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Exposure to particles due to local and non-local sources in Stockholm - Estimates based on modelling and measurements 1997 - 2006

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A report from activities in TRAPART and IMPORT
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CONTENTS

1. Preface

This report presents results from two research projects - TRAPART (“Traffic related particles relative and absolute impact on quantifiable health endpoints”) and IMPORT (”Comparing health effects due to local and regional pollution in Stockholm”). They have both been financed by the Swedish Road Administration, Energy Agency and Environmental Protection Agency as part of the agreement of the EMFO program.

TRAPART has focussed on exposure and health effects of local traffic related sources and IMPORT has focussed on exposure and health effects of non-local sources. Christer Johansson at the Department of Applied Environmental Science, Stockholm university has been principal investigator of IMPORT and Bertil Forsberg at the Department of Public Health and Clinical Medicine, Umeå university, has been the principal investigator of TRAPART. Tom Bellander at Occupational and Environmental Health (Stockholm County Council) has been responsible for the analysis of health effects due to non-local sources.

This report presents the exposure assessments that has been carried out by Christer Johansson & Camilla Andersson at ITM and Robert Bergström (SMHI, Norrköping). As part of TRAPART Patricia Krecl (ITM) has developed and tested an instrument for measurements of black carbon.

Stockholm, September 2008

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2. Summary

2.1 *Objectives*

Optimal air pollution abatement strategies aiming at minimizing health effects, requires knowledge of the impact on people's health of different sources and different compounds. Both local sources and non-local (e.g. long-range transported air pollutants) need to be considered. TRAPART ("Traffic related particles relative and absolute impact on quantifiable health endpoints") focussed on health effects due to exposure of local traffic related sources and IMPORT ("Comparing health effects due to local and regional pollution in Stockholm") has focussed on health effects due to exposure of non-local sources. In this report we present calculations of the exposure and discuss the different source contributions. Health assessments will be presented in separate reports.

2.2 *Exposure due to LRT*

Exposure to long-range transported (LRT) particulate components was estimated using an Eulerian, three-dimensional, chemistry and transport model covering Europe (the MATCH model developed by SMHI). Daily mean contributions due to emissions from different sectors in different parts of Europe were estimated. Calculations were made for 7 years (1997 – 2003).

One of the largest contributors to LRT particulate exposure is the secondary inorganic aerosol (SIA). SIA is formed by emissions of nitrogen oxides (NOx) and sulphur oxides (SOx), which are emitted mainly from road traffic, energy production and shipping. SIA is also due to emissions of ammonia (NH_3) mainly from agricultural activities. Another contributor to the LRT aerosol exposure is primary emitted particles mainly from road transport, shipping and other combustion processes. According to the emission inventory used here most SOx (46% of total emissions) and primary PM (ca 50%) is emitted in eastern Europe, whereas western Europe is the largest source region for NH_3, NOx, non-methane volatile organic carbon (NMVOC) and carbon monoxide (CO).

For primary PM the largest contribution to the **population exposure in Sweden** was estimated to be due to emissions in Sweden; 40%, 44% and 48% for elemental carbon (EC), organic carbon (OC) and inorganic compounds (IPM), respectively as averages for 1997-2003. Primary PM emissions in eastern and western Europe contribute with between 11% and 21% to EC, OC and IPM. Eastern and western European emissions make the largest contributions to secondary organic aerosols. For sulphate (SO_4^{2-}) the largest contributor is eastern Europe (28 %), but western Europe (23 %) and international shipping (22 %) are also large contributors. For nitrate (NO_3^-) western Europe is the largest contributor (32 %). For ammonium (NH_4^+) western Europe is also the largest contributor (30 %), but the contribution from eastern Europe (25 %) and Sweden (21 %) is also important. Hence, for secondary inorganic aerosols the largest emitters of precursor gases also contribute most to population exposure in Sweden, whereas for primary PM Swedish emissions are only a few percent of total emissions, but they are the most important for the exposure.

Calculated **exposure for the Greater Stockholm region** indicate that for SIA the largest contribution is from eastern (27%) and western (23%) Europe, whereas the Swedish contribution is smaller and of the same size as the international shipping contribution. For primary PM2.5 Swedish emissions are most important for the exposure (55% of total primary PM2.5). According to the calculations the largest contributor to the particulate mass (PM2.5) is nitrate followed by sulphate. It should be noted that the secondarily formed organic compounds and soil dust emissions are not included in the calculations.

2.3 Exposure due to local road traffic

The contribution of **local road traffic to exposure** of particles and NOx were estimated using both measurements and air quality dispersion modelling. For PM, emissions are due to both combustion processes and mechanical processes. Combustion lead to formation of ultrafine particles (<0.1 μm diameter), whereas mechanical processes lead to micrometer sized particles (>1 μm). Modelling of variations in daily exposure of coarse particles due to local emissions of road dust is connected with quite large uncertainties, mainly due to the decisive influence of road wetness on emissions. Daily variations in road wetness is very difficult to predict accurately enough. Therefore, local contributions to exposure of coarse particles were estimated based on the difference between urban background levels in central Stockholm and rural level in Aspvreten. Comparison of rural measurements at Aspvreten and Norr Malma showed however that there are uncertainties in determining representative background levels due to both measurement errors and sometimes important spatial gradients in long-range transported levels. Another difficulty is to know the representativity of a single monitoring station in the city for the population exposure. It is expected, though, that temporal variations in coarse particle exposure is less important for exposure variations in the population than variations due local geographical gradients.

Vehicle exhaust particle exposure was assessed using a combination of measurements and modelling. Emission factors of particles from vehicle exhaust is less well known than for NOx. Model calculations of local vehicle exhaust exposure using NOx as indicator can be translated into PM (exhaust) if the PM/NOx ratio is known for a certain time period. Measurements of both black carbon (BC) and particle number concentrations at Hornsgatan (Stockholm) indicate that vehicle exhaust emissions of particles are decreasing more rapidly than NOx. This is not consistent with emission factors according to the EVA model of the Swedish road administration, which indicate that the emission ratio of exhaust PM/NOx has been about the same since 1998. This means that the trends in the ratio of exhaust particle/NOx need to be considered for long term estimates of exposure due to emissions of particles from vehicle exhaust using NOx as indicator,. For short term exposure (day to day variations within a year or two) NOx is a good indicator for exposure due to particle emissions from vehicle exhaust.

The PM/NOx ratio in the emissions varies depending on the source. For emissions from residential wood burning the mean ratio is around 1000 mg PM per g NOx, and for vehicle exhaust emissions it is around 20 mg PM per g NOx. The overall PM/NOx-ratio at different sites in a city will then vary depending on the relative importance of the contribution from different sources. At sites dominated by vehicle exhaust emissions the ratio will be around 20 mg g^{-1}. Measurements of BC and total particle number concentration, can be used as indicators of particles emitted from vehicle exhaust. For health impact assessments of local vehicle exhaust the calculated NOx exposure may then be related to BC or total particle number using simultaneous measurements of these parameters.

3. Introduction

The health effects of air pollution due to emissions from road traffic are well documented in numerous epidemiological and experimental studies. Although studies indicate that some components of PM, especially combustion-derived particles, are more toxic than others, it is still not possible to quantify the contribution to health effects from different components (WHO, 2004). Most epidemiological studies have characterized particle exposure at city level using one, or a few, fixed monitoring sites. This is the case for many U.S. cohort studies commonly used to estimate long-term mortality effects. In The Harvard Six Cities Study the strongest relation between daily mortality and PM was associated with combustion sources such as traffic, coal and residual oil (Laden et al, 2000). In the US NMMAPS the effect of PM10 on cardiovascular hospital admissions was found to increase with PM_{10} originating from highway emissions (Janssen et al, 2002).

