous measurements including air pollution and meteorological measurements are part of the system.

The Air Quality Management Association of Stockholm and Uppsala is a non-profit organisation that co-ordinate activities for cost-effective assessment and supervision of the air quality and deposition of air pollutants within the region. The members consist of about 30 municipalities and the two county councils. The activity is operated by the members in co-operation with the county administrative board in the counties of Stockholm and Uppsala.

2. Air pollutants in Stockholm

Air pollution has been monitored in Stockholm since the middle of the 1960's. Originally SO_2 and black smoke was studied. Today there is a network of monitoring stations that include measurements of e. g. NOx, CO, SO_2, PAH and a number of meteorological parameters. During the last few years particulate matter (PM_{10}) has also been measured, and starting from 1998 continuous measurements of organic and elemental carbon and benzene, toluene and xylenes have been included at one station.

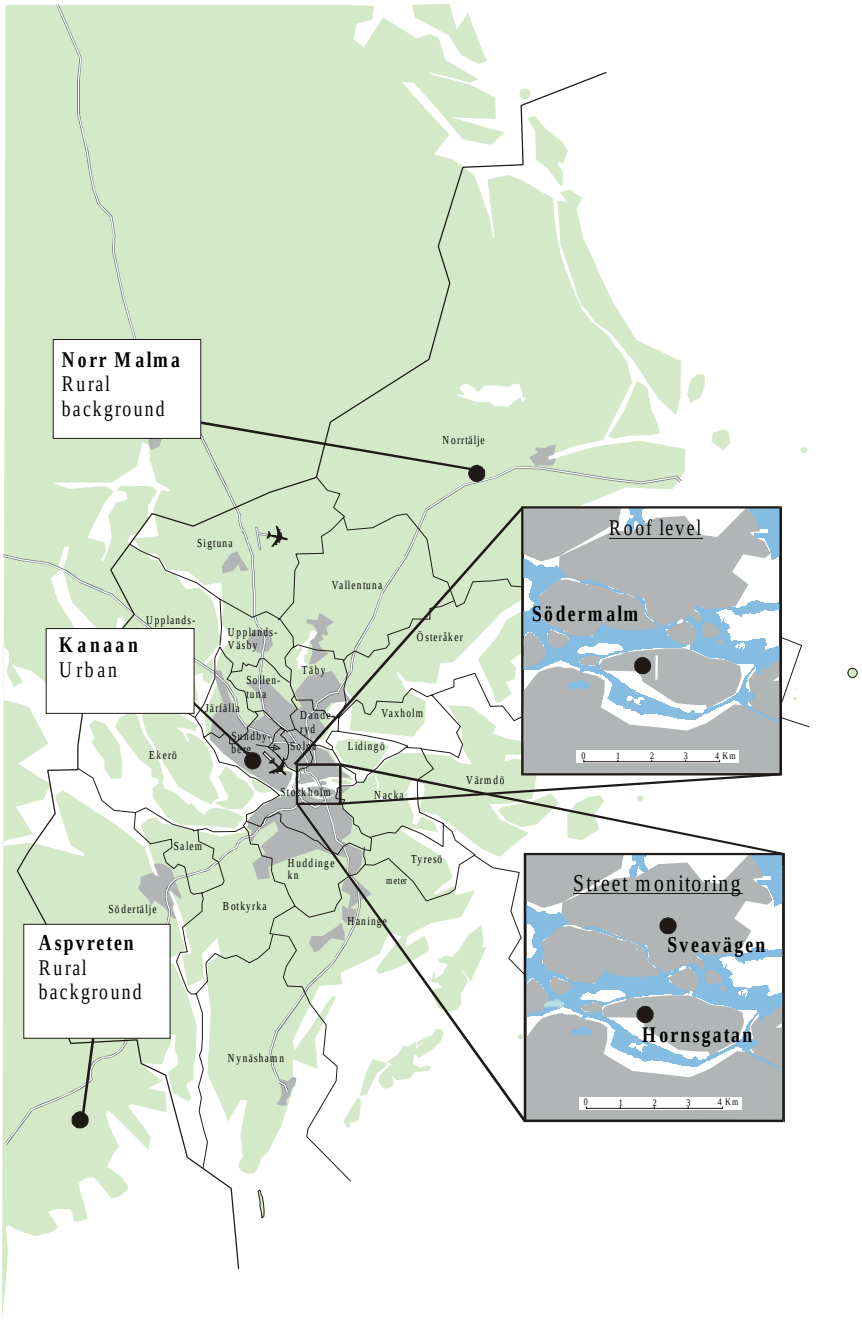


Figure 1. Air pollution monitoring stations in the county of Stockholm.

The monitoring network includes both roof level and street level measurements and several different methods are employed. Nitrogen dioxide and sulphur dioxide are measured using both passive samplers producing monthly averages and active instruments producing hourly averages. Polycyclic aromatic hydrocarbons are measured during the spring (April to June) at one street level station (Hornsgatan). Apart from the monitoring stations indicated in the figure above, several short-term (up to one year) campaigns provide complementing information on the geographical distribution of some air pollutants, mainly NO_2, SO_2 and some monoaromatic compounds (bensene, toluene and xylenes).

There are five meteorological masts in the region. They provide hourly data of a number of parameters. In the southern part of Stockholm there is a 50 metre high mast equipped with several meteorological instruments. The parameters are: wind speed (two heights), wind direction (one height), temperature (absolute and difference between two heights), global radiation, relative humidity, precipitation amount (hourly). In the top of the mast there is a sonic anemometer that gives information on the turbulence.

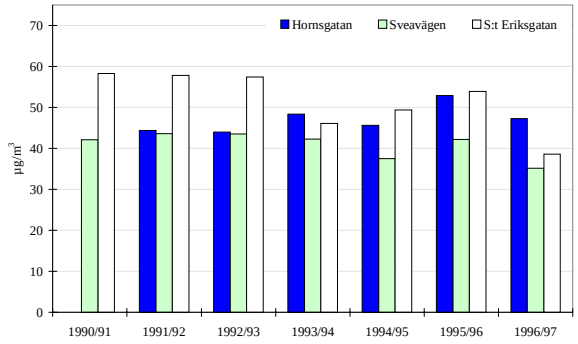
In addition to the monitoring stations mentioned above there are several stations outside the city of Stockholm. These include both air pollutants and meteorological parameters and provide important information on the background levels and input to air quality dispersion models (Luftvårdsförbundet, 1997).

In general there has been a decreasing trend of air pollutant levels in Stockholm during the past decades. For SO_2 and CO the trend has continued during the 90's (Figure 2). For the nitrogen oxides (sum of NO and NO_2) there was a decrease in the winter season mean values between 1990/91 and 1994/95, but then the concentrations increased in 1995/96 to even higher levels than in the early 1990's. The increased concentrations are probably due to a combination of relatively cold winter and an increase in both the total traffic flows as well as in the number of heavy trucks on Hornsgatan (Burman and Höglund, 1998). The health limit value for NO_2 is still exceeded in streets with dense traffic in central Stockholm. For sulphur dioxide (SO_2) and carbon monoxide (CO) the concentrations are well below the health limit values. For ozone there is no significant trend in the winter season mean values.

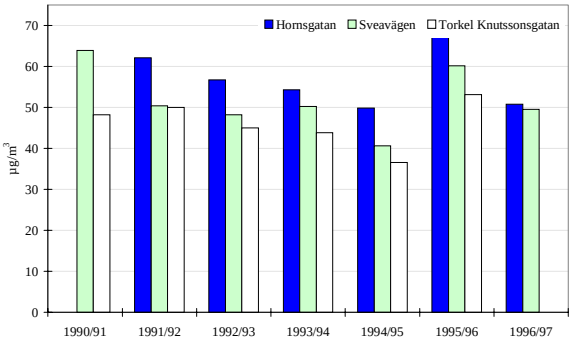
Particulate matter or simply particulates, consist of solid or liquid particles that are small enough to remain in the air for hours to weeks. Most measurements in Stockholm and in other cities have been done collecting the particles on a filter and determining the blackening of the filter paper by measuring the light reflectance. This is a measure of the "black smoke" content, it is not a good measure of particulate matter. Very few long-term measurements have been made. Thus it is difficult to see if there is an increasing or decreasing trend for particles. It seems that the levels in Gothenburg have been halved since the middle of the 70´s (Brandberg, 1995).

PM_{10} (particles with a diameter less 10 µm) have been measured at a busy street in the city centre of Stockholm and at roof level since 1992 and 1993 respectively. The average levels are between 30 and 40 µg/m^3 in street canyons in the city of Stockholm and 10 to 20 µg/m^3 at roof level. The measurements show no systematic trend. There is no limit value for the concentration of particles in urban air in Sweden. There are, however, guide lines stating that the 24-hour mean value should not be higher than 110 µg/m^3 as a 98 percentile, and the winter season mean value should not be higher than 50 µg/m^3 (October to March). Much

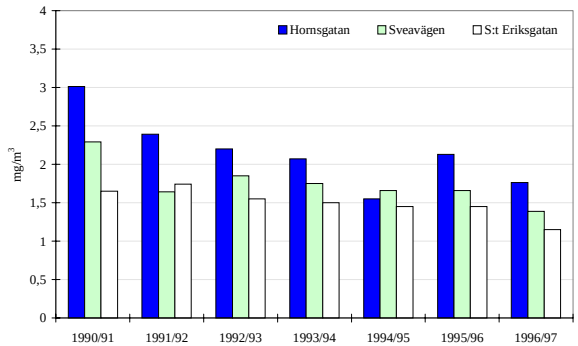
lower limit values have recently been introduced for $PM_{2.5}$ in the USA. Also in Europe a decrease of present quality standards for the Member States of EU has been suggested (see http://www.environ.se).



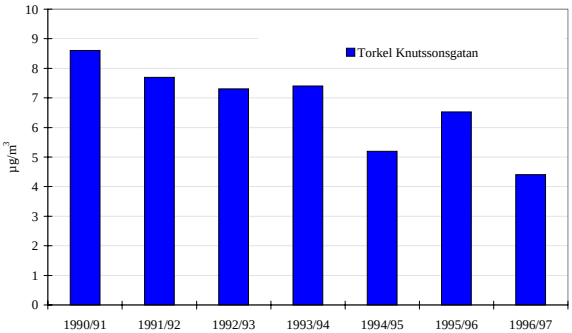
Nitrogen dioxide (street level)



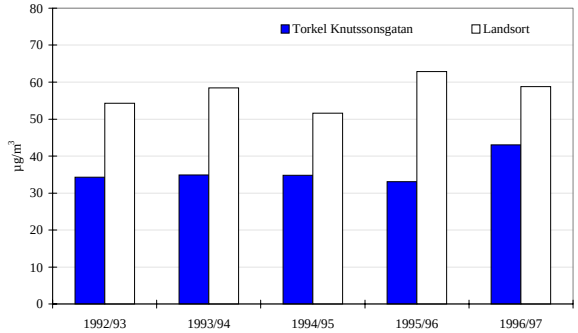
Nitrogen oxides, NO plus NO_2 (Hornsgatan and Sveav. are street level; Torkel Knuts. g. is roof level)



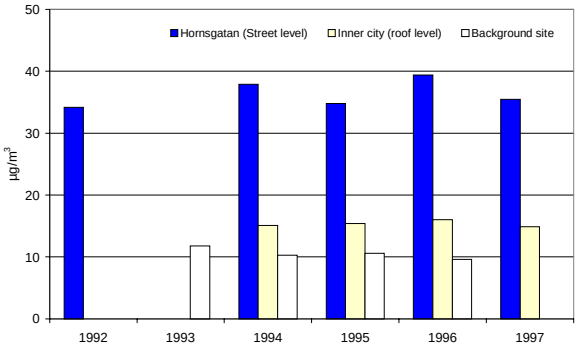
Carbon monoxide (street level)



Sulphur dioxide (roof level)



Ozone (roof level)



Particulate matter (PM_{10})

Figure 2. Air pollutant levels during the 1990's in Stockholm. All values except PM_{10} data are winter season mean values (October – March). All data are from the air pollution supervision of the Environment and Health Protection Administration of Stockholm and the Air Quality Management Association of Stockholm and Uppsala (Slb-analys, 1998; http://www.slb.mf.stockholm.se).

The current Swedish recommended guide line values for PM_{10} are probably not exceeded anywhere in Stockholm, but the new values that may be adopted for all EU countries will be exceeded. The annual mean particle concentrations of PM_{10} at

Swedish background sites range from 3 to 12 µg/m^3 (Kyrklund et al, 1998). Simultaneous wintertime measurements at a number of sites have demonstrated the importance of long-range transport for the PM_{10} concentrations across Europe (Hoek et al., 1997).

In Swedish cities, mean concentrations range from 10 to 40 µg/m^3. At roof level the range is 10 to 20 µg/m^3. Highest concentrations are found at street level with intensive traffic. The mean background concentration of PM_{10} at Aspvreten is around 10 µg/m^3.

Volatile and semi-volatile organic compounds such as bensene, formaldehyde and polycyclic aromatic hydrocarbons are also present at levels close to or above recommended health limit values in Stockholm. Likewise, ozone concentrations exceed the health limit of 110 µg/m^3 (8-hour average) on several occasions during the summer season.

Two different size classes will be reported here — fine and coarse particles. Fine particles are measured using a cut point at 2.5 µm ($PM_{2.5}$; PM = particulate matter). Most measurements in Stockholm are made using a 10 µm inlet (PM_{10}). The difference between the PM_{10} and $PM_{2.5}$ concentrations are often referred to as coarse particles, even though, this is only a fraction of all particles in the whole coarse particle size range as defined in the figure below. See also ISO 7708, 1995 for definitions and terminology regarding particle size fractions.

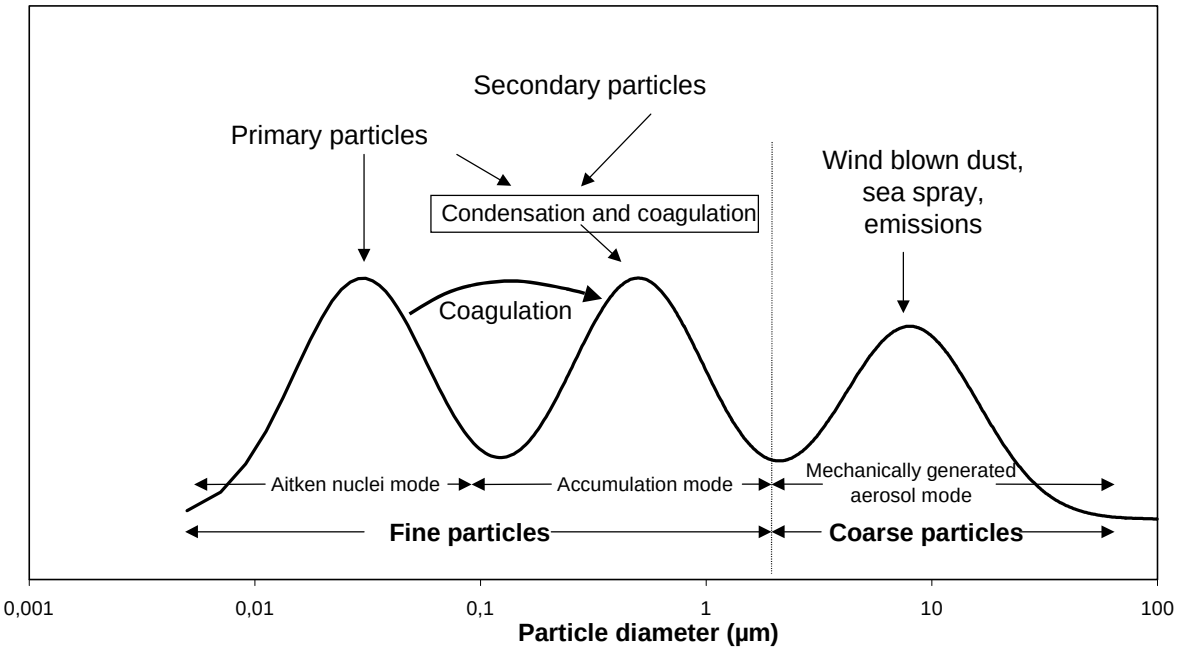


Figure 3. Schematic illustration of the size distribution of particle mass and the principal sources of different size ranges.

The urban aerosol consists of a complex mixture of both primary and secondary organic and inorganic compounds. Suspended particles are found in different sub-ranges due to their release, formation and atmospheric transformation. The

particles in the nuclei-mode (<0.08 µm) are emitted directly from combustion sources. The lifetime of these particles in polluted areas is typically less than an hour. They coagulate with larger particles or act as nuclei for cloud or fog droplets. The particles in the accumulation mode originate from coagulation of smaller particles, primary emission or from gas to particle transformation processes. In the coarse mode the particles are larger than about 2.5 µm and this mode includes particles from wind blown (or traffic generated) dust (sand/salt, wear from tires, brakes, road etc.) and sea salt. Some pollen and spores are also included in this size range.

Different atmospheric processes lead to formation of secondary inorganic and organic compounds that end up in particulate material. Atmospheric transformations include oxidation of SO_2 to sulphate, NO_x to nitrate and the dissolution of ammonia in cloud or fog droplets to form ammonium. Atmospheric oxidation of reactive organic compounds may lead to formation of a secondary organic aerosol (Odum et al., 1997). The chemical and physical processes involved in gas/particle transformations depend on many meteorological and chemical variables and are not completely understood.

Finally, it should be noted that long-range transport of particulate matter is an important source of particulate matter in Scandinavia.

3. Emission database

The quality of the emission data is usually the key factor determining the accuracy of the dispersion calculations in urban areas. This section gives a brief description of the emission database (EDB) used for the air quality simulations. The information in the emission database is provided by representatives from municipalities and regional authorities in the counties of Stockholm and Uppsala. The database is administered at the Stockholm Air Quality and Noise Analysis of the Environment and Health Protection Administration in Stockholm. The information is updated once a year.

3.1 Road transport

Road transport is one of the most important sources of air pollutants in the Stockholm region. The estimates of total traffic volumes are primarily based on measurements *in situ*. Such measurements are of different kinds: regular automatic traffic counting by the local traffic and street authorities within municipalities, automatic traffic counting on main roads by the Swedish National Road Administration and manual surveys of traffic volumes. Traffic volumes on major roads are taken from information provided by the Swedish National Road Administration. For other roads information in the database relies on the traffic volumes estimated from the measurements administered by the local municipalities.

Table 1. Information used to calculate emissions from road traffic in the emission database.

Static information			**Dynamic information**
Links[1)]	**Road types**[3)]	**Vehicles**[4)]	
Road type Total traffic volume Sign-posted traffic speed Percent heavy traffic Number of lanes Number of stops Correction factor[2)]	The relative composition of different vehicle types	Heavy duty vehicles Heavy duty vehicles with trailor Light duty vehicles Emission factors for each vehicle at different speeds	Daily variation of traffic flow of different vehicle types and road types Monthly variation of traffic flow of different vehicle types and road types Future scenarios where the relative composition of different vehicle types changes

[1)] There are about 4500 links in the emission database for the province of Stockholm.
[2)] A correction for the slope of the link. Usually not used.
3) There are about 45 different road types in the province of Stockholm.
4) There are nine different vehicle types (see text).