In the European APHEA2 study the risk coefficient for PM10 on daily mortality increased with mean NO_2 concentration, indicating that road traffic exhaust related PM had a larger effect than the average PM exposure (Katsouyanni et al, 2001). Other studies have suggested that traffic related levels of black smoke and/or elemental and organic carbon are strongly associated with mortality or morbidity (Roemer and van Wijnen, 2001, Hoek et al, 2002). In addition, the epidemiological studies that capture the gradients in exposure to local traffic pollutants indicate an important effect of local traffic emissions, resulting in high relative risks (Roemer and van Wijnen, 2001, Hoek et al, 2002). Of particular interest is a Norwegian study of 16 000 men from Oslo, of whom 25 % died during the follow up, which used modeled nitrogen oxides (NO_x) in the residential area as the exposure indicator (Nafstad et al, 2004). NO_x was estimated for 1 km grid squares and a street contribution added for the largest streets. When the median concentration of NO_x for 1974-78 was used (10.7 $\mu g/m^3$), the relative risk for total non-violent mortality was 8 % per 10 $\mu g/m^3$ (6-11% with a 95% CI). Toxicological studies tend to confirm that combustion-derived, ultrafine, metal and organic-rich particles, such as those derived from vehicle exhaust, are effective in causing inflammation of the lung (WHO, 2004).

NO_x is a good indicator of motor vehicle exhaust in urban areas (e.g. Gidhagen et al., 2004a). Due to its long atmospheric lifetime (days) it may be considered as inert and modeled without considering complex photochemical processes. In Swedish studies NO_2 has been used as marker for traffic exhaust and significant effects on acute effects on asthmatics have been observed (Forsberg et al., 1998). Long-term exposure of traffic exhaust (with NO_2 as marker) has also been associated with respiratory problems (Forsberg et al., 1997), lung cancer (Nyberg et al., 2000), deaths in myocardial infarction (Rosenlund et al., 2006) and adult asthma (Modig et al., 2006).

Road traffic contributes to atmospheric particle pollution in several ways. Emissions of particles and combustion gases lead to increased concentrations of ultrafine particles (< 100 nm). These particles usually only cause a small increase in the local mass concentration expressed as PM_{10} or $PM_{2.5}$, but a large increase in the particle number concentration (PNC) (Johansson et al, 2005; 2007). PNC is a far better indicator of traffic exhaust emissions than PM_{10} (mass) which is strongly influenced by non-exhaust particle sources and the background aerosol (Johansson et al., 2007; Norman and Johansson, 2006). Due to the common major source (traffic exhaust) there is a good correlation between PNC and NO_x in Stockholm (Gidhagen et al, 2005; Olivares et al., 2006). Road traffic particles also emanate from wear of road surface, brake linings and tires. In Stockholm the local contribution to the PM_{10} levels of road dust is approximately 10 times higher than the mass concentration of exhaust particles (Johansson et al, 2005; 2007).

Aerosol concentration data from 31 European sites showed that particle number concentrations (PNC) increase more than proportionally to PM mass concentrations going from rural sites to kerb side locations (van Dingenen et al., 2004), indicating the increasing importance of local traffic emissions for PNC relative to PM_{10}. Even though there is a fairly good correlation between $PM_{2.5}$ (or PM_{10}) and PNC at clean sites, poor correlation is generally found at polluted sites (van Dingenen et al., 2004; Keywood et al., 1999), indicating different processes controlling local emissions of coarse and ultrafine particles at polluted sites. PM10 emissions and concentrations, but not PNC, at kerb side are controlled by road wetness (Johansson et al., 2007). Annual mean urban background PM10 levels are relatively uniformly distributed over a city, due to the importance of long range transport. For PNC, local sources often dominate the concentrations resulting in large temporal and spatial gradients in the concentrations (Johansson et al., 2007). Despite these differences in the origin of PM10 and PNC, the spatial gradients of annual mean concentrations due to local sources are of equal magnitude due to the common source, namely traffic. Thus, people in different areas experiencing a factor of 2 different annual PM10 exposure *due to local sources* (not total exposure) will also experience a factor of 2 different exposure in terms of PNC (Johansson et al., 2007). This implies that health impact studies based solely on spatial differences in annual exposure due to local sources to PM10 may not separate differences in health effects due to ultrafine and coarse particles. On the other hand, health effect assessments based on time series exposure analysis of PM10 and PNC, should be able to observe differences in health effects of ultrafine particles versus coarse particles.

4. Objectives and implementation

The purpose of IMPORT and TRAPART is to estimate the long term PM exposure of the population in Stockholm region (1997 – 2003) in order to assess the importance of different sources to different health outcomes. In IMPORT the main focus is to assess the effects on human health of the long-range transported air pollution and in TRAPART the effects of local sources are studied. The importance of the contributions of long-range transported versus local sources (mainly vehicle exhaust and road wear) need to be assessed to be able to connect health effects to the different sources.

Local traffic NOx exposure is modelled and used as a tracer for vehicle exhaust and related to different particle measures (PM10, PM2.5, PM1 and BC) via local measurements. The contribution from long-range transport to PM exposure in Stockholm is investigated by European scale dispersion modelling. The MATCH model is used to estimate the contribution of four European regions (Sweden, Denmark-Norway-Finland, western Europe and eastern Europe) and international shipping.

NOx and total particle number concentrations were used as indicators for exposure and health impact of local vehicle exhaust particles in an earlier research project (SNAP/PASTA). One aim of TRAPART was to see if black carbon is a better indicator of health effects due to local exhaust particles. In this project we therefore initialized measurements of black carbon by developing and validating a custom built BC instrument. It should be relatively cheap and easy to operate in order to evaluate spatial variations in an urban area.

5. Methods

It was decided to create a set of time series of data based only on measurements in Stockholm and at background sites. Data should be representative for population exposure to non-local and local sources. These time series will be used in statistical analyses together with different markers for health effects (e g hospital visits). The data series should be divided to reflect the following local sources:

- Road dust (includes road wear and other non-exhaust coarse particles)
- Vehicle exhaust particles (all gasoline and diesel vehicles)
- Non-local PM (all non-local source contributions)

An important question concerns the time coverage and quality of the measurement data. In this report we compare the measurement data available at the rural sites Aspvreten and Norr Malma. For local sources we use measurement data from urban sites in Stockholm.

5.1 Instruments

Particle mass concentration measurements were performed using automatic TEOM instruments (Tapered Element Oscillating Microbalance, model 1400, Rupprecht and Pataschnik) equipped with PM10 and PM2.5 inlets. NOx is measured at several sites in Stockholm using automatic instruments based on chemiluminescence (ThermoElectron).

The particle number concentration is measured at Hornsgatan and Rosenlundsgatan/Torkel Knutssonsgatan using Condensation Particle Counters. In Stockholm CPC TSI 3022, that has a lower cut-off size of 7nm (N_7) is used. At Aspvreten a DMPS is used to get the size distribution from 10 – 450 nm. Total number concentration is measured with a CPC 3010 (particle >10 nm).

5.1.1 Black carbon

During the last two years black carbon has been measured continuously at Aspvreten and in Stockholm. The same type of instruments are used, based on the light absorption of green light after collection of the particles on a filter. The instruments have been constructed at ITM, but are similar to a commercial instrument called PSAP (Particulate Soot Absorption Photometer; Bond et al., 1999). In Stockholm both PSAP's and commercial Aethalometers are used at street (Hornsgatan) and roof-top (Torkel Knutssonsgatan). The measurement principle of these instruments is the same and relies on the light absorbing properties of the carbonaceous aerosols. The method is based on the integrating plate technique (Lin et al., 1973) that measures the attenuation of light transmitted through particles that are continuously collected on a filter. A vacuum pump draws air through the instrument while the accumulated particles are illuminated by a light source at wavelength λ. The light transmission is monitored by two detectors to correct for drift in electronics, variation of light source intensity or any variation in the filter properties (e.g., relative humidity content). One detector measures the light intensity I transmitted through the filter where the particles are deposited (sample spot) and the second one measures the light intensity I_o transmitted through a non-exposed filter area (reference spot). By using the Beer-Lambert-Bouguer law, the attenuation coefficient of the particles collected on the filter b_{ATN} is calculated as