All information on the traffic volumes is described in the emission database as line sources, digitised manually on maps by representatives from the different

municipalities. The geographic accuracy obtained is a few meters. The traffic network is divided into short links for which traffic volume, speed, vehicle composition etc. are defined. For each link there is also information on the traffic flow described by the number of stops per kilometre on the link. Within the county of Stockholm there are about 4000 to 5 000 links. The length of each link varies from a few tens of metres to a few kilometres.

Each link is connected to a road type. About 45 different road types are being used. The road types represent different geographical locations, different vehicle compositions and/or different temporal variation of the traffic volumes. All data are given as annual averages. Information on the temporal variation is based on local measurements or estimates provided by regional authorities.

3.1.1 Vehicle emissions

The emission database contains three main types of vehicles: light-duty vehicles (passenger cars), heavy-duty vehicles (trucks) with and without trailor. For each category there are three sub-classes that have different emissions depending on the vehicle age and exhaust after-treatment. Buses are included among the heavy duty trucks.

Emission factors for the vehicles are based on the EVA-model (Hammarström and Karlsson, 1991). The highest specific emission (i.e. per vehicle-km) of both NOx and fine particulate matter comes from heavy-duty diesel fuelled vehicles and the lowest from light-duty vehicles with catalytic converters.

Table 2. Vehicle types in the emission database.

Type of vehicle	**Vehicle subset**	
Passenger car	1	Without catalytic converter; corresponds to the vehicle composition of 1986
	2	Catalytic converter (old type), environmental class 3
	3	Catalytic converter, environmental class 1
Truck	1	Corresponds to the vehicle composition of 1986
	2	Environmental class 3
	3	Environmental class 1
Truck with trailer	1	Corresponds to the vehicle composition of 1986
	2	Environmental class 3
	3	Environmental class 1

Table 3 shows the mean emission factors for particulate matter and NO_x for different types of vehicles in Stockholm. Note that the values represent the mean emission factors for different driving conditions in the city of Stockholm. Since the number of diesel driven passenger cars is very small compared to petrol driven cars they have not been defined separately in the EDB. But for PM the specific emission factors are about 10 times higher than petrol driven cars without catalysts. For the city of Stockholm, diesel passenger cars are estimated to contribute with 5% of the total traffic volume by passenger cars. This means that

diesel driven passenger cars contribute with about 30% of the total exhaust emission from road traffic in the city of Stockholm.

Both for particulate matter and NO_x the specific emission factors for heavy-duty vehicles are much larger compared to those for cars. For PM the emission factor for heavy-duty trucks (average of all trucks) is about 30 times higher than the corresponding factor for cars (average of all types of cars). For NO_x the corresponding emission factor is about 26 times higher for trucks than for cars.

For the city of Stockholm the exhaust emission of PM from trucks is about 60% of the total exhaust emission from all vehicles. For NO_x about 40% of the total exhaust vehicle emission comes from trucks.

Most of the exhaust particulate matter from diesel and gasoline vehicles is less than 2.5 µm (70% to 95% of the total mass) and almost all is less than 10 µm (EPA, 1985).

Table 3. Emission factors for trucks and cars in the city of Stockholm. Note that the values are weighted averages for the traffic composition and driving conditions in the city of Stockholm.

	Emission factor (g per vehicle km)	
Vehicle type	**Particulate matter**	**NO_x**
Passenger car[2)] without catalyst (corresponding to vehicle composition 1986)	0.019	2.4
Passenger car[2)] with old catalyst (1990 model car)	0.0054	0.3
Passenger car[2)] with new catalyst (1997 model car)	0.0053	0.2
Truck[1)], old type	0.27	7.4
Truck[1)], Environmental class 3	0.22	4.7
Truck[1)], Environmental class 1	0.071	3.6
Truck w trailor[1)], old type	0.60	24
Truck w trailor[1)], Environmental class 3	0.26	15
Truck w trailor[1)], Environmental class 1	0.13	12

[1)] >3.5 and < 24 tons gross vehicle weight.

[2)] Gasoline driven cars only. For diesel driven cars see text.

3.1.2 Cold start emissions

When vehicles are first started it takes some time before the engine reaches its normal operating temperature. During this period the emissions are substantially higher than when the engine is hot. The problem increases the colder it is, though it can be reduced using an engine pre-heater (Björklund, 1996). This is true both for petrol-engine vehicles without catalysts and those with three-way catalytic converters. Catalytic converters do not operate effectively until they have reached their normal operating temperature. During driving in heavily congested traffic in cold weather the vehicles may not reach their full operating temperature throughout their journey (Hutchinson and Clewley, 1996).

The additional emissions due to cold starts in the model are large for small roads (a journey of about 10-20 km for each cold start) and almost zero for motorways (a journey of about 300 km for each cold start). Cold start emissions are implemented as line sources distributed along different road types.

3.1.3 Re-suspension of road dust

Particles on the roads will be re-suspended mainly due to the turbulence generated by the vehicles. When the roadways are dry, this re-suspension is important and may be 10 times larger than the direct emission from the exhaust pipe (Johansson, 1993). Measurements during wintertime in Oslo have shown that the contribution from re-suspension was about 3 g/vehicle-km, when the roadway was dry.

The origin and nature of street dust was investigated in Madrid and Oslo between 1990 and 1994 (de Miguel et al, 1997). Two distinct ”sources” could be identified:

‘Urban’ elements include exhaust and wear of tyres and breaks plus weathering and corrosion of building material.

‘Natural’ elements include sanding, salting, transport onto the road by the wind and by tyres.

Road dust may come from mechanical wear of tyres and brake lining of the vehicles, debris from vehicle loads, influx of soil material, wear of the road surface (especially due to the use of studded tyres) and sanding/salting of roads. The wear of the road surface increases with the moisture and is 2-6 times larger for a wet road than for a dry one (Folkeson, 1992). It also increases after salting of the road, since then the road surface remains wet for longer periods. Vehicle speed, the power on the shaft, the tyre pressure and air temperature also affect road wear. As the temperature decreases the tyres become less elastic, with the result that the roadway gets more worn (NTNU, 1997). The size of many of these particles is larger than 10 μm, but the contribution to the PM_{10} levels in air may be substantial.

Recently, Westerlund (1998) studied the chemical content of brake lining and estimated the production of metals in the urban environment due to this wear. The amount and type of brake wear consumed by different types of vehicles was estimated based on a large inquiry for the city of Stockholm (1997). Based on this information the emission factor for passenger cars is estimated to be 10 to 20 mg per vehicle-kilometre, which is the same magnitude as the emission factor for exhaust emission of particles from passenger cars without catalysts in the city of Stockholm. It is somewhat higher than the estimate by US EPA of about 8 mg/vkm. Buses and trucks have been estimated to emit 80 to 110 mg/vkm in Stockholm. This is between 30% and 50% of the exhaust emission from these vehicles. The brake wear emissions include all sizes of particles but according to US EPA 98% of the total particle mass emitted is less than 10 μm and 39% is less than 2.5 μm (EPA, 1985). Since we are not aware of any studies for Swedish conditions and it is still not clear how large fraction that will actually become airborne, brake wear particles is not included in our calculations presented here.

Table 4. Comparison of emissions of particulate matter due to wear of brake lining and vehicle exhaust for the city of Stockholm.

Vehicle type	Wear of brake lining (mg/vkm)	Exhaust emission (mg/vkm)
Passenger car without catalyst	10 — 20	Ca 20
Passenger car with catalyst	10 — 20	5 — 6
Truck	80 — 90	200 — 300
Bus	110	200 — 300

The re-suspension due to the turbulence generated by the vehicles without any influence from meteorological factors is calculated as:

$$E_{resusp}^{vehicle} = \frac{\%cars}{100} \cdot 6.7 \cdot \left(\frac{v}{110}\right)^2 + \frac{\%trucks\ /\ buses}{100} \cdot 6.7 \cdot \left(\frac{v}{110}\right)^{0.5} \quad (1)$$

In this expression re-suspension for cars is proportional to the square of the vehicle speed (v in km/h) and for trucks to the square root of the speed (Sehmel, 1984). At 110 km/h both cars and trucks emit 6.7 mg/vkm. The emissions due to re-suspension, with consideration of the influence of meteorology are then calculated as (Bringfelt et al., 1997):

$$E_{resusp} = E_{resusp}^{vehicle} \cdot f_{qe} \quad (2)$$

where f_{qe} is a function that reduces the re-suspension depending on the conditions of the road surface, meteorological conditions and dust depot. The wetness of the road is calculated from hourly data on precipitation, temperature, dew point, net radiation and wind speed. The model has been tested using data from two cities in Sweden (Gothenburg and Norrköping) and a complete description is given by Bringfelt et al. (1997).

Most of the total re-suspended matter consists of large particles with high settling velocity that are deposited on the roadway or within about ten metres (Folkeson, 1976; Lygren & Gjessing, 1984; Lygren et al, 1984). Only a small fraction of the particles are transported farther away from the road (NVF, 1992). Data reported by US EPA suggest that of the total suspended particulate matter from road dust on the order of 20% is PM_{10} and 10% $PM_{2.5}$ (EPA, 1985). These percentages will depend on a number of factors (road surface material, sand/salt load, type of tyres, amount and composition of the traffic etc.). Particle size, topography, wind speed, road design and vegetation density are important for how the particles will be dispersed.

3.2 Emissions from ships

In the EDB emissions from ships are divided into emissions from pleasure boats, the ferries, merchant ships and other ships (cargo ships, sightseeing boats,

ships belonging to the marine and customs, ice-breakers etc.). The emissions from ferries and merchant ships are described as a series of point sources dotted along the major passages and a special point source with different temporal variation in the harbours. The timetable for the ferries to Finland and the Baltic States are used for the temporal variation of the emissions from ferries. The merchant ships are larger than 300 gross weight tons, except ferries. The emission rates for NO_x and PM are based on Skogö (1992).

The emissions from pleasure boats are calculated on the basis of boat and motor types and the sizes of the engines according to SNV (1990). The number of boats is taken from the now discontinued registry of pleasure boats. The emissions are distributed along the coastlines of the municipalities and are assumed to occur during daytime in the summer months.

Figure 4 shows the relative contributions from the different types of ships to the total emissions in the county of Stockholm. For NO_x almost 90% of the emission comes from the ferries, whereas for particulate matter a substantial fraction (65%) is emitted from pleasure boats.

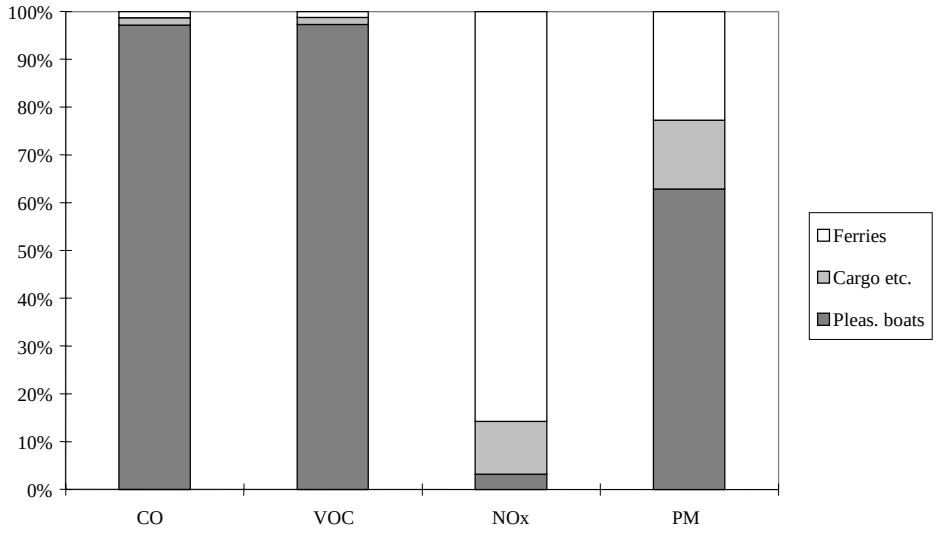


Figure 4. Emissions of carbon monoxide, volatile hydrocarbons, nitrogen oxides and particulate matter in the county of Stockholm 1995.

Most of the pleasure boats use two-stroke engines, which do not combust more than 75% of the fuel (Burman and Johansson, 1997). Sailing boats usually use diesel engines. They emit about 5% of the total particle emissions from pleasure boats, while two-stroke engines emit about 85%. Since the emissions take place below water surface, there might be some scavenging in the water, which means less emission to the air.

3.3 Motorised tools and off-road vehicles

Emissions from motorised tools, such as air compressors, snowmobiles, power-saws and lawn-movers are not regulated by law. Working machinery include both vehicles and equipment, such as excavators, used for earth moving, building and

civil engineering construction. These diesel engines are often running during long periods. Therefore, they may emit significant amounts of particles and NO_x, for example at road and house construction sites (Björklund, 1996). Since the geographic positions of these vary during the year, especially for the ones used at road-works, they are not included in our dispersion calculations.

The emissions from off-road vehicles and motorised tools in the municipalities in the county of Stockholm are estimated based on national data according to SNV (1990) and SNV (1993), respectively. Since there is no information on the geographical distribution of the emissions, 19.2% of the total national emission was attributed to the county of Stockholm, i. e. the same share as the population. The emissions are described as area sources — 24% attributed to industrial areas and 76% according to population statistics of the different municipalities. There is a large uncertainty in the actual emissions. This source is particularly problematic in dispersion modelling on local scales, since it is very difficult to define the geographic distribution and the temporal variation. It is not included in the model calculations presented here due to lack of information (see further discussion below).

3.4 Residential heating

3.4.1 Wood combustion

Emissions from wood combustion for residential heating depend on the type of boiler and how the wood is combusted. Five to six percent (5-6%) of the boilers in Sweden are environmentally authorized (NUTEK, Naturvårdsverket, 1993) and 39% are connected to an accumulation tank. In the emission database for the counties of Stockholm and Uppsala, the emissions are allocated to residential areas that are not connected to central district heating (Ekström et al, 1995). The extent of wood combustion in each municipality was estimated by STOSEB, 1992 (an association between energy production companies in the county of Stockholm).

The emissions of particulate matter from small boilers vary from 200 to 1500 mg/MJ (Kyrklund et al, 1998). The difference between combustion using accumulator (40 mg/MJ) and direct combustion (900 mg/MJ) depends on the combustion temperature. The temperature in direct combustion is too low to burn the wood completely, while accumulated combustion can occur at a higher temperature. Only 42% of the newly installed boilers are in accordance with environmental recommendations. The emission factor for particulate matter used in the emission database of Stockholm is 900 mg/MJ. The uncertainty may be as large as a factor of 10.

There is also a large uncertainty in the total use of wood as fuel for small-scale heating. Although the amount of wood-heated houses is relatively small in urban areas, they may account for as much as 25% of the total wood consumption in Sweden. The total use of wood for domestic heating in Sweden has been estimated to 12 TWh or 2.1 - 2.8 Mtons per year in 1994, but it may be 3 to 7 times higher (Kyrklund et al., 1998).

3.4.2 Fuel oil for residential heating

The emissions from houses heated with oil boilers are described as area sources allocated to residential areas (Ekström et al, 1995). The emissions are assumed to be 50 - 100 mg/MJ (depending on the oil quality) for NO_x and 20 mg/MJ for particulate matter, respectively. The amount of light fuel oil is obtained from Statistics Sweden (SCB).

3.5 Large combustion sources

The largest energy production plants require special authorisation for operation and have to provide a report on the emissions every year, either to the environmental agencies of the municipalities or to the regional authorities. The emissions are usually calculated from in situ measurements. This information is included in the emission database. For energy production plants larger than 10 MW, actual monthly fuel consumption during the year is input to the emission database.

For smaller central-heating plants (< 10 MW) serving large groups of buildings or hospitals, information is collected about the total fuel consumption and type of fuel during the year. Emissions are then calculated using standard emission factors for different types of fuel. The temporal variation of the emissions (daily and monthly) is either specifically obtained from the individual power plant or otherwise given as standard formulas. Information on the smaller central-heating plants is obtained from representatives of the environmental agencies in the municipalities.

3.6 Construction dust

Construction and maintenance work, including new building construction, demolition and redevelopment, major building refurbishment and routine maintenance is another source of dust. Even quite modest operations, such as rubbing down paint prior to redecoration, produces considerable quantities of fine dust. Dust is also produced as a result of highway maintenance and other civil engineering works (Hutchinson and Clewley, 1996).

Street dust samples collected in the proximity of buildings being renovated show high concentrations of Ca, suggesting that cement dust is a major component of street dust in these locations (de Miguel et al, 1997). According to de Miguel et al. (1997), paint flakes from façades are likely responsible for the unusually high lead content, while corrosion of metal structures can explain the anomaly for some of the other trace elements.

The majority of the construction dust concists of particulate matter larger than 10 µm in size. Given the difficulty of estimating the emissions and the fact that the geographic positions of the emissions vary, they are not included in the dispersion calculations.

3.7 Air traffic

In the emission database, air traffic is described by a variation during the day, the week and the year. Only the airports of Arlanda and Bromma are included

(Ekström et al, 1995). The LTO-phase emissions are included as area sources placed at the ground surface. This will tend to overestimate the contribution from aircrafts to the concentration, since a large fraction of the emission occurs higher up in the atmosphere.

Other important sources connected to airports include:

- Ground support vehicles (aircraft service vehicles, baggage handling vehicles etc.)
- Road traffic serving the airport (passenger transports)
- Airport heating systems

All these sources have been included as area sources in the emission database.

The discharge of particulate matter from aeroplanes consists of two kinds of very small particles (much smaller than 2,5μm), soot and engine wear (Ström, 1998). Emissions of particulate matter from aeroplane engines have been studied by Petzold and Schröder (1996). The measurements were performed in the exhausts of a Rolls Royce/SNECMA M45H Mk501 engine, which emits about five times more particles than most modern engines. The emissions varied only by a factor less than two at between 11% and 70% of maximum power, which indicates that the variations of the emissions during a flight, except take-off, are relatively small. According to Ström (1998), the emission (g total coal/kg fuel) is at least ten times higher for take-off than for the rest of the flight. The total fuel consumption for the LTO-phase (landing and take-off) at Arlanda and Bromma has been calculated by Anette Pålsson at FFA (The aeronautical research institute of Sweden).

Information on the emissions of NO_x from Arlanda and Stockholm is obtained from the Swedish Board of Civil Aviation. The calculations are carried out according to regulations set by the Swedish National Franchise Board for Environmental Protection. Data for the different aircrafts are collected from the airplane companies at Arlanda Airport and from the US Federal Aviation Administration's Aircraft Engine Emission Database. The aircrafts are assumed to be equipped with the most common engine for each type of aircraft. Unknown aircraft types are assumed to have the same emissions as similar known models.

3.8 Total emissions of NO_x and particulates in Stockholm

The total emissions of particulate matter (< 10 µm diameter) and NO_x from different sources in 1996 are presented in Table 5. For particulate matter the dominating source is small-scale wood burning. The estimate is, however, very uncertain due to lack of information on emission factors, large variation in the emissions from different types of boilers depending on burning conditions and also uncertainties regarding the amount of wood burned. For particulate matter from road traffic, about equal amounts are emitted directly as exhaust gases and indirectly as re-suspended road dust. The latter emission is estimated from calculated concentrations using the re-suspension model and is not included in the table.

For NO_x, road traffic is the main source, contributing with almost 40% of the total emissions both in the city and in the county of Stockholm. As stated earlier a very large fraction of the vehicle exhaust emissions of NO_x and PM come from heavy-duty vehicles. For NO_x light-duty vehicles without catalysts contribute with about 40% in the city of Stockholm. Another important source of both NO_x and PM is off-road vehicles. They contribute with more than 30% of the total NO_x emissions in the city of Stockholm and almost 20% of the total PM_{10} emissions (excluding re-suspension).

The geographical distribution of the total emissions in Greater Stockholm is presented on the maps below.

Table 5. Total emissions of NO_x and particulate matter (PM_{10}) in Stockholm (1996).

Source	County of Stockholm				City of Stockholm			
	PM_{10}		NO_x		PM_{10}		NO_x	
	Ton/year	*%*	Ton/year	*%*	Ton/year	*%*	Ton/year	*%*
Road traffic	220[3]	*7*	13 000	*37*	90[4]	*10*	3 900	*36*
Ferries	190	*6*	8 000	*23*	30	*3*	900	*8*
Energy production	2 400[1]	*72*	4 400	*13*	630[2]	*68*	2 300	*21*
Industrial processes	80	*2*			20	*2*		
Off-road vehicles and motorised tools	400	*12*	9 000	*26*	150	*16*	3 600	*34*
Air traffic	30	*1*	700	*2*	1	*0*	-	*0*
Total	≈ 3 300	*100*	≈ 35 000	*100*	≈ 920	*100*	≈ 11 000	*100*

[1] Wood burning is estimated to emit 1 900 ton/year but the uncertainty may be as high as a factor of 10.
[2] Wood burning is estimated to emit 320 ton/year.
[3] This value does not include brake wear particles and re-suspension of road dust. The latter may be of about equal magnitude (about 200 ton/year), see under Results below.
[4] This value does not include brake wear particles and re-suspension of road dust.

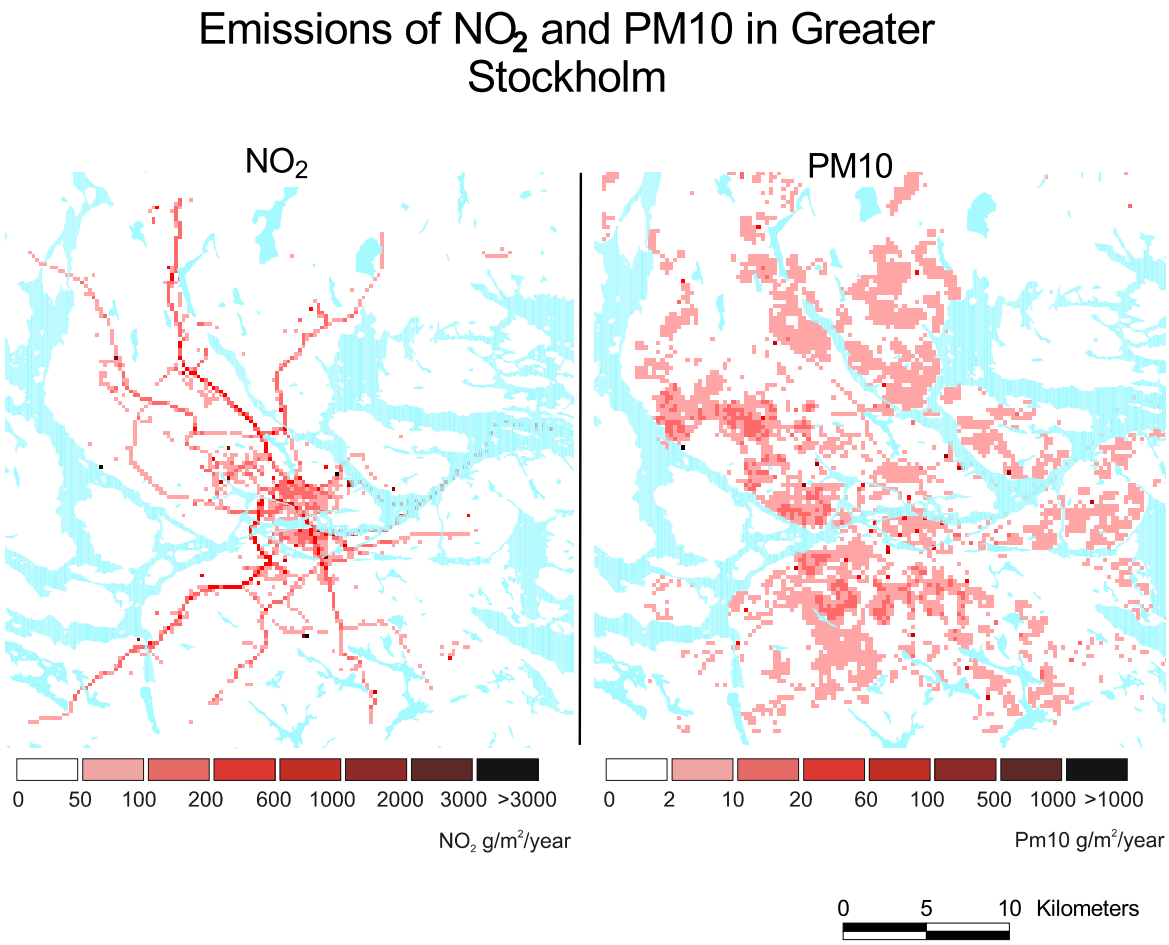


Figure 2. Total emissions of NO2 and PM10 in Greater Stockholm during 1996. Emission of PM10 due to resuspension is not included.

4. Validation of emission data

Since emission data represent the main uncertainty in air quality model calculations on the urban scale, it is very important to validate the emissions calculated from the information in the database. For the purpose of modelling it is not only important to accurately quantify the sources but also the temporal and spatial distributions of the emissions. As already indicated above, some sources such as off-road vehicles are extremely difficult to localise, since e. g. construction and maintenance work is not stationary. The problem of validation of emission data have recently been addressed by the European Topic centre on Air Emissions of the European Environment Agency (Cirillo et al., 1996).

For Stockholm different cross checks have been made in order to evaluate the uncertainty in the emission data:

- Calculated vehicle fuel consumption has been compared with fuel deliveries to the region.
- Fuel oil consumption for energy production has been compared with deliveries to the region.

As can be seen in Table 6 there is, in general, an excellent agreement between estimated fuel consumption from the emission database and the deliveries of fuels to the county of Stockholm according to Statistics Sweden (SCB). The total amount of gasoline *delivered* to the region was 890 000 m^3 during 1996. The amount of gasoline *sold* at gasoline stations (reported from each individual station) was 856 000 m^3 according to the emission database. Based on the traffic volume in the county of Stockholm and fuel consumption factors for different vehicle types at different driving conditions, the total vehicle gasoline consumption is estimated to 734 000 m^3. Gasoline is also used in pleasure boats, working tools, motorcycles and mopeds. Altogether, approximately 800 000 m^3 gasoline is estimated to be consumed in the region. This is in reasonable agreement with the amount sold.

The corresponding amount of diesel consumption, based on traffic volumes and diesel fuel consumption of mainly heavy duty vehicles (excluding off-road machinery), is about 200 000 m^3. This agrees well with the diesel delivered for road transport, 208 000 m^3 according to SCB, indicating that the heavy duty traffic volume data in the EDB-96 is of good quality.

The total amount of fuel oil delivered was 1 275 000 m^3 in 1996. This may be compared with the corresponding value used to calculate emissions due to burning of oil, which is 1 360 000 m^3 in EDB-96. However, some of the information on fuel consumption in the EDB is based on data from SCB (mainly residential heating), so the values in this comparison are not completely independent.

The amount of diesel used by working machinery is probably the single largest uncertainty in the EDB. There are no statistics available that would make it possible to validate the emissions from working machinery and the uncertainty may be several tens of percent. It should be noted, though, that the impact of these emissions on the ambient concentrations will only be of local importance and not so important for larger areas. For dispersion calculations the geographic distribution of the emissions is critical and since such information is not available, working machinery is not included in the calculations presented here.

Table 6. Comparison of fuel deliveries according to the national statistical office in Sweden (Statistics Sweden, SCB) and the consumption according to the emission database for 1996 (EDB-96). All numbers are for the county of Stockholm. Unit: m^3.

	Fuel deliveries 1996 According to SCB	**Fuel consumption According to the EDB-1996**
Gasoline, total	890 000	856 000 (sold amount[2])
Light duty vehicles		734 000[1]
Pleasure boats		33 000
Working tools		1 750
Motorcycles and mopeds		not available
Diesel, total	296 000	—
Road transport	208 000	205 000
Fuel oil, total	1 275 000	1 360 000
Grade 1 (EO 1)	725 000	—
Grade 2-5 (EO 2-5)	550 000	—

[1] Based on fuel consumption factors (litre gasoline per vehicle km) and traffic volumes (vehicle kilometres) in the emission database.

[2] Amount sold at gasoline stations in the region. This value is used to calculate the emissions of volatile hydrocarbons from gasoline stations.

Another cross check of the emissions of NO_x and PM may be obtained by using recent data on the specific emissions from heavy duty vehicles (NTM, 1998). According to these data 25 - 35 g NO_x is emitted per liter diesel. Assuming that 182 000 liters of diesel is used by heavy duty vehicles (based on EDB-96) gives 4 500 – 6 500 tons of NO_x from heavy duty vehicles in the county of Stockholm. This may be compared with 3 500 tons NO_x according to EDB-96, based on traffic volumes, driving conditions and emission factors according to the National Road and Traffic Authority.

A corresponding comparison may be made for particulate matter. The specific emission for heavy duty vehicles is 0.5 - 1 g PM per liter diesel (NTM, 1998). Using the total amount of diesel used by heavy duty vehicles (182 000 m^3), this gives a total emission of about 90 - 180 tons of PM for the county. This is in reasonable agreement with 140 tons PM according to EDB-96, based on traffic volumes, driving conditions and emission factors according to the National Road and Traffic Authority.

In most cases emissions from road traffic are calculated from emission factors determined for single vehicles in dynamometer tests under certain driving conditions specified in standardised driving cycles. In order to determine the accuracy of the estimated emissions from road traffic, measurements under real-world driving conditions are needed. Vehicle emission factors used in the emission data base for Stockholm have been compared with measured emission factors for

NO_x, CO and some volatile organic compounds based on traffic tunnel studies (Johansson et al., 1996; Johansson et al., 1997). A comparison has been made of the emission factors for different types of vehicles used in the emission database with factors calculated from measurements in a road traffic tunnel. It showed a discrepancy of up to 50% for NO_x. Although, this comparison is only valid for the type of driving that prevail in the tunnel, it indicates that even though the estimates of traffic volumes and fuel consumption are of good quality there may be uncertainties in the specific emission factors that should be considered.

No validations of the emissions of particulate matter have been made. Another way to assess the accuracy of the emission database is to compare with results from source-receptor models. This is particularly useful for particulate matter and such a project is currently (1998) being implemented in Stockholm.

5. The dispersion model

A Gaussian dispersion model is used to simulate the distribution of ground concentrations over the area. Hourly concentrations are calculated based on a wind field obtained from a diagnostic wind model. The wind is assumed to be more or less constant during that period and the concentrations are assumed to be in steady-state.

The concentration due to the emission from a single source, Q, is calculated as (SMHI, 1997):

$$C = \frac{Q}{2\pi\sigma_y\sigma_z U} e^{-y^2/2\sigma_y^2} \left[e^{-(z-h_e)^2/2\sigma_z^2} + e^{-(z+h_e)^2/2\sigma_z^2} + e^{-(z+h_e-2h)^2/2\sigma_z^2} \right]$$

where Q describes the source emission, y and z are the distances to the sources in the horizontal and vertical direction, respectively. U is the horizontal wind speed (along the x co-ordinate). The dispersion parameters, σ_y and σ_z, are calculated on the basis of meteorological measurements, extrapolated using a wind model (see below). The resolution of the wind field depends on the topographic information, which range from 500 metres to 2 km. If the map is zoomed up to a smaller area, the wind field will be linearly interpolated within the original grid.

The model does not resolve individual buildings. Instead, surface structures like houses and trees enter the model through local roughness values and through the wind field. In an open area the calculation height is 2 m above ground level. Over a city the simulation will reflect the concentrations at 2 meters above roof height. A special treatment of the Gauss model plume length is introduced to avoid unrealistically long plumes. This length depends on the stability and persistency of weather conditions. A detailed description of the model is given in the Airviro User Documentation (SMHI, 1993).

5.1 Photo-chemistry and other processes affecting the NO_2 and PM_{10} levels in the city

5.1.1 Calculation of NO_2 concentrations

The model calculations do not explicitly consider chemical reactions. Most of the NO_x (sum of NO and NO_2) is emitted as NO, nitrogen monoxide. NO is rapidly oxidised in the atmosphere by reaction with ozone (O_3) to NO_2 (nitrogen dioxide). The relative amounts of NO and NO_2 therefore depend on the ozone level and are also dependent on incoming solar radiation and air temperature. Close to the traffic inside a street canyon, NO may be the dominating form of NO_x. At roof level, NO_2 concentrations are generally higher than NO.

The dispersion model is used to calculate NO_x concentrations (mean and 98-percentile concentrations of hourly mean values). The removal of NO_x from the atmosphere occurs as a result of mainly two processes: NO_2 is oxidised to nitric acid or removed by dry deposition on ground surfaces. However, both processes are

slow giving a lifetime of NO_x of a few days. Since this is much longer than the time scale for dispersion in this region, both processes have been neglected. Annual mean and daily 98-percentile concentrations of NO_2 is obtained from an exponential expression[3] based on measured NO_x levels and NO_2 at different locations. Both urban street sites and background sites have been used in order to obtain an expression that can be used for any geographical area in the region. The daily 98-percentile concentrations are obtained from a linear relation between the hourly mean 98-percentile concentrations and the daily mean 98-percentile concentrations.

The left panel of Figure 6 shows a comparison of the measured annual mean NO_2 concentrations at three sites in central Stockholm with that calculated from measured NO_x concentrations according to the empirical expression. The right panel of Figure 6 shows a comparison of the measured daily 98-percentile NO_2 concentrations with calculated hourly 98-percentile concentrations. In both cases the difference between calculated and measured data are less 10%.

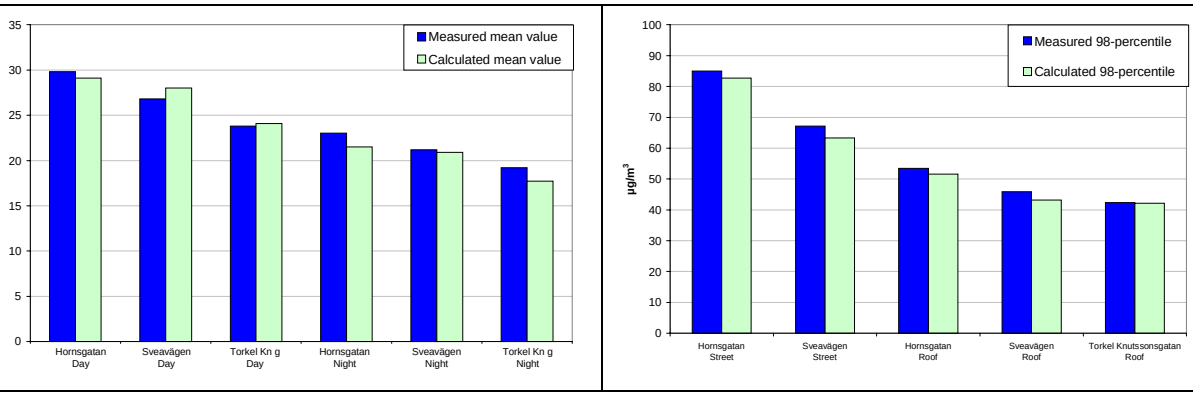


Figure 6. Comparison between measured annual mean and daily 98-percentile concentrations. Data are from three measurement stations in the city of Stockholm in the period January 1993 to September 1998.

5.1.2 Calculation of PM_{10} concentrations

The modelling of particulate matter is complicated by the need to consider not only primary particulate matter (i. e. PM emitted in combustion processes etc.), but also secondary particulate matter formed through physical and chemical reactions. Under certain conditions nucleation is a source of new particles. Even though, a large number of ultra-fine particles can be generated in urban areas, their mass is sufficiently small that they do not significantly alter the total aerosol mass concentration on the time-scale for dispersion of air pollutants in urban areas (Wexler et al, 1994, and Lurman et al, 1997).

Condensation/evaporation of inorganic and organic compounds increases or decreases the particle mass and changes the size of the particles. Two main sources of condensation/evaporation are sulphur dioxide (SO_2) and nitric acid

$$[NO_2] = [NO_x]^{\left(A+\frac{B}{C+[NO_x]}\right)}$$

[3] where $[NO_2]$ and $[NO_x]$ are concentrations of NO_2 and NO_x.
A, B and C are empirical constants.

(HNO_3). Emitted SO_2 is oxidised to sulphate, which condense onto droplets and particles. Most particulate sulphate is found as a combination of sulphuric acid, ammonium bisulphite (NH_4SO_4), and ammonium sulphate ($(NH_4)_2SO_4$). The time-scale for gas-phase transformation of SO_2 to sulphate range from less than 0.01% (of gaseous SO_2) per hour to 5% per hour, depending on meteorological and photochemical conditions. The contribution of locally emitted SO_2 in Stockholm is assumed to be negligible for PM_{10} concentrations in this region.

Nitric acid, formed as a result of the oxidation of NO_2, may be absorbed on droplets and particles. Particulate nitrate (in the form of ammonium nitrate) may evaporate from the particles to the air, due to equilibrium between ammonium nitrate and gaseous nitric acid and ammonia. Nitric acid may also react with sodium chloride and alkaline dust particles forming other particulate nitrate species. The rate of gas-phase conversion of NO_2 to nitric acid ranges from less than 1% per hour to 90% per hour. Emitted NO_2 may thus, under favourable conditions, be converted to particulate nitrate and thereby contribute to the local PM_{10} levels in Stockholm. This is not considered in the model calculations. Measurements show that the levels of nitric acid and particulate nitrate are around 0.5 $\mu g/m^3$ higher in central Stockholm compared to rural (background) areas. Whether this is due to local formation of nitrate from NO_2 or primary emission is not known, but it gives an upper limit to the effect of local NO_2 to nitrate conversion.

There is also a contribution to the PM mass from organic compounds, either primary emitted or formed in photochemical processes (secondary organic compounds). Recent studies by Wexler et al (1994) and Lurmann et al. (1997) show that the contribution from secondary organic material to the fine particle mass in San Francisco is small compared to direct emission of organic material. Since it may safely be assumed that photochemical processes are less important in Stockholm compared to San Francisco, formation of secondary organic compounds should be even smaller in Stockholm. Primary semi-volatile organic compounds may also be adsorbed or absorbed on the aerosol particles, but since there is very limited information on the levels of these compounds in Stockholm this is not considered in our calculations.

Coagulation processes also affect the size-distribution of the aerosol. However, since wet and dry deposition of particles is not considered here, the size distribution is not taken into account. Coagulation does not affect the total mass of particulate matter. When haze or fog exists particles grow to larger sizes (Svenningsson et al., 1992). Particle growth and shrinkage are determined by the amount of water transferred to and from the aerosol. These processes depend on the relative humidity, temperature and chemical composition of the aerosol. Deposition of fog droplets may be large. In addition, fog reduces the photolytic reactions and increases the reaction rates above the fog layers (Lurmann et al, 1997). The importance of such processes for particle concentrations in Stockholm has not been assessed.