$$b_{ATN}(\lambda) = \frac{A}{Q.\Delta t} \ln\left[\frac{\left(I(\lambda)/I_o(\lambda)\right)_{t-\Delta t}}{\left(I(\lambda)/I_o(\lambda)\right)_t}\right] \ ,$$

where A is the sample spot area, Q is the flow rate, t is the current time, and Δt is the time interval (Bond et al., 1999). Filter-based absorption measurements need to be corrected for some instrumental artifacts to obtain the absorption coefficient of the airborne particles b_{ap}. As described by Weingartner et al., 2003, there are two main instrumental artifacts that change the optical properties of the particles embedded in the filter with respect to the properties of the same particles in the airborne state. The first artifact is the enhanced light absorption of the deposited particles produced by multiple scattering of the light beam at the fiber filters in the unloaded filter (C factor). The second artifact is caused by the instrument response to the increasing filter mass loading (R factor). As the filter gradually loads up with particles, there is a reduction of the filter multiple scattering. This effect can be partially compensated if light-scattering particles are also present in the filter, contributing with additional light scattering and, thus, decreasing the light transmission (Bond et al., 1999; Weingartner et al., 2003). Following Bond et al. (1999), b_{ap} is computed as

$$b_{ap} = b_{ATN} / \left[C.R(Tr)\right] - k.b_{sp} \ ,$$

where Tr is the filter transmission relative to an unloaded filter ($Tr = \frac{(I/I_o)}{(I/I_o)_{t=0}}$), k is the scattering correction factor and b_{sp} is the aerosol light scattering coefficient. The BC mass specific absorption cross section σ_a relates the absorption coefficient of the aerosol to the BC mass concentration M_{BC}. The calculation of σ_a requires the determination of M_{BC} by an independent method (e.g., thermo-optical measurements).

5.1.1.1 The aethalometers

Aethalometer series 8100 (Magee Scientific Co., Berkeley, USA) were employed to measure the absorption coefficient (Hansen et al., 1984). Aerosol particles are deposited on a Q250F filter tape (quartz fibers with a polyester backing, Pall Corporation, Ann Arbor, USA) while being illuminated by a 880 nm LED. The area of the sample spot is 1.67 cm^2. The tape is automatically advanced when the loading of the filter attenuates 25% of the original transmitted light. The aethalometers are operated on the same sampling inlet as the TEOM, switching beteen PM2.5 and PM10. Average values of 15-min are logged.

5.1.1.2 PSAP's

Custom-built particle soot absorption photometers constructed at the Department of Applied Environmental Science, Stockholm University measures the absorption coefficient by using a 525 nm LED. Aerosol particles were collected on 47 mm Tissuglass E70-2075W filters (Pall Corporation, Ann Arbor, USA) with a circular sample spot. A detailed description and validation of the instrument is given by Krecl et al. (2007).

5.1.1.3 Comparison of PSAP and Athalometer

A comparison of the commercial Aethalometer and the custom built PSAP is shown in Figure 1. The aethalometer BC concentrations were computed using a σ_{ATN} value provided by the manufacturer (16.6 m^2 g^{-1}). The two methods were found to be highly correlated ($R^2 = 0.92$). If the mass specific attenuation cross section σ_{ATN} (13.4 m^2 g^{-1}) obtained for this particular site and time period is used to calculate the aethalometer BC mass concentrations, and the PSAP absorption coefficients are converted into BC mass concentrations employing $\sigma_{a_{PSAP}} = 20.09$ m^2 g^{-1}, the linear regression equation becomes $M_{BC,PSAP} = 0.98 M_{BC,aeth}$. The correlation is linear over the whole measuring domain, even if there is relatively more scatter at low values. Since both aethalometer and PSAP measurements were normalized with respect to M_{EC}, any difference observed between them might be due to sampling or instrumental differences such as uncertainties in flow measurements, sample spot area, signal to noise ratios.

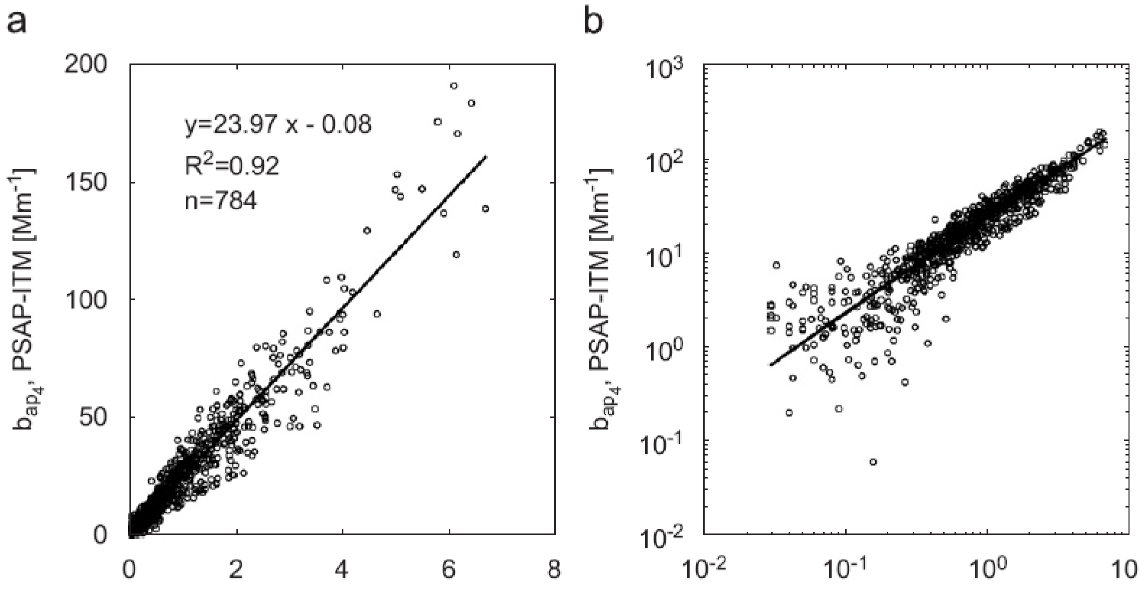

Figure 1. Comparison of measured BC concentrations using a commercial Aethalometer and a custom built PSAP (from Krecl et al., 2007).

5.2 *Measurement stations*

5.2.1 Rural (background) levels

ITM performs various studies of air pollution at Aspvreten, about 80 km south of Stockholm at the Baltic coast. The station is included in the national Swedish air-monitoring network and is also used in various research projects. The station is located away from any local sources and is representative for the regional background.

Figure 2. Locations of the rural measurement stations at Aspvreten (run by ITM, Stockholm university) and Norr Malma (run by SLB Environment and Health Administration, Stockholm).

The station is well equipped with instruments for determination of the physical and chemical properties of the aerosol. It is also equipped with meteorological instruments as well as basic instrumentation for gaseous compounds. At present (2008) the continuous monitoring program includes: particle number size distribution (from 10 to 500 nm), particle mass in two fractions (PM10 and PM2.5), particulate carbonaceous material (organic carbon, OC, and elemental carbon, EC), black carbon (“soot”), O_3 and meteorology (temperature, relative humidity, wind, global radiation and pressure).

According to the data from Aspvreten, PM10 has decreased from around 17 µg/m^3 1990 to about 8 µg/m^3 in 2007, reflecting the decreasing emissions in Europe during the last decades. However, for the period 1998 – 2004 there is no obvious systematic downward trend; it is only the last year (2007) that the levels are lower. PM2.5 levels have been approximately the same during the period 1998 – 2007;

between 7 and 9 µg/m³. Data from Aspvreten have been reported in several publications and reports (e. g. Forsberg et al., 2005; Tunved et al., 2005; Gidhagen et al., 2005; Areskoug et al., 2004; Areskoug, 2004; Nyquist et al., 2002; Alsberg et al., 2001).

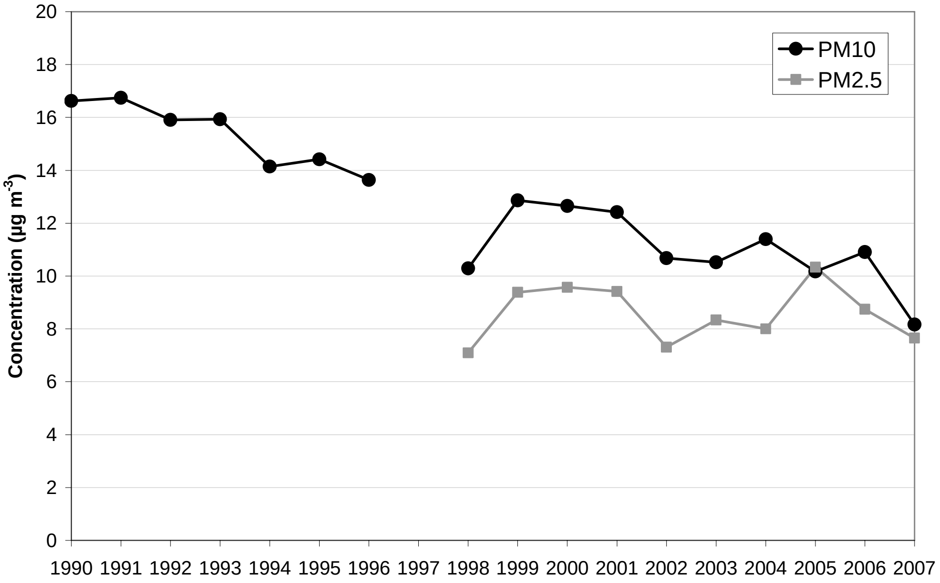

Figure 3. Trends in PM10 and PM2.5 levels at Aspvreten 1990 – 2007.

Another rural station, Norr Malma, run by SLB-analys at Stockholm Environment and Health Administration is located about 70 km NNE of Stockholm (Figure 2). At this station PM10 and PM2.5 has been measured since June 2005. Also NOx and O_3 is measured since 1994. Meteorological measurements are performed in a 12 m mast. It includes wind speed, wind direction, temperature, temperature difference, global radiation and relative humidity. All data are reported annually and reports as well as a more detailed description of the station are available at http://www.slb.nu/lvf. Annual average PM10 and PM2.5 data for the urban stations are presented in Figure 4 and Figure 5.

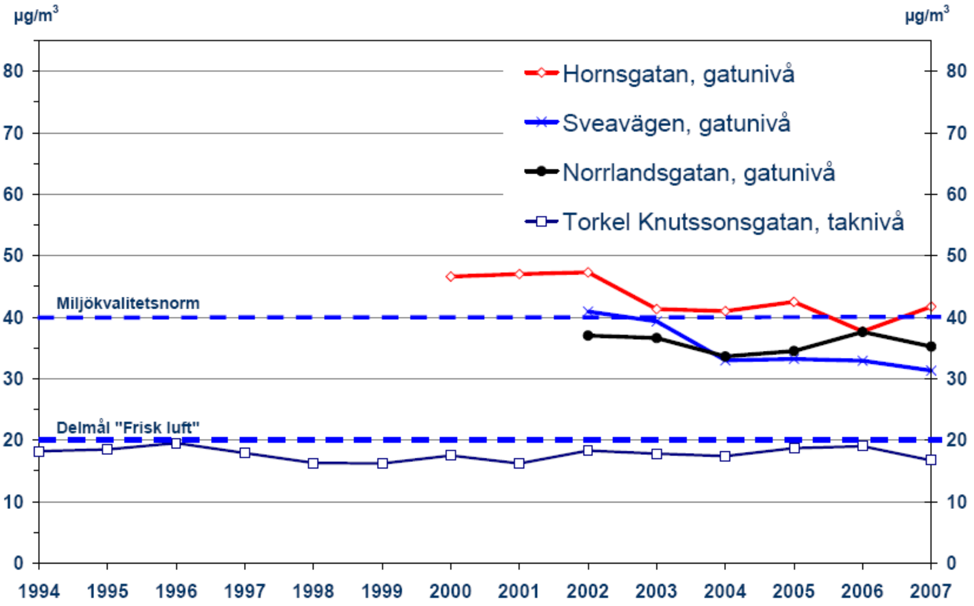

Figure 4. Trend in PM10 levels in Stockholm 1994-2007.

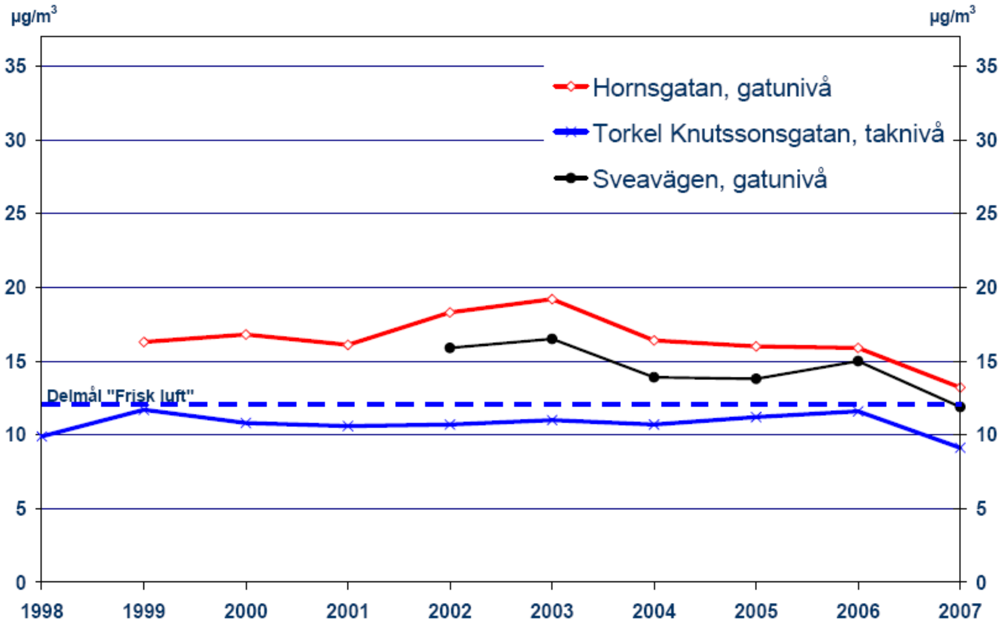

Figure 5. Trend in PM2.5 levels in Stockholm, 1998-2007.

5.2.2 Intercomparison of PM measurement methods at Aspvreten

Several intercomparison studies of gravimetric PM measurements and other instruments for automatic measurements have been performed at Aspvreten (Nyquist et al., 2002; CEN, 2006; Johansson, 2003; Ferm & Hansson, 2003). In 2002 an intercomparison between a number of gravimetric and automatic methods for PM2.5 was performed by the European Committee for Standardisation, CEN (CEN, 2006). An analysis of variance (F-test) of the data from the automatic instruments shows that the variance between the methods is larger than the residual variance in the whole data set at 95% confidence level indicating that the different methods give dissimilar results (the variation between the different methods is significantly larger than that of the whole data set). The same was found for the gravimetric data set.

Ferm and Hansson (2003) cite a number of studies were TEOM instruments have been compared to gravimetric methods and were TEOM tend to give lower PM10 levels due to several problems. The slope and intercept of PM10 levels according to gravimetric (IVL) versus TEOM are 0,84 and 5,5 respectively (Figure 6) for Aspvreten (Ferm and Hansson, 2003). Similar slope and intercept was found in another study at Aspvreten using gravimetric (Kleinfiltergerät) and TEOM (Figure 7). The intercomparison of TEOM and a gravimetric method at Aspvreten presented by Ferm and Hansson (2003) showed that measurements of PM10 using TEOM may be corrected by first eliminating the built-in standard instrumental corrections (factory settings) and then multiplying the values by 1,3: PM10(corrected) = (PM10(uncorrected)*1,03 – 3)*1,3. Both studies show quite large variabilities.