Finally, as shown later in this report, the calculations based on the local primary emissions and considering the import of particles to the region (based on measurements) yield a good agreement with local measurements of PM_{10}. This indicates that physical and chemical particle transformation processes, not considered in these calculations, are small.

6. Meteorological data and the wind model

Meteorological measurements at different sites within the region serve as input for the dispersion calculations. Two different types of data are used for NO_2 and PM_{10} calculations, climatological and sequential real-time data, respectively.

6.1 Climatological data for NO_2 calculations

For NO_2 a climatological database is used (statistical meteorological data). Input data to the database is almost 10 years of meteorological measurements at a 50 metre high mast at Högdalen in southern Stockholm. From these data subsets of (climatological) data are generated in order to describe typical weather conditions in the region for different time periods. The climatologies consist of weather cases representing different stabilities for different wind directions. For the SHAPE project separate climatologies for night and day have been created. The climatological database contain all necessary meteorological data, including wind speed and direction, stability, temperature and frequency of occurrence for this type of weather.

6.2 Real-time sequential data for calculations of PM_{10} levels

For PM_{10} it is not possible to use climatological data consisting of separate weather cases. Real-time sequential data need to be used for the re-suspension model, which contain parameters that are dependent on the history of the weather (e g number of preceding sanding and salting occasions in the city).

Real-time data are obtained from the same meteorological stations as climatological data and the processing of the data is the same except that the frequency of occurrence is not used since all cases are considered. This means that the calculations of the PM_{10} concentrations require much longer computer time and much more post-processing in order to obtain the average concentration fields.

6.3 The wind model

Since meteorological data are available only at a limited number of sites and representative only for the local conditions close to the measurements, a wind model is used to generate all necessary data for all grid cells of the calculation domain. The horizontal large-scale wind is assumed to be uniform in the model. The area-wide wind field is estimated assuming geostrophic balance. The model is not mass conservative and vertical circulation can not be described, since it is a one-layer model. The model can not describe the sea breeze. But it can diagnose it, if mast data detect it.

7. Calculation areas and time periods

Figure 7 below shows the geographical areas for which calculations have been performed. The resolution refers to the topographic resolution and the resolution obtained in the wind model. It should be noted that the emission data are given at a much higher resolution.

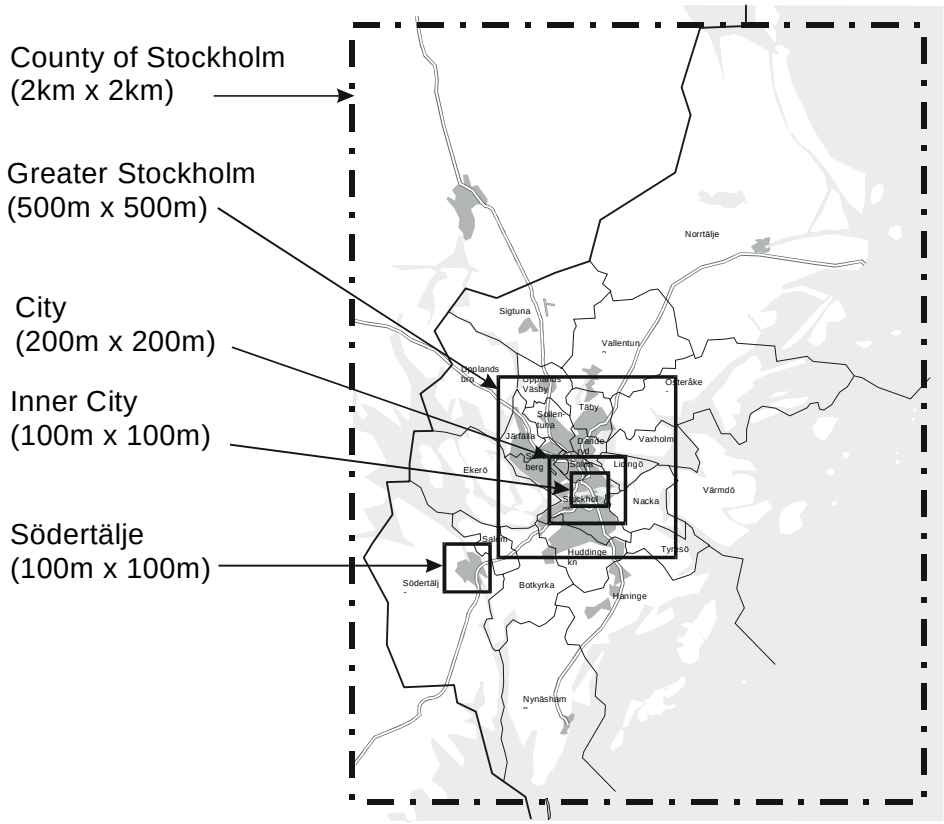


Figure 7. Calculation areas and resolution used.

Five different areas with different geographical resolutions were defined for the calculations:

1. County of Stockholm (298 000 m^2, 2 km x 2 km)
2. Greater Stockholm (71 000 m^2, 500 m x 500 m)
3. City of Stockholm (28 000 m^2, 200 m x 200 m)
4. Inner city of Stockholm (14 000 m^2, 100 m x 100 m)
5. City of Södertälje (17 900 m^2, 100 m x 100 m)

Separate calculations were performed for each of the areas. There was no nesting of the calculations. Since only local sources inside of the area were considered for each calculation different background[4] values was needed to obtain

[4] "Background concentration" is the contribution to the concentration from sources outside the area considered in the calculations (both local and distant sources may be important).

the total concentration in the respective areas. This is mainly important for the "city" and "inner city" area. For "Greater Stockholm", "County of Stockholm" and "Södertälje" there was essentially the same background concentration, which was obtained from rural monitoring stations. For Stockholm city and the inner city of Stockholm separate local background values was obtained from calculations using all sources except the ones within respective area. The background concentrations were then added to the concentrations calculated for each area. In this way the geographical variation of the background concentrations due to local sources was correctly taken into account. This procedure is, however, expected to overestimate the 98-percentile values.

Both annual averages and 98-percentiles of hourly and daily mean values have been calculated using emissions and meteorological data for daytime (07 – 19) and night-time (20 – 06) separately. In this way, the calculations use meteorological data that correspond to daytime and night-time population. Calculated concentrations for night-time population include also emissions and meteorology for daytime weekends and holidays. It should be noted that the 98-percentile of the daily mean values presented are not true daily mean values since only 12 hours (night or day) instead of 24 hours were considered in the values.

All model calculations for NO_2 are summarised in Table 7. Separate calculations have been performed in order to evaluate the contribution from road traffic to the NO_2 concentrations.

Table 7. Overview of calculations made for NO_2.

Value	Sources	Time resolution	Time period[1)]	Calculation areas
Mean and 98-percentile	All sources and road traffic separately	Hour and day	Yearly value day and night	All areas shown in Figure 7

[1)] Representative weather types from 8 years of meteorological data (climatological data, see text).

Calculations of annual concentrations of PM_{10} have been made for the same areas as for NO_2 except for the inner city of Stockholm, for which no calculations have been made. Meteorological data were from the period April 1995 to March 1996. Separate calculations have made for road traffic (sum of vehicle exhaust emissions and road dust).

Table 8. Overview of calculations made for PM_{10}.

Value	Sources	Time resolution	Time period[1)]	Calculation areas
Mean and 98-percentile	All sources and road traffic separately	Hour Day	Annual value day and night (April 1995 to March 1996)	All areas shown in Figure 7

[1)] Meteorological data from April 1995 to March 1996 (hourly mean values).

8. Population data

Data on the stationary population for 1995 have been obtained from the national office for Statistics, Sweden (SCB, Örebro). The data are separated between males and females and are also divided in 5 age classes of 20 years each (0-19, 20-39, 40-59, 60-79 and older than 79). In addition, there are separate data available for daytime and night-time population. Data have been provided for 100 m, 200 m, 500 m and 2000 m resolutions as defined in Figure 7.

8.1.1 Night-time population

The night-time population is based on the home addresses and is assumed to be representative also for the weekend population.

8.1.2 Daytime population

The daytime population for a certain area (grid) is defined as:

The night-time population in the area minus those living in the area but working somewhere else plus those working in the area but living somewhere else.

Statistics Sweden has data on employees according to the location of their place of work. Problems occur when a workplace is not connected to a property. Therefore, so called “NYKO”-areas are used. A NYKO-area varies in size in and between the municipalities but is usually between a few hundred metres (in densely populated parts of the municipality) to a couple of kilometres. The population is given for the central point in the NYKO-area. The error introduced in this way depends on the size of the NYKO-area. However, 94% of the population data are geographically distributed according to properties /addresses) and 6% according to NYKO-areas. About 10% of the daytime population in the county has not been possible to distribute in the gridnet. This value varies between municipalities and is between 8% (for Stockholm) and up to 20%-30%.

Only stationary population data is used since it is beyond the scope of the work presented in this report to differentiate more carefully between the time people spend in different environments. There are a number of uncertainties of using these data for exposure calculations:

1. Children staying at day-care centres and schools during daytime are not included in the daytime population
2. People at hospitals, shops etc. are not included correctly
3. Unemployed, people on leave, house-working people etc. are not included correctly

Another important factor to consider in exposure estimates is the differences between indoor and outdoor concentrations. This will be discussed later in the report.

Total population data for the different areas is summarised in Table 9. There are about 1.7 million people in the whole county. Almost 80%, or 1.3 million people, live in Greater Stockholm. Three percent (approximately 60 000 people) live in Södertälje.

It is interesting to note that there is a substantial difference between the daytime and night-time population in the City of Stockholm. During the day there are about 20% more people compared to night-time. For the Inner City area there are almost 50% more people during the day compared to night. For the other areas only small differences are noted.

The spatially averaged population density range between 33 individuals per square meter for the inner city of Stockholm during the day to 3.3 per square meter for the Södertälje area during the night.

Table 9. Summary of population and population density (number per square meter) data for the different areas considered.

	Inner city	City of Stockholm	Greater Stockholm	Södertälje	Whole county
Day					
Population	**465 104**	**768 240**	**1 357 705**	**62 235**	**1 682 371**
Density	**33**	**27**	**19**	**3.5**	**5.6**
Night					
Population	**313 281**	**625 826**	**1 337 237**	**59 939**	**1 725 341**
Density	**22**	**22**	**19**	**3.3**	**5.8**
Day/night ratio	**1.48**	**1.23**	**1.015**	**1.038**	**0.98**

9. Results

9.1 NO_2 concentrations

9.1.1 Impact of spatial resolution on calculated NO_2 concentrations

Before presenting the results it is interesting to consider the impact of spatial resolution on the calculated NO_2 concentrations. Obviously, a coarse resolution tends to smear out the variations more than a fine resolution, but there is a limit to the influence of the resolution. This may be illustrated by comparing the resulting NO_2 concentrations in the inner city of Stockholm using 2 km, 500 m, 200 m and 100 m resolution. The inner city of Stockholm is the area with the highest emissions and where the largest impact of the resolution is expected to occur. Using the same background value to compensate for sources outside of the inner city area, the range of the values differ due to the grid resolution.

The standard deviation of the daytime mean NO_2 concentrations of all grid cells in the inner city area decreases by 70%, from 6.1 to 1.8 µg/m^3. The maximum values decrease and the minimum values increase. The same pattern is seen for the 98-percentile of the 12-hour daytime concentrations. The impact of the resolution on the night-time concentrations is not so large due to lower variability in the emissions.

Table 10. Comparison of standard deviations, maximum and minimum values of calculated grid cell NO_2 concentrations within the inner city of Stockholm using different spatial grid resolutions (100 m to 2 km). Only daytime values.

		Mean, day	98-percentile, day
100 x 100 m	Standard deviation	6.1	6.1
	Min — Max	14 — 45	29 — 65
200 x 200 m	Standard deviation	5.5	6.2
	Min — Max	14 — 40	30 — 63
500 x 500 m	Standard deviation	4.1	4.3
	Min — Max	15 — 35	35 — 60
2 km x 2 km	Standard deviation	1.8	2.5
	Min — Max	20 — 32	43 — 60

For NO_2 concentrations at roof level an increase of the resolution beyond 100 x 100 metres is not expected to improve the description of the NO_2 pattern. This is mainly due to the fact that NO_2 is not primarily coming from direct emissions, but is formed as a result of the oxidation of NO by O_3.

It seems likely, though, that in order to achieve a more detailed description of the variation of NO_2 concentrations at finer spatial resolution than 100 metres, more sophisticated models would be needed, in order to take into account the inhomogeneities of the urban surface area (roughness, temperature, heat fluxes and humidity fluxes, which generate the development of internal boundary layers and local breezes). The buildings generate a thick roughness sub-layer occupying most of the atmospheric surface layer and a roughness sub-layer (just above the roofs), where both Monin-Obukhov theory and the homogeneity assumption are invalid. Canyon-like streets canalise the flow in the lowest layers of the dense city centres. Street canyon concentrations in Stockholm are discussed in "9.2 Population exposure to NO_2" below.

9.1.2 Calculated roof level concentrations

Calculated average roof-level concentrations of NO_2 in the county of Stockholm vary between 4 and 45 µg/m^3 during the day (Figure 8 and Table 11). During night-time they range from 4 to 37 µg/m^3. The minimum concentrations correspond to background concentrations due to sources outside the borders of the county. The highest concentrations occur in the central part of the city.

Table 11. Spatial averaged NO_2 concentrations in the county of Stockholm. All values correspond to one (climatological) year. Unit: µg NO_2/m^3.

Value	Time	Concentration interval Maximum and minimum value	Inner city of Stockholm (100 x 100 m) Ave ± Std.	City of Stockholm (200 x 200 m) Ave ± Std.	Greater Stockholm (500 x 500 m) Ave ± Std.	City of Södertälje (100 x100 m) Ave ± Std.	Whole county (2 x 2 km) Ave ± Std.
Mean	Day	45 — 4.1	27 ± 6.1	21 ± 6.3	12 ± 5.6	12 ± 4.9	5.1 ± 2.5
	Night	37 — 4.0	20 ± 4.8	16 ± 4.4	9.8 ± 3.7	8.7 ± 2.8	4.7 ± 1.7
98-perc (1-h mean)	Day	87 — 12	58 ± 8.1	51 ± 9.0	44 ± 11	35 ± 11	17 ± 7.2
	Night	83 — 12	51 ± 7.6	44 ± 8.4	39 ± 9.1	28 ± 9.3	16 ± + 6.3
98-perc (12-h mean)	Day	65 — 9.3	44 ± 6.1	42 ± 7.6	33 ± 8.0	28 ± 9.2	13 ± 5.3
	Night	57 — 9.2	38 ± 5.7	36 ± 7.1	29 ± 6.8	25 ± 7.9	13 ± 6.6

The 98-percentile concentrations (based on 12-hour average concentrations) are a factor of 2 - 3 higher than the mean values. The concentrations range from 9 and 65 µg/m^3 during the day and 9 to 57 µg/m^3 during the night. The 98-percentiles of the hourly average concentrations range from 12 to 87 µg/m^3 during the day and 12 to 83 µg/m^3 during night-time.

The largest difference between day and night occur in the inner city and is caused by differences in traffic emissions. The contribution from road traffic is shown in Figure 8 and summarised in Table 12.

Table 12. Spatial average concentrations (µg/m^3) due to all sources and only road traffic and percentage contributions from road traffic to the total NO_2 concentrations. Note that the spatial resolutions are different for different areas.

	Inner City of Stockholm (100 m)		City of Stockholm (200 m)		Greater Stockholm (500 m)		Södertälje (100 m)		Whole County (2 km)	
	Day	**Night**	**Day**	**Night**	**Day**	**Night**	**Day**	**Night**	**Day**	**Night**
All sources	27	20	21	16	12	9.8	12	8.7	5.1	4.7
Only road traffic	17	11	13	6.9	6.3	3.8	5	2.6	0.8	0.5
Percent from road traffic	65%	54%	60%	43%	52%	39%	43%	30%	15%	10%

Highest road traffic contribution to the average NO_2 concentrations occur in central Stockholm, which is the most densely trafficked area of the county. The spatial average contribution from road traffic in the inner city of Stockholm is 17 µg/m^3 during the day and 11 µg/m^3 during the night. On average for the Inner City of Stockholm, road traffic contributes with 65% and 54% to the total mean NO_2 concentrations during day and night, respectively (Table 12). The contribution decrease as both the area considered increase and the spatial resolution becomes less detailed.

Note that all calculated values in Table 12 correspond to roof level. Evidently the traffic contribution is higher in street canyons and close to roads.

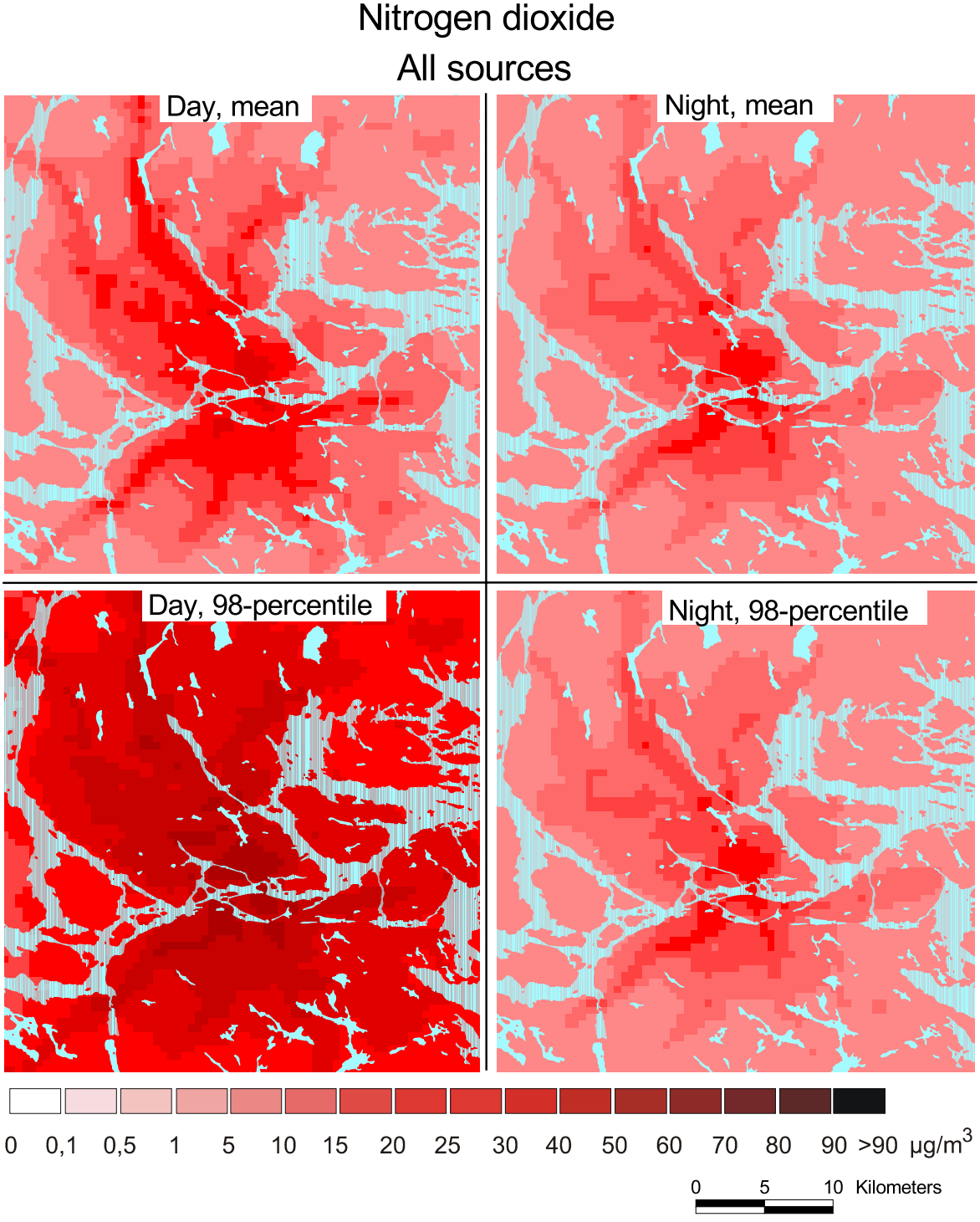


Figure 4. Concentrations of NO2 during daytime (07 to 19) and nighttime (20 to 06) in Greater Stockholm area. Note that the 98-percentile values correspond to daily mean concentrations. The values represent roof level in built up areas and about 2 m above ground in open areas.