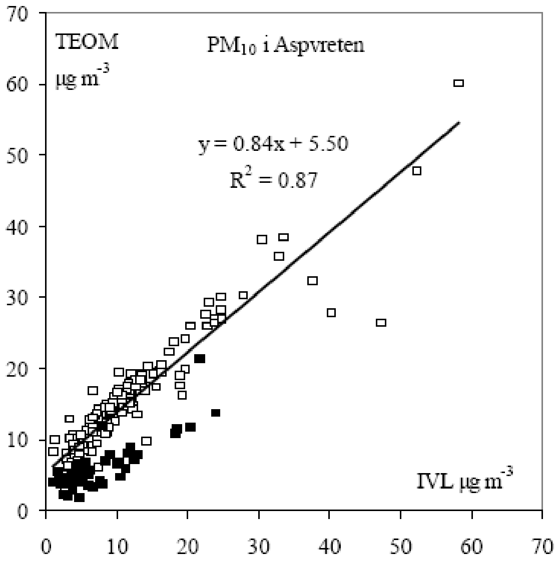

Figure 6. Comparison of PM10 levels measured using a TEOM and a gravimetric (IVL) method with mass concentration determined according to the standard procedure of the reference method (EN1234-1[1]). Black squares are data from November that is not included in the regression (Ferm & Hansson, 2003). Daily mean values.

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[1] SS-EN 12341 "Air quality – Determination of the PM10 fraction of suspended particulate matter – Reference method and field test procedure to demonstrate reference equivalence of measurement methods."

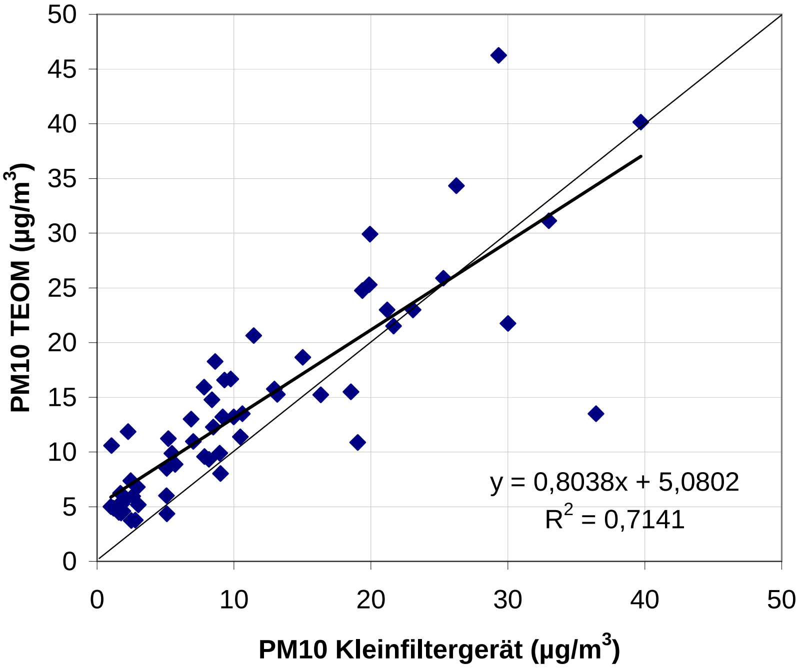

Figure 7. Comparison of PM10 levels measured using a TEOM and a gravimetric (KFG) method with mass concentration determined according to the standard procedure of the reference method (EN1234-1). Daily mean values.

Figure 8 shows a comparison of gravimetric PM2.5 measurements and TEOM, again with similar slope and intercept.

Both for PM10 and PM2.5 there can be large deviations between PM measurements using the gravimetric method and automatic TEOM measurements. This variability can be due to

- Measurement errors due to different precision of the methods and/or variability due to differences in the sampling inlet or sampling lines

- Measurement errors due to the two methods being more or less sensitive to variations in the chemical composition of the particles

It is likely that the main differences between the gravimetric method and TEOM occur due to water taken up by the particles during the weighing of the filters with the gravimetric method or volatilization of organic and inorganic compounds during the measurements with the TEOM. At Aspvreten volatilization of ammonium nitrate in the TEOM may (occasionally) explain a large part of the deviation between the TEOM and the gravimetric method (see discussion in Ferm and Hansson, 2003).

Comparisons of gravimetric and TEOM measurements of PM10 have also been made at Hornsgatan with similar discrepancies as for Aspvreten (Johansson, 2003, see Figure 8).

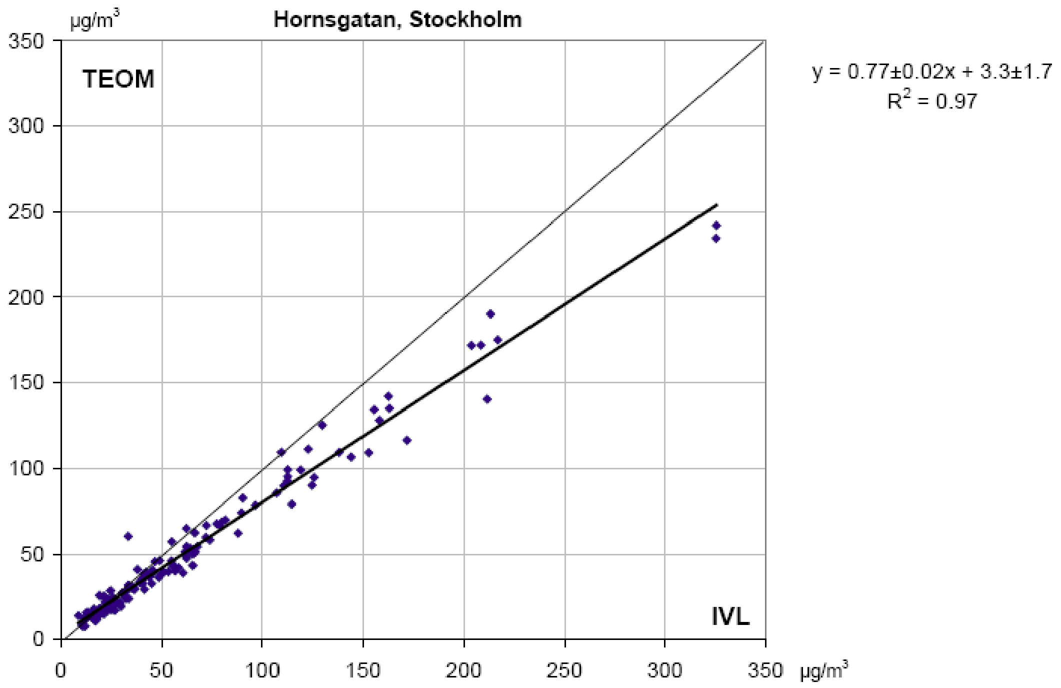

Figure 8. Comparison of PM10 measurements using a gravimetric (IVL) method according to EN1234-1 and an automatic TEOM at Hornsgatan in Stockholm (Johansson, 2003). Daily mean values.

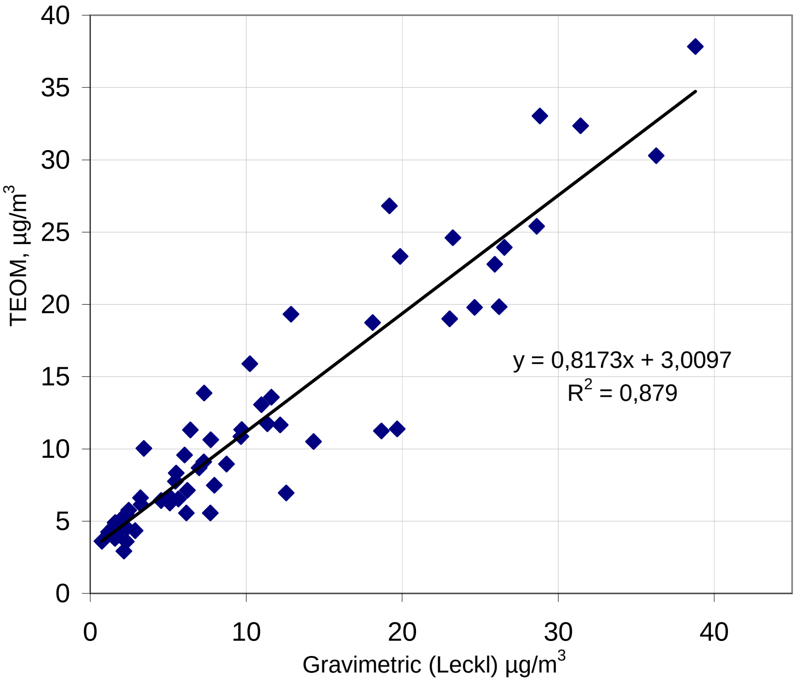

Figure 9. Comparison of gravimetric (Leckl sampler and weighing according to EN1234-1) and automatic (TEOM, R&P) PM2.5 measurements at Aspvreten. Daily mean values.

5.2.3 Urban background

Rosenlundsgatan and Torkel Knutssonsgatan are both roof top sites in central Stockholm that can be regarded as representative for the urban background since they are not directly affected by nearby local vehicle emissions. This is also verified by urban scale modelling of particle number concentrations by Gidhagen et al. (2005). In 2005 the site at Rosenlundsgatan was closed and all instruments were moved to Torkel Knutssonsgatan. Comparisons between the PM10 concentrations at these two sites by running TEOM instruments in parallel, have shown excellent agreement. Comparisons between data from Torkel Knutssonsgatan using point measurements of NO_2 and O_3 with a DOAS instrument (along a path from Rosenlundsgatan to Torkel knutssonsgatan) also show very good agreement. The measurement program at these two sites include: NOx (NO and NO_2), PM10, PM2.5, total particle number concentration (PNC).

5.2.4 Street canyon

Hornsgatan is a 24 m wide, four lane street surrounded by 24 m high houses on both sides, thus being a rather symmetric street canyon with a unity width/height ratio (Gidhagen et al., 2005). Traffic intensity (2003) is about 35 000 vehicles per day during weekdays, with an average of 5% of heavy duty vehicles (HDV:s), mostly ethanol buses. Of the light duty vehicles (LDV:s), there is an average of 5% diesel fuelled cars, mainly taxis.

Air intakes for NOx monitoring (Chemiluminescence analyzers, Thermo Electron) are placed at both sides 1.5 m from the façades, at 3 m height and also at roof level at a building some 500 m east of Hornsgatan, in a less trafficked area (urban background). Close to the NOx air intake at the North side of the street, a trailer is parked with a CPC3022 instrument (TSI Inc.) measuring total number concentrations. An identical instrument is located at the urban background station Rosenlundsgatan, about 600 m from Hornsgatan and at 30 m height.

5.3 Data available

Table 1 summarize the data available at the rural stations Aspvreten, Norr Malma and at the urban background station in central Stockholm. Non-local PM10 and PM2.5 concentrations may be obtained from urban background levels after subtracting the rural levels measured at Aspvreten or Norr Malma. The longest time series is for PM10 at Aspvreten (data from 1990), but only from 2005 at Norr Malma. Total particle number are available at both Aspvreten and Stockholm simultaneously since 2001. Simultaneous black carbon measurements were initialized within the TRAPART project and are available since 2004/2005.

Table 1. Available data from rural and urban background stations to assess the contribution to the exposure of non-local sources. Dates are start of measurements and all data are stored as hourly mean values (for some parameters 15 min mean values) from which daily mean values are calculated depending on the number of hours with data. All measurements are still running but in some cases there have been periods without measurements.

Parameter	**Aspvreten** **Rural background**	**Norr Malma** **Rural background**	**Stockholm** **Urban background**	**Stockholm** **Kerbside**[2]
PM10	1990	2005	1994	2000
PM2.5	1998	2005	1998	2000
NOx	-	1994	1982	1992
Total particle number[1]	2000	-	2001	2005
Particle number size distribution[2]	2000	-	only short periods	2007 (before this date only short periods)
Black carbon	2004	-	2006	2005
Ozone	1985	1994	1982	2008

[1] Lower cut-off at Aspvreten is 10 nm and in Stockholm the cut-off is 7 nm. [2] Range 10 – 450 nm.
[2] Dates refer to Hornsgatan, but for some parameters measurements take place at several kerb side sites.

5.4 Dispersion models

5.4.1 Local modelling

NOx concentrations were calculated using a wind model and a Gaussian air quality dispersion model, both part of the Airviro Air Quality Management System (SMHI, Norrköping, Sweden; http://airviro.smhi.se). Meteorological conditions were based on meteorological measurements (15 minute averages) in a 50 meters high mast located in the southern part of Stockholm. The wind field for the whole model domain was calculated based on the concept first described by Danard (1976). This concept assumes that small scale winds can be seen as a local adaptation of large scale winds (free winds) due to local fluxes of heat and momentum from the sea or earth surface. Any non-linear interaction between the scales is neglected. It is also assumed that the adaptation process is very fast and that horizontal processes can be described by non-linear equations while the vertical processes can be parameterised as linear functions. The large scale winds as well as vertical fluxes of momentum and temperature are estimated from profile measurements in one or several meteorological masts (called principal masts). For the model domain analysed in this study (35 km^2) only one principal mast is used. This is located in the southern part of the city. Topography and land use data for the Danard model are given by 500 meter resolution. Since the topography of Stockholm is relatively smooth, without dominating ridges or valleys, the free wind can be assumed to be horizontally uniform in the whole domain.

The dispersion calculations were performed on a 100 meter resolution (122 500 receptor points). The higher resolution in the dispersion calculations compared to the wind model (500 m) is justified by the fact that the emission data has much higher spatial resolution (around 10 m) than the wind model. Individual buildings and street canyons are not resolved but treated using a roughness parameter (similar to the treatment used by Gidhagen et al., 2005).

Emission factors for PM10 were obtained from measurements in a street canyon using NOx as tracer (Omstedt et al., 2005). Information on traffic flows, vehicle types etcetera was obtained from the emission inventory of the Regional Air Quality Management Association of Stockholm and Uppsala (Johansson et al., 1999). The inventory includes some 20 000 road links and an annual traffic volume of 12 500 million vehicle km's (data from 2001). The emission factors for particle number concentrations are those suggested by Gidhagen et al. (2005). Chemical and physical transformation processes of particles as well as dry and wet deposition were neglected in the model calculations of annual mean PM10 and number concentrations. See also Gidhagen et al. (2005) regarding the influence of dry deposition and coagulation on particle number concentrations over the urban scale of Stockholm.

5.4.2 Regional modelling

A three dimensional chemistry transport model, the MATCH model (Multi-scale Atmospheric Transport and Chemistry), was used on the European scale. MATCH has previously been used on various spatial scales from 500 m (Gidhagen et al., 2005) to regional scales (e.g. Langner et al., 2005, Engardt et al., 2005, Andersson et al., 2007) to simulate PM, photochemical compounds and deposition of eutrophying and acidifying compounds.

The horizontal model resolution was ca 44 km (0.4°x0.4°). In the vertical 22 layers were used, starting with a near-surface layer of ca 20 m depth and increasing the depth of the layers with height, ending up with the top of the model domain at 5 km. The model contains about 70 chemical components including ozone and its precursors, and nitrogen and sulphur chemistry. The model includes 130 photochemical reactions. Particulate components described in the model are anthropogenic primary emitted particles including elemental carbon (EC), organic matter (OM) and inorganic material (IM) e.g. from road, break and tyre wear. Secondary particulate component include only secondary inorganic aerosol (SIA). Contributions from sulphur emitted naturally from volcanoes and oceans, and sea salt particles are included in the model. Secondary organic aerosol (SOA) and natural dust are not yet implemented in the model. The exclusion of SOA and natural dust means that the model is expected to underestimate total particulate mass concentrations and exposure compared to measurements of PM_{10} and $PM_{2.5}$.

The model was run with meteorology from the European Centre of Medium-range Weather Forcasts (ECMWF). For the years 1997-2000 meteorology from the re-analysis was used. For the years 2001-2003 meteorology from the operational system was used. The horizontal resolution of ERA40 is about 125 km and 40 km for the operational system. The fields were interpolated to the grid used in MATCH. The temporal resolution of 6 hours in the meteorology was interpolated to hourly meteorological fields for use in the model calculations.