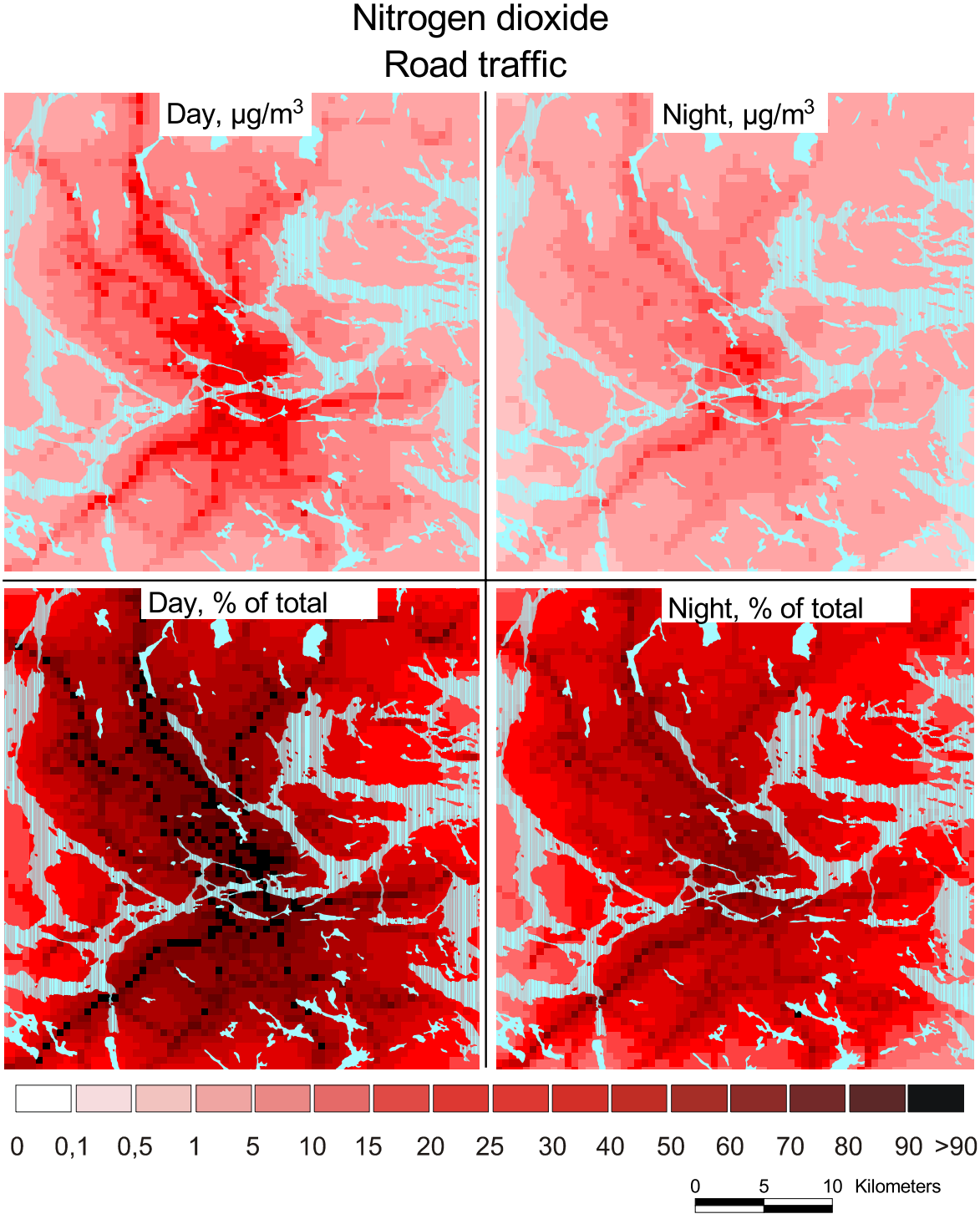


Figure 5. Contribution from road traffic to the concentrations of NO2 during daytime (07 to 19) and nighttime (20 to 06) in Greater Stockholm area. Note that the 98-percentile values correspond to daily mean concentrations. The values are given as % of the total concentration and represent roof level in built up areas and about 2 m above ground in open areas.

9.2 Population exposure to NO_2

9.2.1 All sources

Figure 10 below shows the number of people exposed to different levels of NO_2. The concentration interval is 2 µg/m^3. All calculation areas in the county are included with a geographic resolution between 100 m in the most densely populated area of the inner city and 2000 m in the less populated areas outside of the Greater Stockholm and Södertälje areas.

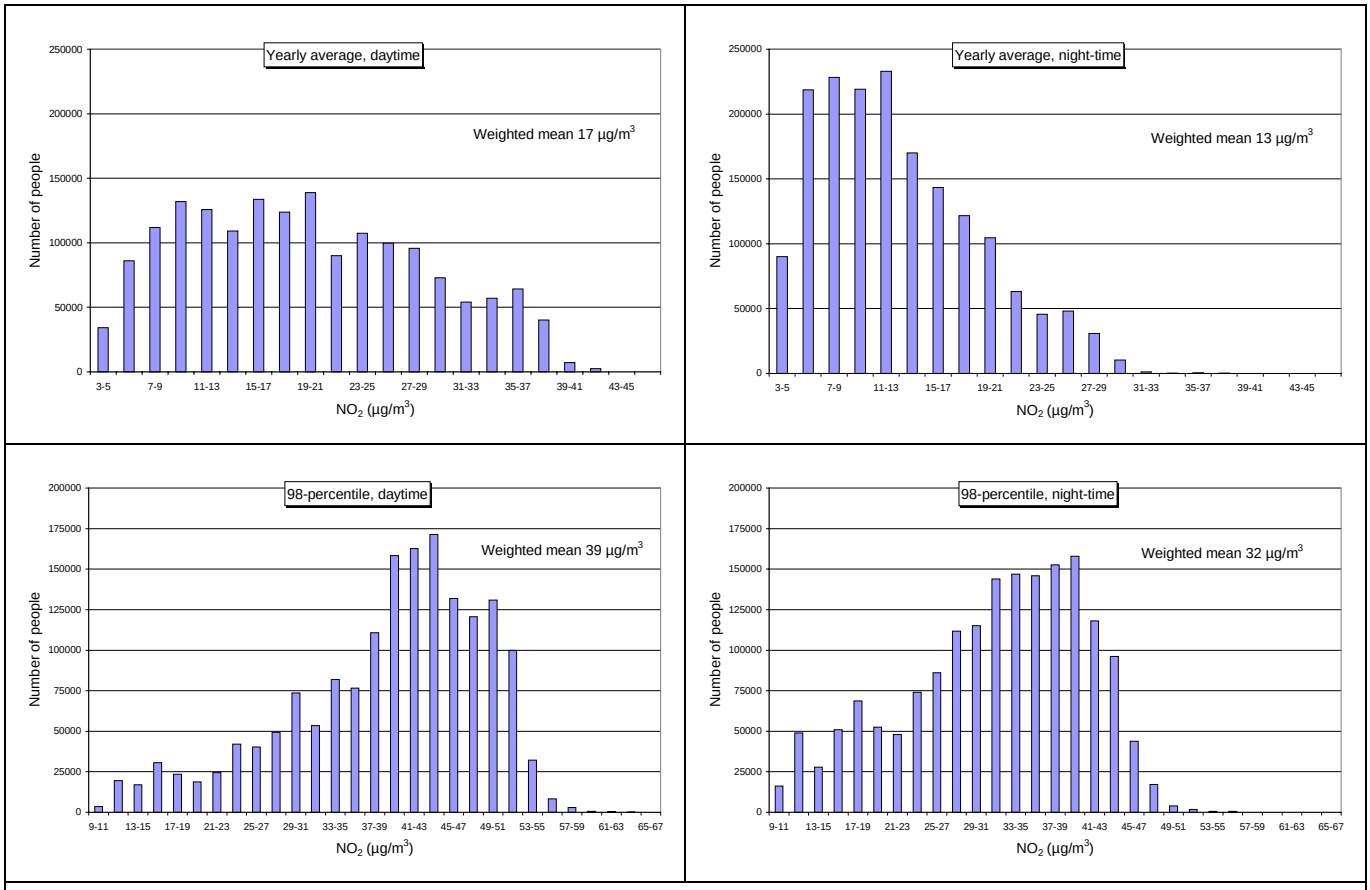


Figure 10. Number of people (at home and at work) exposed to different NO_2 concentrations in the county of Stockholm. Upper panel shows mean concentrations for day and night and lower panel 98-percentiles of 12-hour mean concentrations.

There are large differences in the number of people exposed at different concentrations. As expected, much more people are exposed at higher concentrations during the day compared to the night.

For the whole county of Stockholm the population-weighted[5] average concentrations are 20 and 13 µg/m^3 for the day and night, respectively. The

[5] Population weighted average $\frac{\sum_{\text{Area}} c \cdot P}{\sum_{\text{Area}} P}$; where c is the concentration and P is population.

corresponding 12 hour 98-percentile values are 39 and 32 µg/m^3, for day and night, respectively (see Table 13) and the one hour average 98-percentile concentrations are 63 and 53 µg/m^3, for day and night, respectively.

The weighted concentrations are higher than the non-weighted since there is a significant spatial co-variation between the concentrations and the number of people, i. e. most people live or work in areas where the highest concentrations are observed. No significant difference can be seen in the mean values for men and women.

About 7 000 people are exposed to more than 40 µg/m^3 as a mean value during the day[6]. This corresponds to 0.6% of the daytime population in the county (Figure 11). During the night no one is exposed to more than 40 µg/m^3. For the 98-percentile there are about 600 people, corresponding to 0.06% of the population, who are exposed to more than 60 µg/m^3 during the day and none during the night.

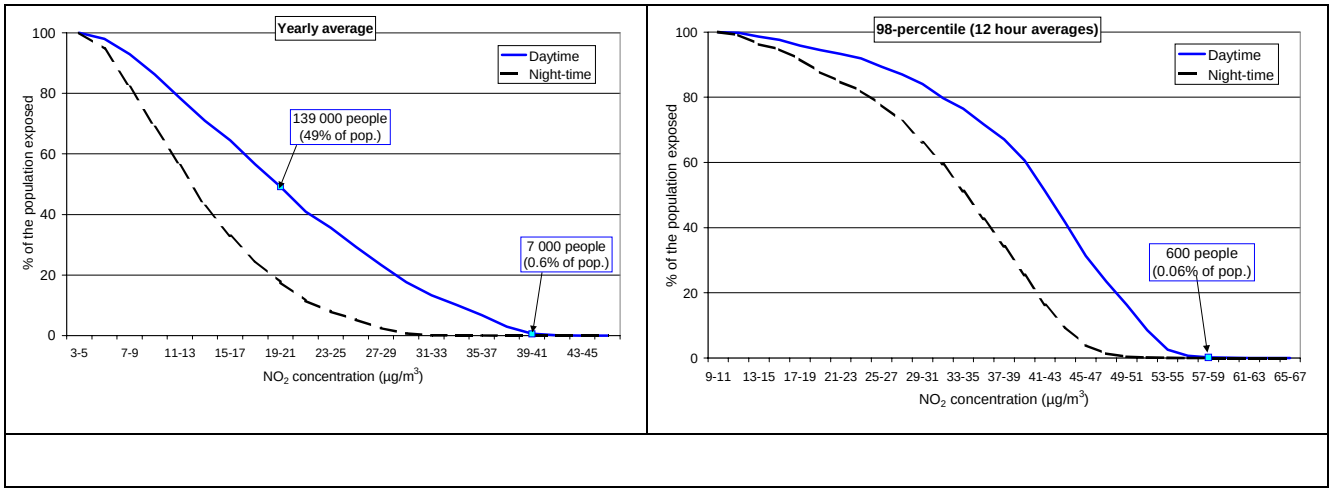


Figure 11. Percent of the total population in the county exposed to higher concentrations than indicated on the x-axes.

For the inner city of Stockholm (100 m by 100 m geographical resolution), i. e. the calculation area with the highest population density and most concentrated emissions, the population weighted average concentrations are 30 and 22 µg/m^3 for day and night, respectively. Within this area 1.3% of the population are exposed to higher concentrations than 40 µg/m^3 during the day. The 98-percentile concentrations are 47 and 41 µg/m^3, for day and night. About 0.1% of the population in this area are exposed to more than 60 µg/m^3 during the day.

[6] These concentrations may be compared with the new limit values in Sweden — (i) 40 µg/m^3 as an annual average; (ii) 60 µg/m^3 as a 98-percentile of the daily averages; (iii) 90 µg/m^3 as a 98-percentile of the hourly averages during a year (SFS 1998:897). These values should not be exceeded in urban areas after December 31, 2005. Note, however, that the 98-percentiles of the daily concentrations calculated here (for day and night), are averages for 12 hours (not 24 hours as defined for the limit value).

Table 13. Summary of calculated NO_2 population-weighted concentrations in the county of Stockholm. All data correspond to one year. Unit: µg NO_2/m^3.

			Population-weighted average				
Value	Time	Concentration interval[1)] Maximum — minimum value	Inner city of Stockholm (100 x 100 m)	City of Stockholm (200 x 200 m)	Greater Stockholm (500 x 500 m)	City of Södertälje (100 x100 m)	Whole county (variable spatial resolution)
Mean	Day	45 — 4.1	30	27	20	16	20
	Night	37 — 4.0	22	19	14	10	13
98-perc (1-h Mean)	Day	87 — 12	63	58	58	46	48
	Night	83 — 12	55	49	48	35	40
98-perc (12-h mean)	Day	65 — 9.3	47	48	43	38	39
	Night	57 — 9.2	41	41	36	30	32

[1)] Not weighted.

There are very few earlier estimates of population exposure. Steen and Cooper (1992) estimated that about 13% of the residents of Stockholm are exposed to NO_2 levels exceeding 110 µg/m^3 as a 98-percentile of the hourly average concentrations during a winter season. This estimate was also based on day- and night-time population data and dispersion model calculations, but the geographic resolution and emission data base were not as detailed as in the present study. According to the calculations presented in this report, using emission data for 1995, no one is exposed to more than 110 µg/m^3 (as a 98-percentile of the hourly mean values during one year).

As discussed above (see Population data) a more complete population exposure estimate should consider peoples' activity and time of exposure in different environments. According to Leksell and Löfgren (1995), a substantial part of the population exposure (inhaled dose) of NO_2 may be due to exposure in the traffic environment (inside cars, pedestrians, people on bicycle etc.).

Measurements at Hornsgatan and Sveavägen in central Stockholm show that at street level, in busy streets, the annual mean concentrations of NO_2 are a factor of 1.6 to 2 higher. The 98-percentiles of the daily mean concentrations are a factor of 1.3 to 1.6 higher. The difference is higher during the day due to the larger influence on the concentrations from traffic.

In general, the difference between street canyon concentrations and roof level concentrations depends on the traffic composition, traffic volume, street width, house heights and the direction of the street (compared to the wind direction). For NO_2 it is somewhat more complicated than for other less reactive compounds. The NO_2 concentrations within a street canyon are highly dependent on the ozone concentrations. Most NO_x is emitted as NO, which is oxidised to NO_2 by ozone within a few minutes, depending on the ozone concentration. The ozone concentration at street level in a street canyon depends on the roof level concentration and atmospheric mixing processes inside the canyon. Therefore, the highest concentrations of NO_2 at street level in the city are usually recorded during the spring to early summer when the ozone concentrations are highest.

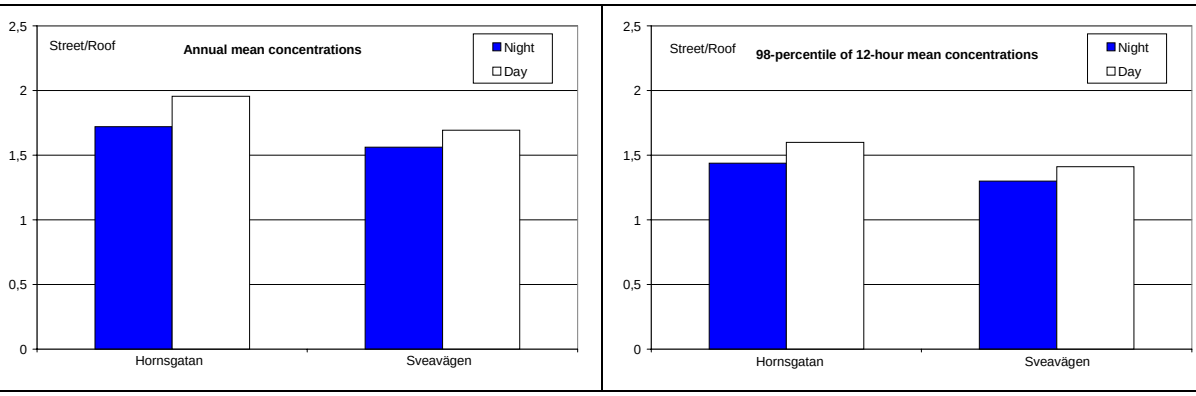


Figure 12. Ratio of street and roof level NO_2 concentrations at two busy streets in central Stockholm. Left panel shows annual mean concentrations and right panel shows 98-percentile values for 12-hour mean concentrations.

A comparison of the measured NO_x concentrations at Hornsgatan and Sveavägen shows that the amount of NO_2 of the total NO_x concentration is 30% at Sveavägen and only 20% at Hornsgatan. The most likely explanation for this difference has been found to be differences between the width of the street canyons at these sites. The canyon at Sveavägen is wider compared to Hornsgatan. Therefore ozone is more efficiently mixed into the canyon at Sveavägen, resulting in more efficient oxidation of NO to NO_2.

For population exposure it is not only the outdoor street level concentrations that are important, but also the indoor concentrations at different floors in the house depending on the location of the air ventilation intakes. A more detailed estimate of population exposure would thus also need to consider the number of people living and working at different heights in high-rise buildings in the city and the type of ventilation system in the building.

However, since population activity data is not available for this project a more detailed exposure estimate has not been made.

9.2.2 Exposure due to road traffic

Table 14 below, shows the contribution to the NO_2 exposure due to local road traffic for the different calculation areas. Note that only road traffic within the calculation area is included in the calculations.

The largest contributions from road traffic occur in the inner city of Stockholm; 74% of the yearly population-weighted average daytime NO_2 concentration is due to local road traffic. During night the contribution is 62%. The road traffic contribution decreases as the area increases due to the increasing importance of other sources for the NO_2 concentrations (including contribution from sources outside the region).

Table 14. Yearly average NO_2 concentrations ($\mu g/m^3$) due to local road traffic. Numbers in parenthesis indicate percentage contribution from road traffic to the total population weighted average concentrations.

	Population-weighted average				
Time	Inner city of Stockholm (100 x 100 m)	City of Stockholm (200 x 200 m)	Greater Stockholm (500 x 500 m)	City of Södertälje (100 x100 m)	Whole county (2 km x 2 km)
Day	22 (74%)	20 (73%)	14 (71%)	9.6 (60%)	1.3 (23%)
Night	14 (62%)	9.6 (51%)	7.4 (53%)	4.7 (45%)	1.1 (19%)

The weighted average concentrations due to road traffic presented in Table 14 may be compared with the non-weighted concentrations presented above in Table 12. For the inner city of Stockholm the daytime road traffic contribution increases from 65% for the non-weighted contribution to 74% for the population-weighted contribution. The largest impact is observed for Greater Stockholm area for which road traffic contribution increases from 52% to 71% during the day. This is due to the spatial co-variation of traffic emissions and population density.

9.3 Concentrations of PM_{10}

Compared to NO_2 the PM_{10} concentrations show much less difference between different parts of the region and between day and night. The time averaged concentrations of PM_{10} in the county of Stockholm vary between 11 and 28 µg/m^3 seen over the total area. The average night-time concentrations vary between 11 and 18 µg/m^3 in the area (Figure 13 and Table 15). The reason for the small spatial variations is the high background levels of PM_{10}, about 11 µg/m^3, which is mainly due to long-range transport. As discussed above, for NO_2, the concentrations will be higher in street canyons and close to roads with busy traffic.

Table 15. Summary of calculated PM_{10} concentrations in the county of Stockholm. All values correspond to one year (April 1995 to March 1996).

Value	Time	Concentration interval Maximum and minimum value	Inner city of Stockholm (100 x 100 m) Ave ± Std.	City of Stockholm (200 x 200 m) Ave ± Std.	Greater Stockholm (500 x 500 m) Ave ± Std.	City of Södertälje (100 x100 m) Ave ± Std.	Whole county (2 x 2 km) Ave ± Std.
Mean	Day	28 — 11	14 ± 1.1	13 ± 1.0	12 ± 1.1	13 ± 2.0	11 ± 0.46
	Night	18 — 11	14 ± 1.1	13 ± 0.91	12 ± 0.98	12 ± 1.0	11 ± 0.43
98-perc (12-h mean)	Day	57 — 30	38 ± 3.3	37 ± 3.7	37 ± 1.0	37 ± 1.9	36 ± 0.42
	Night	57 — 30	36 ± 2.9	37 ± 4.2	37 ± 0.90	37 ± 0.97	36 ± 0.39

Spatially averaged values for Greater Stockholm are 12 and 33 µg/m^3, as a yearly average and 98-percentile of the 12-hour averages. In the most densely trafficked area of Stockholm, the total local road traffic contribution is bout 5 µg/m^3 (roof level). Re-suspension of road dust contributes with about 2-3 µg/m^3, which is 15-20 % of the total particle concentration, including the background concentration. The highest road dust contribution occurs during the spring. In the model, the only parameters that cause a geographical variation of re-suspension are traffic flow, vehicle speed and vehicle composition. Sanding/salting and meteorology are assumed to have the same effect on all streets in the area. One might expect significant differences in sanding/salting between different roads. Precipitation amount may also vary over the region.

The spatially averaged annual contribution from road traffic within the inner City of Stockholm to the total PM_{10} concentration is about 16% during the day and 10% during the night (Table 16). These values represent roof level and include the PM_{10} concentration due to sources all outside the inner city area (including import from outside the borders of the county). During daytime, the spatially averaged road traffic contribution is 65% of the total contribution due to local sources in the Inner City area (Table 16). The corresponding value for night-time is 42%. Locally, at street level, the road traffic contribution will higher.

Table 16. Percentage contributions from road traffic (including both re-suspension and vehicle exhaust emissions) to the total PM_{10} concentrations.

	Inner City of Stockholm (100 m)		City of Stockholm (200 m)		Greater Stockholm (500 m)		Södertälje (100 m)		Whole County (2 km)	
	Day	Night	Day	Night	Day	Night	Day	Night	Day	Night
All sources (incl. import)	14	14	13	13	12	12	13	12	11	11
Only local road traffic	2.3	1.5	1.6	1.0	0.83	0.46	0.55	0.30	0.041	0.068
% from road traffic (incl. import)	16%	10%	12%	8%	7%	4%	4%	2%	0.3%	0.6%
% from road traffic (excl. import)	65%	42%	66%	48%	62%	36%	29%	25%	20%	34%

In residential areas wood combustion and other heating processes are the dominating local sources of PM. In residential areas south of Stockholm the highest contribution from wood burning is between 2 and 6 $\mu g/m^3$. This corresponds to up to 50% of the total PM_{10} concentration during the night. In the inner city of Stockholm wood combustion contributes with up to 10% of the total concentrations.

It should be noted, though, that in the emission database the local emissions due to residential wood burning are kept constant during day and night. In reality, one might expect somewhat higher emissions from residential wood burning during the night. This would increase the night-time population exposure.

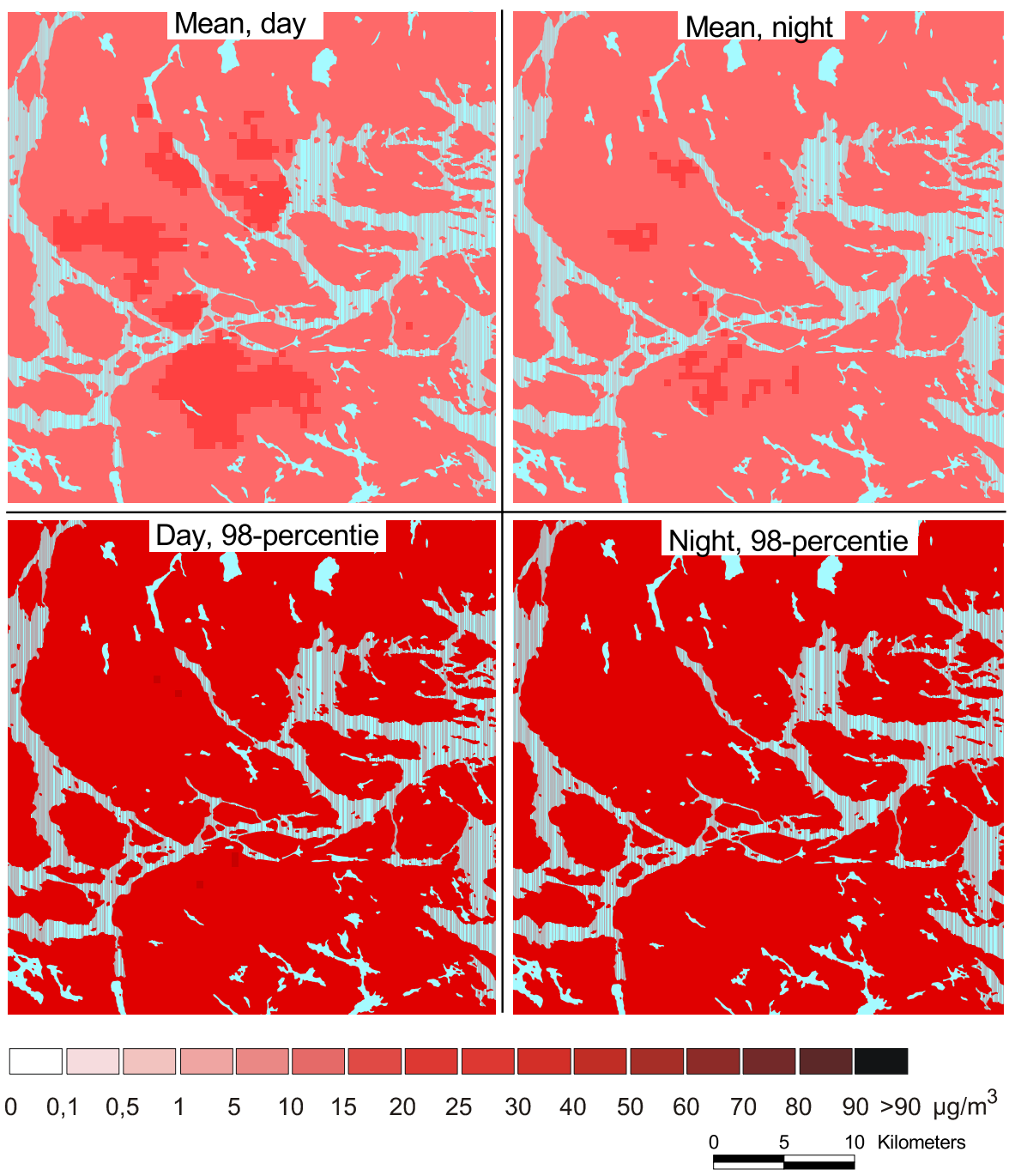


Figure 7. Concentrations of PM10 during daytime (07 to 19) and nighttime (20 to 06) in Greater Stockholm area. Note that the 98-percentile values correspond to daily mean concentrations. The values represent roof level in built up areas and about 2 m above ground in open areas.

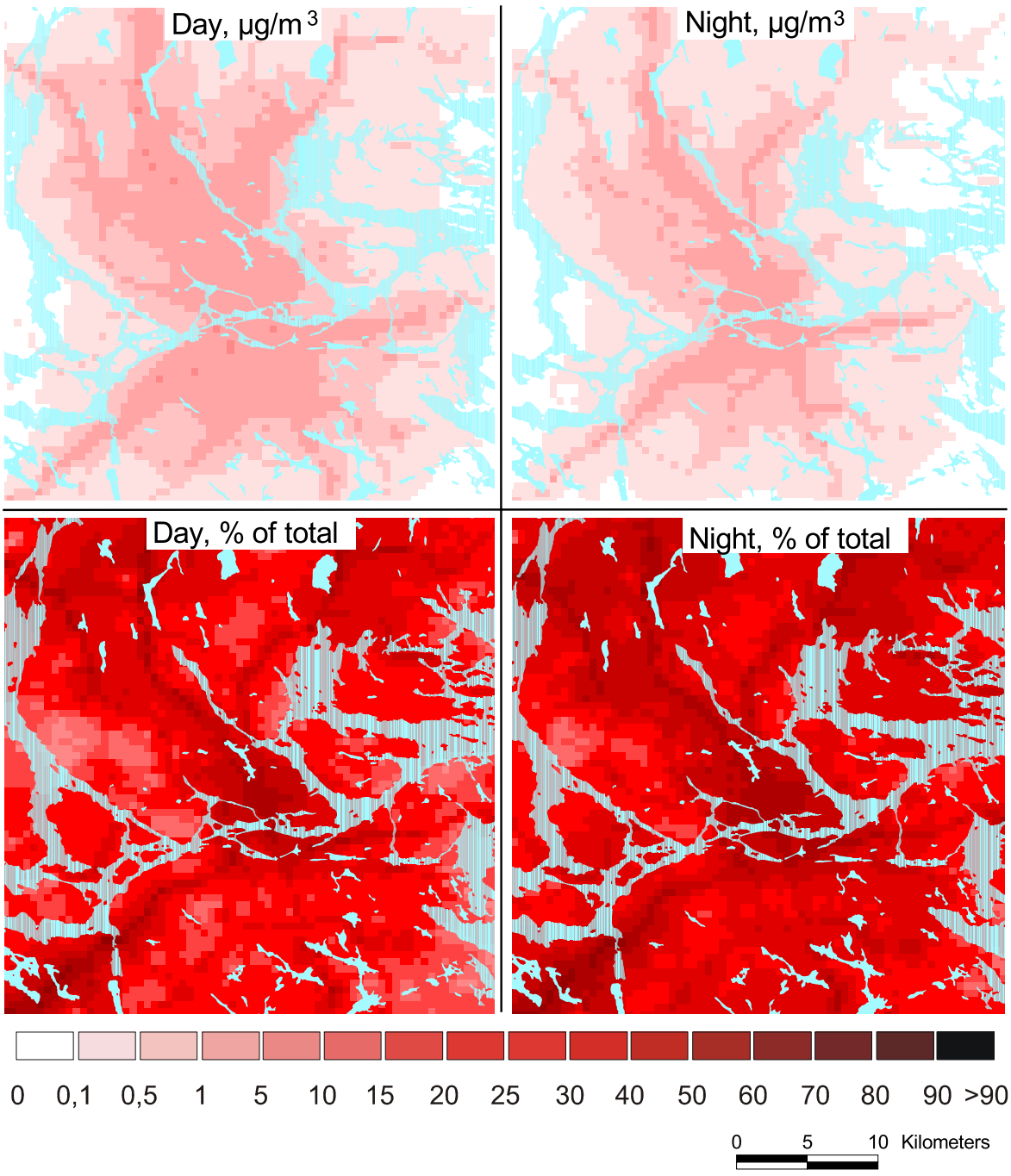


Figure 8. Contribution from road traffic to the concentrations of PM10 during daytime (07 to 19) and nighttime (20 to 06) in Greater Stockholm area. Note that the 98-percentile values correspond to daily mean concentrations. The values are given as % of the total concentration and represent roof level in built up areas and about 2 m above ground in open areas.

9.4 Concentrations of $PM_{2.5}$

The model calculations presented above represent only PM_{10} concentrations. From the health effects point of view, fine particle concentrations ($PM_{2.5}$) might be more appropriate, although it may be debated which measure is the most appropriate — mass, number of particles or surface area of the particles (see Brook et al., 1997, and references therein). In the present model calculations no distinction is made between PM_{10} and $PM_{2.5}$.

Some information on the relative amounts of PM_{10} and $PM_{2.5}$ can be obtained from measurements. Different sources contribute to the concentrations of these two particle-size fractions — sources that vary differently in time and space. Also the removal rates of fine and coarse particles are different (Seinfeld, 1986). Since fine particles travel long distances and are extensively mixed in the atmosphere, they should be distributed evenly over urban areas. Measurements indicate that the concentrations of coarse particles (equal to PM_{10} minus $PM_{2.5}$) may be quite variable from site to site (Wilson and Suh, 1997). This is in agreement with current understanding of particle transport processes.

As shown by Wilson and Suh (1997), PM_{10} and $PM_{2.5}$ concentrations are highly correlated at sites where the concentrations of fine particles dominate the PM_{10} concentration. At sites with high levels of coarse particles (PM_{10} – $PM_{2.5}$) and low levels of fine particles, the correlation between PM_{10} and $PM_{2.5}$ is not as high. Some of the lack of correlation between coarse and fine particles may also be due to poor precision in the measurement of coarse particles.

As pointed out by White (1998), random measurement errors of coarse particle concentrations may obscure the regression of coarse and fine particle concentrations. This would be particularly important in situations when the coarse particles form a large part of the PM_{10} concentrations.

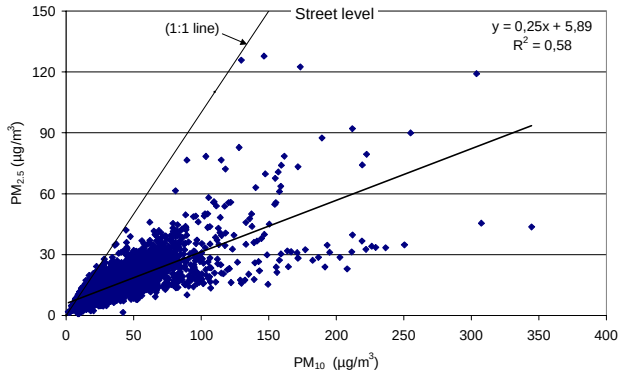


Figure 15: The relation between $PM_{2.5}$ and PM_{10} at street level (Hornsgatan, April to June 1997 and April to June 1998). The line is based on a linear least squares regression estimate forced through the origin.

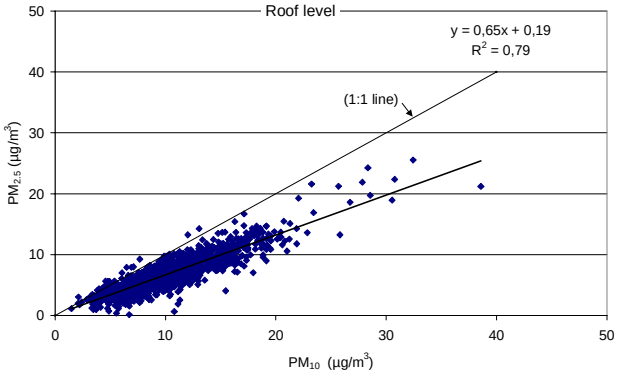


Figure 16: The relation between $PM_{2.5}$ and PM_{10} at roof level (Rosenlundsgatan, April to Sept. 1998). The line is based on a linear least squares regression estimate forced through the origin.

There are very few simultaneous measurements of the concentrations of PM_{10} and $PM_{2.5}$ in Stockholm. An idea of the relation between $PM_{2.5}$ and PM_{10} may be obtained from measurements made at two different sites — at roof level at Rosenlundsgatan in the spring and summer of 1998 and at street level at Hornsgatan during April to June 1997 and April to June 1998 (Figure 15). In addition there are data from Aspvreten (background air) during the spring and summer of 1998.

At street level the $PM_{2.5}$ concentrations are about 43% of the PM_{10} concentrations, whereas at roof level $PM_{2.5}$ concentrations are about 70% of the PM_{10} concentrations (Figure 16 and Figure 15). The larger amount of coarse particles (PM_{10} – $PM_{2.5}$) at street level may be due to re-suspension of road dust (Wallin, 1998).

Note, however, that the measurements presented in Figure 16 and Figure 15 were not taken at the same time period. The roof level data are from April to September 1998, whereas the street level measurements are from the period April to June in 1997 and April to June in 1998. Since the contribution from re-suspension is highest in the spring, the influence of re-suspension may be higher for these data than if other parts of the year would be considered — especially at the street, re-suspension may be very important. This is illustrated in Figure 17, showing the concentrations of coarse particles (measured as PM_{10} minus $PM_{2.5}$) at three different sites during autumn, winter and spring in central Stockholm. As can be seen, there is a substantial difference between the concentrations during the spring as compared to the rest of the year. Even though, some of the difference may be due to different conditions at the sites, these data are consistent with the model calculations that indicate a substantial influence on coarse particle concentrations due to re-suspension during springtime. (The reason for the negative values is that the PM_{10} and $PM_{2.5}$ measurements were not made at exactly the same time, but within a period of a few weeks).