Details on the model and model set-up are given in Andersson et al. (2007) and Andersson et al. (2008). EMEP expert emissions of NOx, SOx, NMVOC, CO, NH3 and primary PM extracted from the EMEP home page (Internet URL http://www.emep.int; version 2006-11-21, Vestreng, 2003) extracted for years available[2].

To distinguish between contribution from different regions in Europe the anthropogenic emissions were divided into four main regions and one sector (which was not included in either region). These were Sweden (SE), Denmark-Norway-Finland (DNF), western Europe (WEU) including also Greece, eastern Europe (EEU) and finally international sea traffic (SEA). The resulting emission scenarios of primary $PM_{2.5}$ according to the division in source regions is shown in Figure 10 for the year 2001. Natural emissions, for example emissions from volcanoes and di-methyl-sulphate (DMS) from oceans were only included in the "total" scenario.

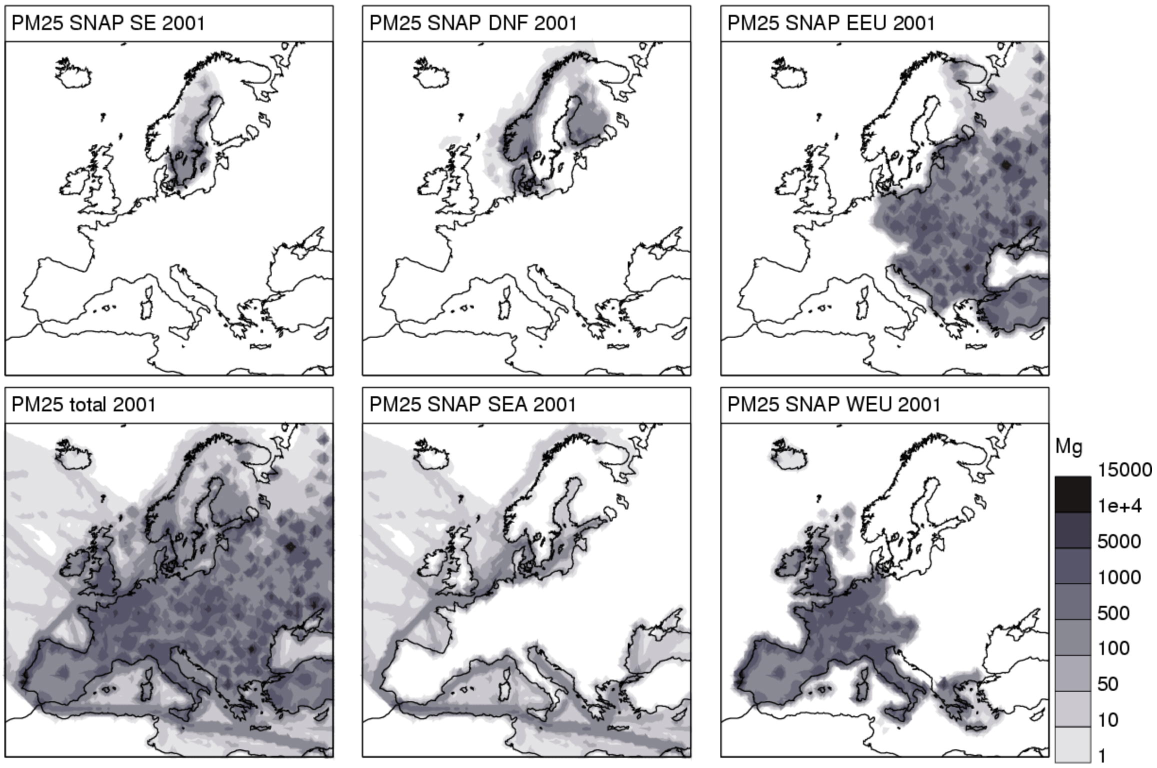

Figure 10. Emissions of fine primary particles (PPM2.5) for the year 2001. Top left is the SE scenario, top middle is DNF scenario, top right is the EEU scenario, bottom right is the WEU scenario, bottom middle the international sea traffic scenario (SEA) and bottom left is the total emissions scenario. Unit: Mg yr^{-1}.

The primary PM scenarios and the photochemistry-scenarios, for calculating the SIA concentrations, were conducted separately. For primary PM the emissions were reduced by 100 % in the corresponding source region and the contribution from one region was therefore the difference between the scenario with total emissions and the scenario with reduced emissions according to

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[2] After quality check of the emission data it was found that some files contained errors important for this study. The erroneous data were replaced by the corresponding data from the nearest available year with acceptable data.

$$c_{contribution} = c_{total} - c_{reduced} \quad .$$

For the SIA scenarios however a 100% reduction in emissions in a certain source region may lead to large non-linear[3] effects. For SIA the emissions were therefore reduced by 20% in the different source regions. A set of calculations were also performed with 10% reduction to check for non-linearities. The result was that for most regions in Europe the long-term average is to a large extent linear (for more details see the Appendix).

6. Measured local and non-local contributions

6.1 PM10 and PM2.5

There are also large scale gradients over Sweden with highest background levels in the south and lowest up north (Areskoug et al., 2001; Forsberg et al., 2005) and similarly but not so clear, some east – west gradients. But these gradients, observed when averaging data over a year or more, are small compared to day to day variations. Table 1 gives an overview of the data available at Aspvreten and Norr Malma. In this study we focus on the period 1998 -2004 and for this period there is no PM10 and PM2.5 data at Norr Malma, only NOx data.

At Norr Malma PM10 and PM2.5 measurements started in June 2005, so from that date there have been parallel measurements at Aspvreten and Norr Malma. These can be used to check the variability of the rural background levels.

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[3] By linear we mean that if we reduce the emissions in one region by 10 % or by 20 % the concentration decrease of the 20 % reduction should be twice as large as the concentration decrease of the 10 % reduction.