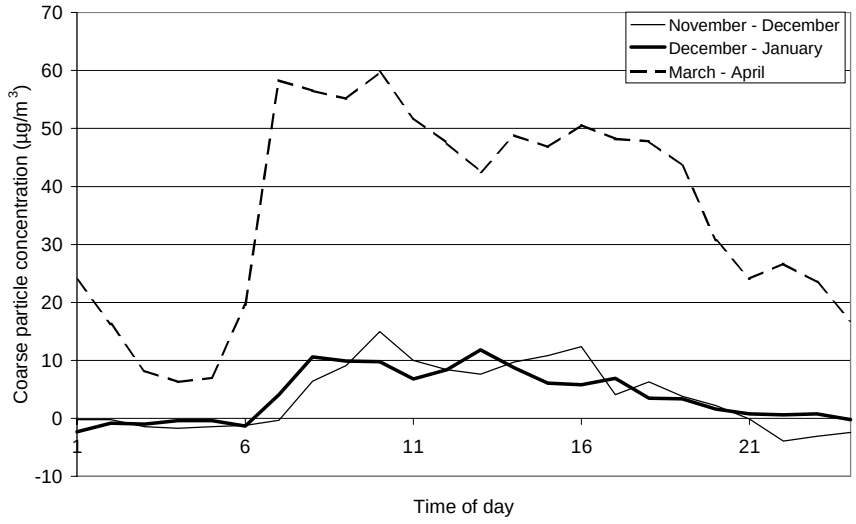


Figure 17. Coarse particle concentrations at three different street level sites in central Stockholm. The measurements of PM_{10} and $PM_{2.5}$ at the sites were not made at exactly the same time, but within the same four to six week period.

Farther out from the central city, and at some distance from main roads, the percentage $PM_{2.5}$ of PM_{10} concentrations may be assumed to increase due to decreased influence on the concentrations of road dust and more efficient removal of coarse particles. In clean, background air at Aspvreten (about 70 km S. Stockholm), fine particles constitute a larger fraction of PM_{10} compared to inside the city. Here, $PM_{2.5}$ is about 75% of PM_{10}. Most of the particles at this site are due to long distance transport, but there might also be some contribution of coarse particles from the Baltic Sea.

It is also important to note that there is a substantial variation of the relative amounts of PM_{10} versus $PM_{2.5}$ depending on the time of the day. At street level the PM_{10} concentrations show much larger difference between day and night compared to $PM_{2.5}$. Figure 18 and Figure 19 shows the average diurnal variation of the hourly mean concentrations of PM_{10} and $PM_{2.5}$ at roof and street level in central Stockholm. The concentration scale has been normalised by dividing with the minimum hourly mean value during the day (the minimum concentration is recorded early in the morning at 4 to 5 AM).

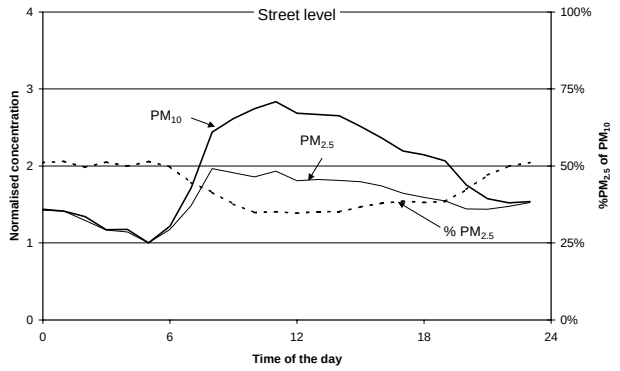


Figure 18. Diurnal variation of normalised concentrations of PM_{10} and $PM_{2.5}$ at street level in April to June in the city of Stockholm. The values are averages of data obtained during 1997 and 1998.

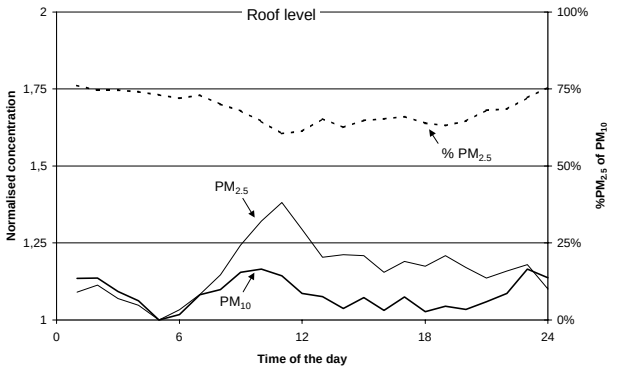


Figure 19. Diurnal variation of normalised concentrations of PM_{10} and $_{PM2.5}$ at roof level (Rosenlundsgatan, April to Sept. 1998).

For the street level, daytime PM_{10} concentrations are about 2.5 times higher than night-time concentrations; $PM_{2.5}$ concentrations are less than a factor of 2 higher during the day compared to night. In the day, $PM_{2.5}$ is about 40% of PM_{10} and increases to about 50% during the night. This is consistent with the fact that coarse particles from re-suspension of road dust, generated by traffic, is higher during the day when the traffic amount is highest. It should be noted that the measurements presented in Figure 18 are only made in spring (April to June), so that the contribution from re-suspension of road dust to the coarse particle fraction may be higher in this data than if other periods of the year would have been considered.

For the roof level, the differences between the concentrations during day and night are much less. For PM_{10} the concentrations are about 10% higher during the day compared to the night and for $PM_{2.5}$ the mean difference is less than 1%. During daytime the $PM_{2.5}$ concentrations are about 65% of the PM_{10} concentrations and during night 72%.

In Table 17 the data from Stockholm and Aspvreten are compared with data from The Netherlands and Canada. Janssen et al (1997) studied daytime PM_{10} and $PM_{2.5}$ concentrations in cities in The Netherlands. The measurements were made simultaneously near a busy road and at a background location. The mean daytime contribution of $PM_{2.5}$ to PM_{10} at a busy street in Arnhem was 58% (28 daytime eight hour averages for one site). The relationship between $PM_{2.5}$ and PM_{10} in Canadian cities was evaluated by Brook et al., 1997. A maximum of ten and a minimum of two years of data are available for 14 urban locations in Canada. On average across all sites, $PM_{2.5}$ accounted for 51% of PM_{10}. The values ranged between 36% and 65%.

The correlation between $PM_{2.5}$ and PM_{10} is high at all locations. The comparatively low value of the percentage $PM_{2.5}$ of PM_{10} in Stockholm may partly be due to the fact that re-suspension of road dust contributes with a large amount of coarse particles during the measurement periods. The yearly average percentage $PM_{2.5}$ of PM_{10} close to streets in Stockholm is therefore probably higher than 40%.

Table 17. Comparison of $PM_{2.5}$ and PM_{10} values in Sweden, Canada and The Netherlands. The values for Stockholm, Arnhem and the Canadian sites represent street level measurements.

	Arnhem[2] (The Netherlands) (1 site)	**Canada, urban (14 sites)**	**Stockholm[1] urban (1 site)**	**Central Sweden Aspvreten rural (1 site)**
PM_{10}, mean ($\mu g/m^3$)	75	28	37	8.6
$PM_{2.5}$, mean ($\mu g/m^3$)	43	14	15	6.0
$PM_{2.5}$ of PM_{10} (%)	58% only day	51%	43% day 40% night 50%	75% day 80% night 70%
Correlation between $PM_{2.5}$ and PM_{10}	-	0.86	0.60	0.86

[1] Only April to June, 1997 and 1998.

[2] Only daytime (8 hour mean values).

9.5 Population exposure to particulate matter

Calculated population exposure PM_{10} concentrations for the whole county of Stockholm are presented in Figure 20. Most people in the area are exposed to more than 11 µg/m^3 of PM_{10} as a mean value for one year but no one is exposed to more than 28 µg/m^3. This narrow range of exposure concentration is due to a weak gradient, caused by the high background concentration and a relatively small impact of local sources on roof level concentrations.

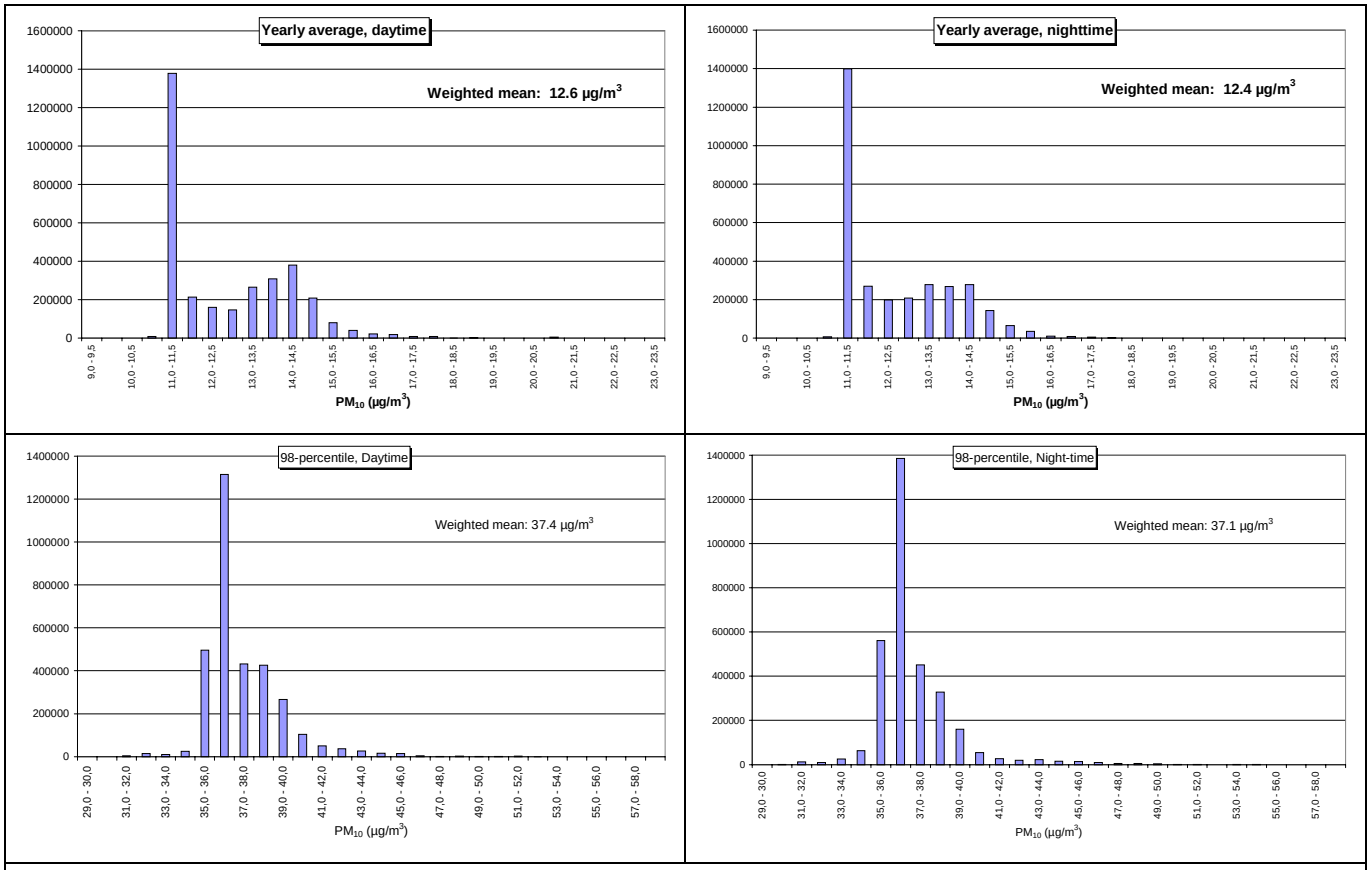


Figure 20. Number of people exposed to different concentrations of PM_{10} in Stockholm county (using 100 m resolution in Södertälje, 500 m in Greater Stockholm and 2 km in the rest of the county). Upper panel shows mean concentrations for day and night and lower panel 98-percentiles of 12-hour mean concentrations. Note the different scales on the axes.

For the 98-percentile concentrations all people are exposed to at least 30 µg/m^3 but no one is exposed to more than 60 µg/m^3.

The population-weighted average concentrations are 12 and 37 µg/m^3 for the mean and 98-percentile, respectively (Table 18). There is essentially no difference between daytime and night-time weighted concentrations. The difference in exposure (both mean and 98-percentile) between men and women is less than 0.5% of the weighted mean concentrations.

For Greater Stockholm with 500 m spatial resolution the population-weighted concentrations are only slightly higher than the concentrations obtained when the whole population in the county is considered.

It should be noted that street canyon concentrations and indoor/outdoor differences have not been considered in the calculated exposure concentrations.

Table 18. Summary of calculated PM_{10} population weighted concentrations in the county of Stockholm and comparison with non-weighted concentrations. All data correspond to one year.

			Population-weighted average				
Value	Time	Concentration interval[1)] Maximum — minimum value	Inner city of Stockholm (100 x 100 m)	City of Stockholm (200 x 200 m)	Greater Stockholm (500 x 500 m)	City of Södertälje (100 x100 m)	Whole county (variable spatial resolution)
Mean	Day	28 — 11	14.4	14.0	13.6	14.8	12.6
	Night	18 — 11	14.5	13.4	13.2	13.4	12.4
98-perc (12-h mean)	Day	57 — 30	38.7	38.2	38.2	39.8	36.1
	Night	57 — 30	36.9	38.2	37.8	38.5	36.1

[1)] Not weighted.

Table 19 shows the mean exposure concentrations due to only local emissions from road traffic. In the Inner City of Stockholm the population-weighted concentrations are 2.6 and 1.7 µg/m^3 for day and night, respectively. The non-weighted concentrations are 10% to 20% lower — 2.3 and 1.5 µg/m^3 for day and night, respectively (see Table 16). If only the local emissions in the calculation area are considered, then local road traffic emissions contribute with between 77% and 48% of the to the population-weighted concentrations due to local sources. If the long-range transported PM_{10} is included, local road traffic emissions contribute with only 10% to 20% to the total population-weighted concentrations.

Table 19. Summary of calculated PM_{10} population weighted concentrations and percentage contribution to the total population weighted concentration due to local road traffic emissions in respective calculation area.

		Population-weighted average			
Time	Value	Inner city of Stockholm (100 x 100 m)	City of Stockholm (200 x 200 m)	Greater Stockholm (500 x 500 m)	City of Södertälje (100 x100 m)
Day	Average	2.6	2.3	1.8	0.94
	% of total concentration (all sources)	18%	16%	13%	6%
	% of contribution due to local sources	77%	77%	71%	25%
Night	Average	1.7	1.4	0.91	0.48
	% of total concentration (all sources)	12%	10%	7%	4%
	% of contribution due to local sources	48%	55%	41%	20%

10. Comparison between measurements and model calculations

Validation is the process whereby the model results are compared in some way with reality. Due to the stochastic nature of atmospheric dispersion processes (turbulence) all model calculations are inexact. That is, even if all input data and the description of the turbulent mixing processes would be exact, the random nature of the turbulence in the atmosphere makes it impossible to calculate the concentrations without any errors. All dispersion models are based on assumptions that simplify the processes determining the atmospheric mixing. Such simplifications involve parameterisations of processes in order to simulate mean values both in space and in time.

Complex models may contain a large number of parameters that require a lot of input data and produce large uncertainties in the final result. Highly simplified models, on the other hand, require only very limited input data but may not be able to describe important features of the atmospheric processes. Obviously, there is an optimum of complexity in models in part depending on the availability of input data and in part on data-processing resources (Zanetti, 1990).

The total uncertainty of the model calculations is very difficult to quantify numerically. The error in one parameter may depend on the absolute value of one or several other parameters or input data. It is, therefore, necessary to compare the model calculations with accurate measurements. There are, however, a number of problems associated with the comparison of model results and measurements:

1. Time and space resolution of the model and the measurements need to be comparable

2. The significance of time and space variations need to be considered

The model calculations are based on time and space averages of emission data as well as meteorological input data. In the emission database used for the county of Stockholm much of the information is accurate with detailed descriptions on how the emissions vary both in space and in time. For some sources, such as residential wood burning, the information is not as accurate. For many sources the time variation is described in a simplified way due to lack of information, and for other sources, such as road traffic, the time variation is taken from a large number of measurements.

Meteorological input data are based on 15-minute mean values obtained from measurements at a few sites and “extrapolated” using the wind model. Input data to the wind model (topography, land use data) are given with a resolution of 500 times 500 metres. This means that the turbulent dispersion processes represent temporal mean values over such grid squares. Another problem might be that most measurements available are not primarily made for the purpose of validating dispersion models. It may therefore be difficult to find suitable data sets for model validation.

In order to assess all the potential sources of errors, a complete validation of model calculations would include a number of different measures:

- Comparison of measured and modelled mean values
- Comparison of measured and modelled extreme values
- Comparison of measured and modelled time and space variation
- Evaluation of the model's capability to predict changes in the concentrations due to changes in emission data or meteorological input data

At this time, all of these points have not been considered here. Since the calculations (at least for NO_2) are based on typical weather situations taken from a period of 8 years it is relevant to consider a few years of measurements that represent different meteorological conditions. Some of the measured variation may of course be caused by differences in the emissions between different years. The calculated values are based on the emissions of 1995.

It should be noted that measured background values have been added to the calculated values, which are based only on the local emissions within the region. For NO_2, background values are taken from Norr Malma. For PM_{10}, background values are taken from Aspvreten.

10.1 Comparison between calculated and measured NO_2 values

The comparison between the measured and calculated NO_2 concentrations in the county of Stockholm is shown in Table 20. In central Stockholm, Hornsgatan and Torkel Knutssonsgatan are only 400 meters apart while Sergel and Sveavägen are 2 to 3 km N of the two other stations (see Figure 1). All four measurements are made at roof level in the central parts of Stockholm. The other measurements presented are from other parts of Stockholm and other municipalities in the county.

Two different techniques to measure NO_2 have been used: (i) Continuous monitoring using commercial chemiluminescence NO_x analysers equipped with Molybdenum converters to convert NO_2 to NO, and (ii) monthly sampling using passive diffusional tubes according to Ferm (1991).

The average NO_2 concentrations measured at Torkel Knutssonsgatan, Sveavägen and Hornsgatan during 1994, 1995, 1996 and 1997 are between 19 and 30 µg/m^3. The calculated values using emission data for 1995 and meteorological data corresponding to the last 8 years are between 22 and 28 µg/m^3. The measured levels at Hornsgatan are systematically higher compared to the two other sites.

For all sites, the mean difference between calculated yearly mean concentrations and measured concentrations is 12% (range –20% to +53%). A linear-least-squares regression of calculated (y) and measured (x) data (16 data points) gives a slope of 0.98 ± 0.07 and an offset of 1.07 ± 1.11 µg/m^3. The correlation coefficient (r^2) is 0.93.

Table 20. Comparison of calculated yearly mean NO_2 concentrations with measured values in the county of Stockholm. A background value of 4 µg/m^3 has been used for the contribution from sources outside the county.

Site	Measurements					Model[1)]
	1994	1995	1996	1997	*Mean 94-97*	
Sveavägen[4)] (Stockholm)	24 ± 9	24 ± 13	26 ± 15	22 ± 13	*24*	28
Sergel[3)] (Stockholm)	-	26	27	27	*27*	28
Torkel Knutsg.[4)] (Stockholm)	24 ± 9	20 ± 13	21 ± 14	19 ± 13	*21*	23
Hornsgatan[4)] (Stockholm)	29 ± 11	27 ± 17	30 ± 18	24 ± 15	*28*	22
TV4-building[3)] (Stockholm)	-	18	17	16	*17*	20
Hjorthagen[3)] (Stockholm)	-	17	18	16	*17*	20
Fiskartorpet[2)] (Stockholm)	-	16	16	16	*16*	18
Sticklinge (Lidingö)[2)]	11	11	12	11	*11*	12
Svulten (Vallentuna)[2)]	-	4.2	4.0	4.0	*4.1*	5.7
Mjölsta (Norrtälje)[2)]	-	3.3	2.8	3.0	*3.0*	4.6
Norr Malma[4)] (Nörrtälje)	3.2 ± 2.3	3.5 ± 3.4	3.9 ± 4.1	3.8 ± 4.2	*3.6*	4.6
Gladö (Huddinge)[2)]	-	4.3	5.5	4.5	*4.7*	6.3
Lämshaga (Värmdö)[2)]	-	8.2	7.9	7.9	*8.0*	6.3
Kanaan[4)] (Stockholm)	11	9	10	8	*9*	8
Farstanäs (Södertälje)[2)]	-	6.5	7.2	7.2	*7.0*	6.2
Alby (Upplands Bro)[2)]	-	4.3	4.8	5.0	*4.7*	5.1

[1)] Using emission data for 1995 and meteorological data representative for typical weather situations during the past 8 years. The value given is a mean value for the surrounding grid cell (500 times 500 metres). See text for further comments.

[2)] Based on monthly sampling using passive diffusion samplers according to Ferm (1991). Data from Länsstyrelsen, Stockholm (see Länsstyrelsen, 1998 and earlier reports in same series).

[3)] Based on monthly sampling using passive diffusion samplers according to Ferm (1991). Data from EHPA, Stockholm (Slb-analys).

[4)] Based on continuous monitoring using commercial chemiluminescense analysers equipped with Molybdenum converters for NO_2 to NO conversion. Data from the Regional Association for Air Quality Management in the counties of Stockholm and Uppsala.

Table 21 shows a comparison of the measured and calculated daily 98-percentiles of the NO_2 concentrations. Only the monitoring stations with continuous monitors can be used for this comparison, since the passive samplers only provide monthly averages. The average measured values are between 42 and 54 µg/m^3, whereas the calculated values range between 49 and 51 µg/m^3.

Table 21. Comparison of calculated daily 98-percentiles of the NO_2 concentrations with measured values in the county of Stockholm.

Site	Measurement					Model[1)]
	1994	1995	1996	1997	*Mean 94 – 97*	
Torkel Knutssonsg. (Stockholm)	47	39	47	35	*42*	49
Hornsgatan (Stockholm)	56	51	62	48	*54*	48
Sveavägen (Stockholm)	46	43	54	41	*46*	51
Norr Malma (Background)	10	10	12	13	*11*	-

[1)] Using emission data for 1995 and meteorological data representative for typical weather situations during the past 8 years. The value given is a mean value for the surrounding grid cell (500 times 500 metres). See text for further comments.

The main uncertainties in the calculations of NO_2 are due to uncertainties in traffic flows and emission factors for heavy-duty vehicles and light duty vehicles without catalysts. These two vehicle types constitute about 50% each of the total emission from road traffic, which is the dominant source of NO_x close to the monitoring stations in central Stockholm. In fact, a closer examination of the traffic flows close to the measurement stations at Hornsgatan and Sveavägen has shown that the traffic flow of heavy-duty vehicles is incorrect in the emission database used for these calculations. At Hornsgatan the heavy-duty vehicle traffic flow is somewhat too high and at Sveavägen it is too low. This may explain some of the difference between the calculated and measured values.

Locally and for limited periods, the errors could be larger due to sources not considered in the calculations such as off-road vehicles, which constitute a large fraction of the total NO_x emissions. It is worth noting that the errors in calculated NO_2 levels are not proportional to the errors in the emissions of NO_x due to an exponential relationship between NO_x and NO_2.

As described earlier, the dispersion model calculates NO_x concentrations. Mean NO_2 levels are obtained from a relation between measured NO_x and NO_2 levels. Likewise, the 98-percentile values are obtained based on a relation between hourly NO_x values and daily 98-percentile values for NO_2. (The dispersion model calculates hourly 98-percentile NO_x values). This means that the measured and calculated values used for the comparison are not completely independent of each other.

10.2 Comparison between calculated and measured PM_{10} values

There is little available data of PM_{10} and $PM_{2.5}$ necessary for the validation of the calculated concentrations. In the Stockholm region there is only one monitoring station with continuous long-term data. Table 20 shows concentrations at Aspvreten (background site) and Rosenlundsgatan (roof level in the central part of Stockholm). The concentrations are remarkably constant in the period 1994 - 1997. The background level is about 10 µg/m^3 and in the city the level is between 14 and 16 µg/m^3. This means that about 70% of the PM_{10} concentrations measured at roof level in central Stockholm are due to sources outside the region.

The difference between the calculated yearly mean value and the measured values is less than 10%. Note, however, that the model calculations include only local sources, which contribute only a few micrograms per cubic meter in the city centre.

Table 22. A comparison of the calculated yearly mean PM_{10} concentrations with the measured values at Rosenlundsgatan in central Stockholm.

Site	Measurement					Model[1)]
	1994	1995	1996	1997	*Mean 94 – 97*	
Aspvreten (Background)	10 ± 6	11 ± 6	10 ± 8	-	*10*	-
Rosenlundsg. (Stockholm)	14 ± 7	15 ± 7	16 ± 8	15 ± 7	*15*	14

[1)] Using emission data for 1995 and meteorological data representative for typical weather situations during the past 8 years. The value given is a mean value for the surrounding grid cell (500 times 500 metres). See text for further comments.

A comparison of the measured and calculated daily 98-percentiles of the PM_{10} concentration is shown in Table 21. As for the mean values there is very little variation of the daily 98-percentiles between different years. The agreement is excellent between the calculated and measured values.

Table 23. Comparison of calculated daily 98-percentiles of the PM_{10} concentration with measured values at Rosenlundsgatan in central Stockholm.

Site	Measurement					Model[1)]
	1994	1995	1996	1997	*Mean 94 – 97*	
Aspvreten (Background)	27	28	34	-	*30*	-
Rosenlundsg. (Stockholm)	38	34	38	35	*36*	36

[1)] Using emission data for 1995 and meteorological data representative for typical weather situations during the past 8 years. The value given is a mean value for the surrounding grid cell (500 times 500 metres). See text for further comments.

The main uncertainties in the calculated PM_{10} values are due to uncertainties in the emission estimates for wood burning and road traffic. The latter includes uncertainties in the emissions from diesel-fuelled vehicles and road dust emissions (re-suspension).

Since there are no measurements of particulate matter in residential areas with extended wood burning, the uncertainty in the calculated values in these areas is relatively high. As mentioned earlier, the emission factors for wood burning are very uncertain.

Another source of uncertainty, even though it is expected to be small as already discussed above, is the contribution of secondary particulate matter formed through physical and chemical reactions.

There is also a lack of good-quality long-term measurements of PM_{10} and $PM_{2.5}$. In addition, the measurements are associated with some uncertainty: Loss of semi-volatile organic and inorganic compounds during filter collection may occur. If semi-volatile organic and inorganic compounds are a substantial fraction of the total mass, this will give rise to errors in the PM measurements. The loss depends on the chemical composition of the particles. Measurements of gaseous ammonia and nitric acid and particulate ammonium and nitrate indicate, however, that these compounds should not represent a large loss of particulate matter in Stockholm (Luftvårdsförbundet, 1995).

11. Relations between indoor and outdoor concentrations

For more accurate population exposure estimates it is necessary to consider the activity pattern of the population and the relations between indoor and outdoor concentrations (Sexton and Ryan, 1988). Studies have shown that people in industrialised countries spend approximately 90% of their time indoors (Levy et al., 1998). In a large scale study in the Los Angeles Basin, most of the variation (60%) in personal exposures to NO_2 was explained by indoor levels (Spengler et al., 1994). The same conclusion was obtained in an international exposure study, which showed much higher correlation between indoor concentrations and exposures than outdoor concentrations and exposures (Levy et al., 1998). It has also been found that the use of a gas stove in the home may increase the personal exposure dramatically (Levy et al., 1998).

Also for particles there are significant indoor sources such as cooking, wood burning, cleaning and other indoor activities. For epidemiological studies it may be particularly relevant to assess the rate and efficiency of infiltration of ambient particles into the indoor environment (Wilson and Suh, 1997). Measurements have shown that the infiltration of fine particles ($PM_{2.5}$) is significantly larger than that for coarse particles (PM_{10} – $PM_{2.5}$) (Wilson and Suh, 1997). $PM_{2.5}$ measurements may thus provide more relevant information for the assessment of health effects of particles.

There are very few studies in Sweden on indoor/outdoor relationships between different air pollutants. Andersson and Ulander (1992) studied the difference between indoor and outdoor concentrations of NO_2. The study included 30 apartments in 8 buildings in Örebro. It was concluded that the difference between indoor and outdoor concentrations could be attributed to the type of ventilation system. Apartments with mechanical ventilation systems with both incoming and outgoing air showed lower indoor concentrations than on the street outside, mainly because ventilation air is taken from the backyard or roof. Apartments ventilated towards the street, with or without mechanical ventilation systems, showed similar concentrations of NO_2 indoor and outdoors.

In a recent study in Stockholm, the outdoor concentrations of PM_{10}, $PM_{2.5}$, NOx, NO_2, O_3, and several volatile organic compounds were compared with corresponding concentrations inside three apartments in central Stockholm (Westerlund and Sjövall, 1997). The results are summarised in Table 24. The outdoor measurements were made 3 m above street level.

The apartments were different with respect to ventilation and their location was different with respect to nearby traffic. There is no building on the opposite side of the street at Hornsgatan 150, and the ventilation air for the apartment is taken from the street. For the apartment at Hornsgatan 104 there are buildings at both sides of the street, but here the ventilation air is taken from a backyard. The apartment at Birger Jarlsgatan has windows towards several different streets and the area is relatively open. The ventilation air is taken both from the streets and from the backyard.

The most polluted outdoor site is Hornsgatan 104, where the highest values are seen for six of the eleven parameters. For the concentrations indoors, Hornsgatan

104 shows the highest ozone levels. The reason is that the ventilation air is taken from the backyard, where ozone concentrations are expected to be higher than at the street. The relatively small impact on indoor concentrations of traffic emissions at Hornsgatan 104, is clearly seen by the relation between CO indoors and outdoors. The indoor CO level is only 30% of the outdoor level. This is about a factor of 2 lower than for the other apartments. Hornsgatan 150, ventilated towards the street, has the highest concentrations for six of the compounds.

Table 24. Summary of measurements indoors and outdoors of three apartments in central Stockholm. Unit: µg/m³ except for CO that is given in mg/m³.

	Hornsgatan 104			Hornsgatan 150			Birger Jarlsgatan 131		
Compound	In-doors	Out-doors	In/Out %	In-doors	Out-doors	In/Out %	In-doors	Out-doors	In/Out %
NO_X	67	317	*21*	102	146	*70*	42	124	*34*
NO_2	19	48	*40*	21	33	*64*	23	44	*52*
CO mg/m³	0.6	2.0	*30*	1.3	1.7	*76*	0.7	1.0	*70*
Ozon	4.4	18	*24*	-	-	-	9.2	42	*22*
Bensene	2.5	8.7	*29*	5.2	9.4	*55*	12	4.7	*255*
Xylene	7.5	24	*31*	24	23	*104*	7.5	10	*75*
Toluene	15	28	*53*	24	31	*77*	13	14	*93*
TVOC	29	67	*43*	71	69	*97*	40	31	*129*
Formaldehyde	12	5.6	*214*	13	4.5	*289*	8.2	2.7	*304*
PM_{10}	12	23	*52*	9.1	14	*65*	14	49	*29*
$PM_{2.5}$	13	20	*65*	7.5	10	*75*	13	16	*81*

Formaldehyde is the only compound that showed systematically higher levels indoors. The relatively high indoor levels of hydrocarbons at Birger Jarlsgatan are due to painting work inside the building during the measurement campaign.

On average the NO_2 concentration indoors is about 50% of the outdoor street level concentration. For PM_{10} the indoor levels are between 30% and 65% of the outdoor levels and for $PM_{2.5}$ the indoor and outdoor levels are more similar — the $PM_{2.5}$ levels indoors vary between 65% and 81% of the outdoor levels. The fact that $PM_{2.5}$ shows more similar concentrations indoors and outdoors compared to PM_{10} (higher indoor to outdoor ratio) is consistent with other studies (see Wilson and Suh, 1997; Larssen et al., 1993).

Indoor concentrations of coarse particles (PM_{10}-$PM_{2.5}$) are lower than outdoor concentrations (Table 25). This is partly explained by a larger loss in the air ventilation system of coarse particles compared to fine particles.

Table 25. Concentrations (µg/m^3) of coarse particles indoors and outdoors at three locations in central Stockholm. Note that the measurements of PM_{10} and $PM_{2.5}$ where made during different time periods. This explains why some $PM_{2.5}$ values may be slightly higher than PM_{10}.

	Indoors		Outdoors	
	PM_{10}	$PM_{2.5}$	PM_{10}	$PM_{2.5}$
Hornsgatan 104, October, 1996	13 ± 7.4	14 ± 2.7	25 ± 10	22 ± 5.4
Hornsgatan 150, November to December, 1996	9.3 ± 4.6	7.5 ± 2.2	15 ± 5.4	11 ± 1.8
Birger Jarlsgatan 131, March, 1997	12 ± 6.4	12 ± 4.9	57 ± 23	21 ± 5.8

It should be noted that the indoor concentrations have been compared with concentrations in the street right outside the apartments. The exposure concentrations presented above for NO_2 and PM_{10} (based only on the model calculations) are representative for roof level concentrations, which are lower than the concentrations at the street. No attempt has, however, been made within the present study, to estimate the total exposure considering also indoor/outdoor differences.

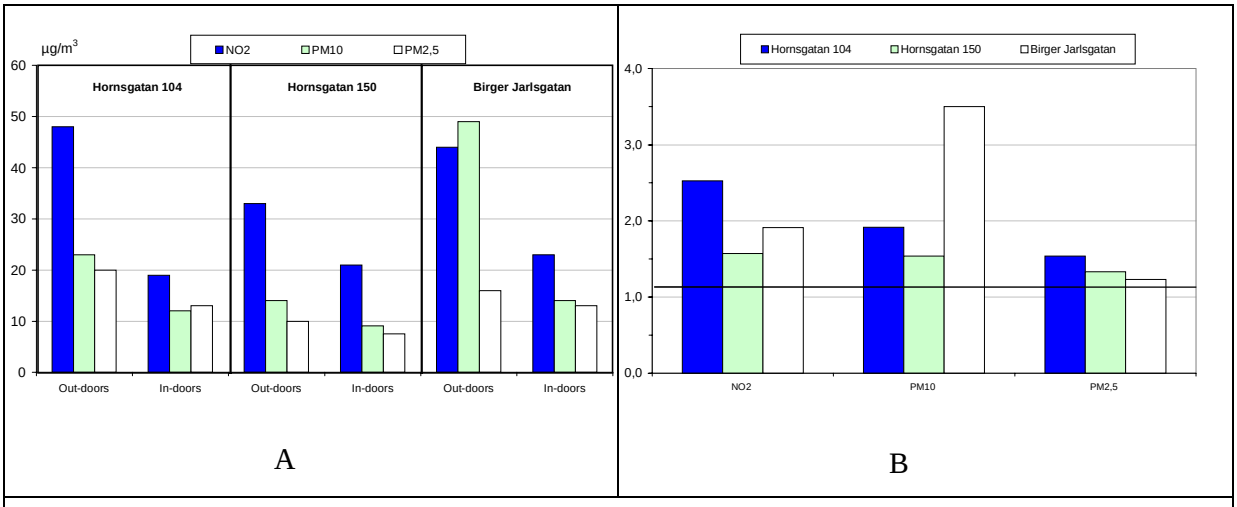


Figure 21.

A. Comparison of indoor and outdoor concentrations of NO_2, PM_{10} and $PM_{2.5}$ in three different apartments in Stockholm. The outdoor concentration measurements were made 3 m above the street right outside the apartments.

B. Quotient of outdoor versus indoor concentration for the different apartments.

12. Conclusions

Mean and extreme values of NO_2 and PM_{10} have been calculated for different areas within the county of Stockholm. The annual mean NO_2 levels range between 4 µg/m^3 and 45 µg/m^3 and the dominating source is road traffic, which contribute with 65% and 54% to the mean level during day and night, in the inner city of Stockholm. The spatial average contribution from road traffic in the inner city of Stockholm is 17 µg/m^3 during the day and 11 µg/m^3 during the night.

The annual mean PM_{10} concentrations range from 11 µg/m^3 to 28 µg/m^3 — a narrow interval caused by a high background concentration and relatively small contributions from local sources. The main sources are long-range transport, road traffic, re-suspension of road dust and wood burning. The spatially averaged local road traffic contribution (only road traffic within the calculation area, including re-suspension of road dust) to the total PM_{10} concentration in the inner city of Stockholm is 16% and 10% for daytime and night-time, respectively. In residential areas wood burning is the most important local source of particulate matter; up to 50% of the total concentration is estimated to be due to local emissions due to wood burning in these areas.

Measurements in central Stockholm indicate that the fine particle fraction ($PM_{2.5}$) contributes with about 70% and 50% to the PM_{10} concentration at roof level and street level, respectively. The $PM_{2.5}$ fraction is somewhat higher during night-time compared to daytime.

There is, generally, a good agreement between model calculations and measurements. In general the calculated NO_2 and PM_{10} concentrations are within 10% to 25% of the measured values in the city of Stockholm. For PM_{10} there is, however, very limited measurement data for comparison. The most important uncertainties in the NO_x calculations are due to emissions from heavy-duty vehicles and non-catalyst light duty vehicles. For particulate matter, emissions from wood burning, heavy-duty diesel vehicles and re-suspension of road dust are probably the most important sources of uncertainty for the calculated concentrations.

The mean population-weighted NO_2 levels are substantially higher for the daytime population as compared to night-time. For the whole county the daytime mean population weighted NO_2 concentration is 20 µg/m^3 and for night-time it is 13 µg/m^3. Local road traffic (i. e. only road traffic sources within the county) contributes with 23% and 19% to the average population weighted concentrations during day and night, respectively. For the most densely populated area (inner city of Stockholm) the yearly average weighted daytime concentration is 30 µg/m^3 and the night-time concentration is 22 µg/m^3. Here, the road traffic contribution is 74% and 62% during day and night, respectively. As expected, the population-weighted road traffic contributions to the total concentrations are higher than the non-weighted ones, due to the co-variation of population density and road traffic emissions.

For PM_{10} the difference in concentrations between day and night is not so large as for NO_2. The mean population-weighted levels are up 10% higher during the day and for the 98-percentile values the difference is only a few percent. It should be noted, though, that in the emission database the local emissions due to residential wood burning are kept constant during day and night. In reality, one might expect somewhat higher emissions from residential wood burning during the night. This would increase the night-time population exposure.

For the total population exposure, it is also necessary to consider the relationships between indoor and outdoor concentrations coupled to the activity pattern of different groups of the population. A study in three apartments in Stockholm show lower NO_2 and PM_{10} levels indoors compared to the measured levels in the street right outside the apartment. $PM_{2.5}$ concentrations show less difference between outdoors and indoors compared to PM_{10}. This is consistent with larger a loss of coarse particles in the air ventilation system. The type of ventilation and the location of the air intakes are important factors controlling indoor air quality. In general, concentrations at roof level are probably more appropriate than those at street level for the exposure estimates.

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