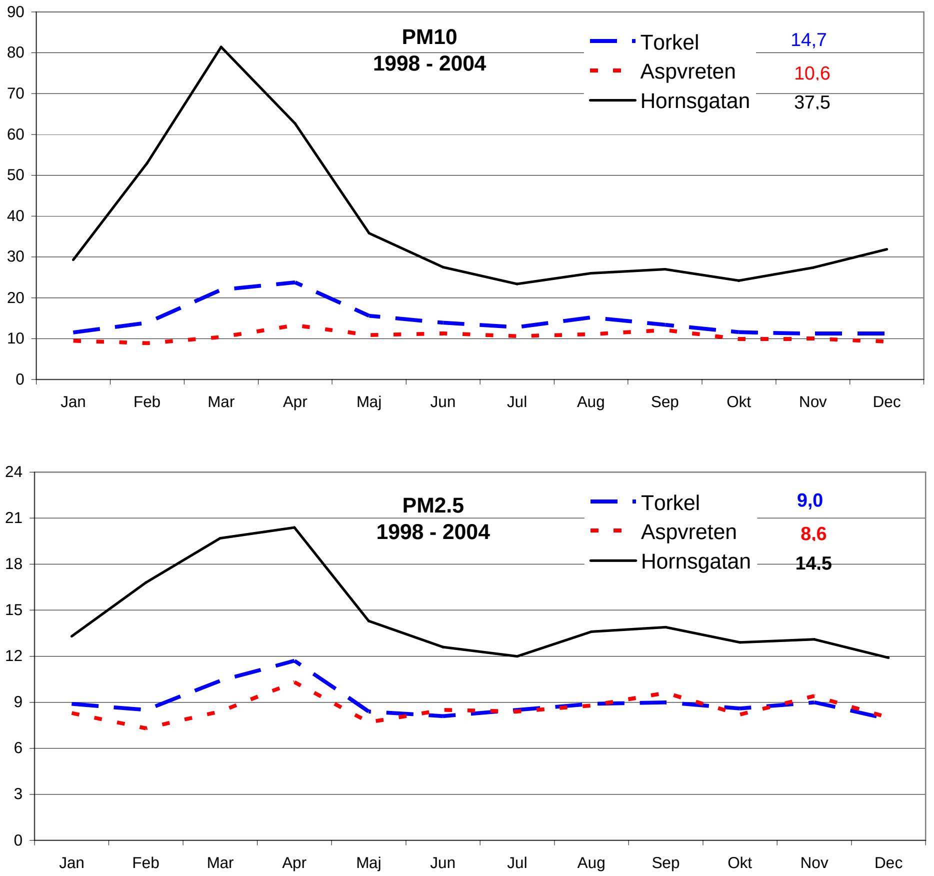
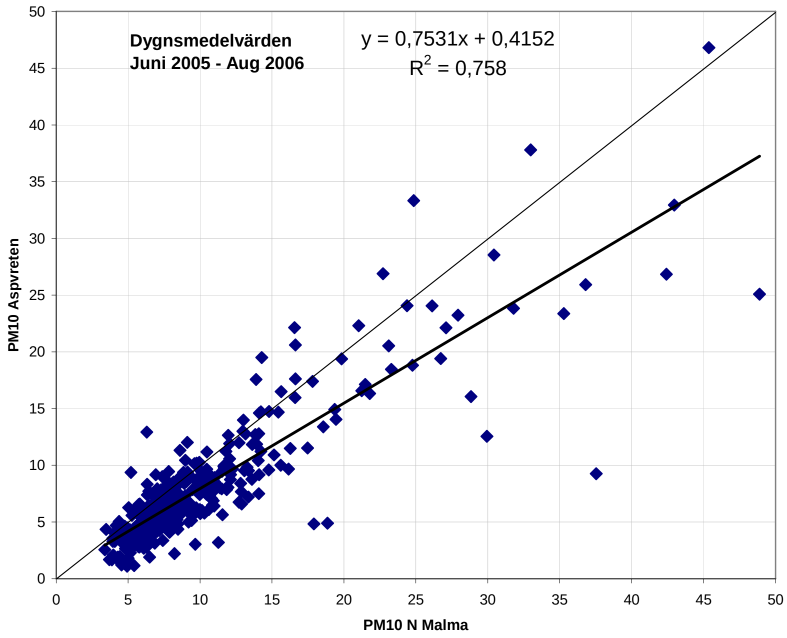


Figure 11. Average monthly variation of PM10 and PM2.5 at Aspvreten , Torkel and Hornsgatan for the period 1998 to 2004.

6.1.1 Comparison of PM10 and PM2.5 measurements at rural sites

During the period June 2005 to August 2006 there are simultaneous PM10 and PM2.5 data from two rural sites and the urban background in Stockholm. It is interesting to see if the two rural sites, located north and south of Stockholm differ significantly. Identical type of instruments have been used at these sites (even though they are of different age).

Indeed there are quite substantial differences in both absolute levels and in diurnal variations observed at these sites (Figure 13). The levels in Stockholm are around 4.5 µg/m^3 higher than at the rural site Norr Malma (see Figure 12), which is due to the local source contributions. But also Norr Malma, north of Stockholm is higher on average compared to Aspvreten and the diurnal variations look quite different, indicating that local sources have some influence at these sites as well. Aspvreten is situated in a forested area, whereas Norr Malma is located in an agricultural area. The exact reasons for the differences observed is not known, but as can be seen in Figure 14 the standard deviations of the hourly mean values are quite large and include zero.



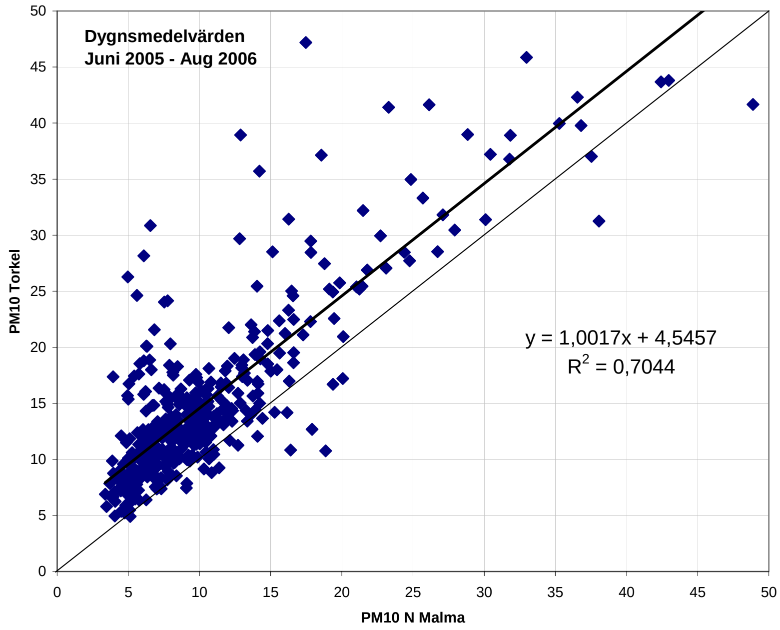


Figure 12. Comparison of daily mean PM10 levels at Norr Malma, Apsvreten and Torkel Knutssonsgatan.

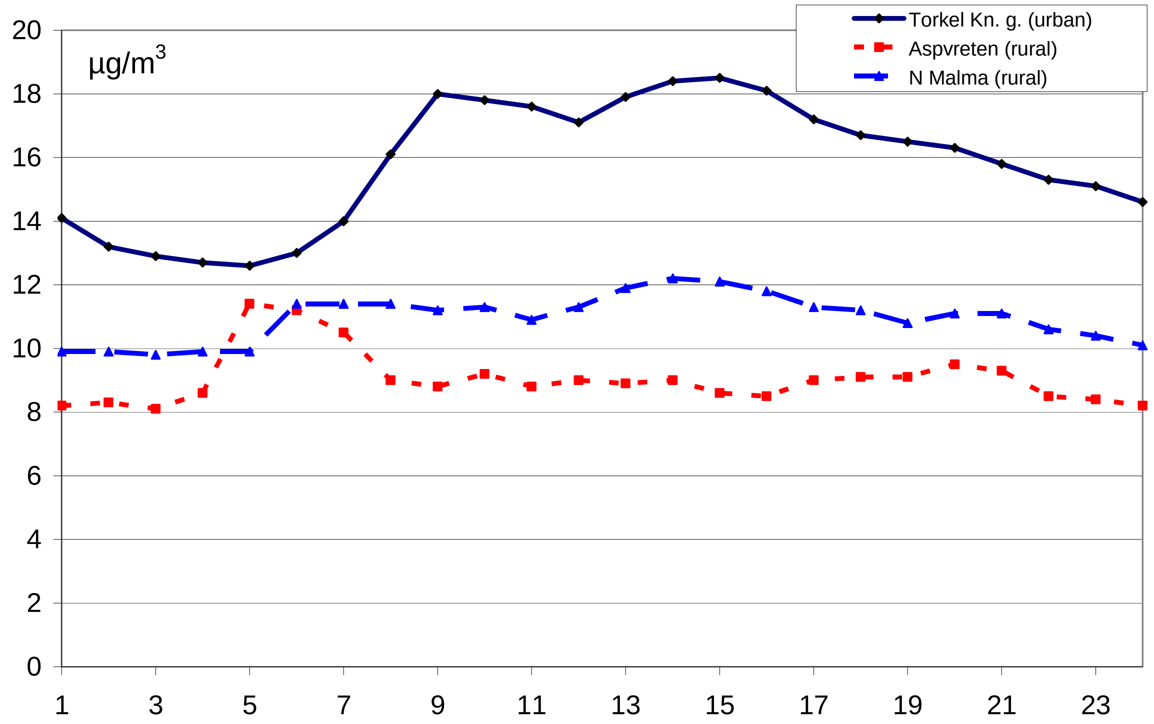


Figure 13. Mean diurnal variation of PM10 in Stockholm (Torkel, urban background) and at two rural background sites (Aspvreten and Norr Malma). Simultaneous data from June 2005 – August 2006 using TEOM instruments. The data have not been corrected.

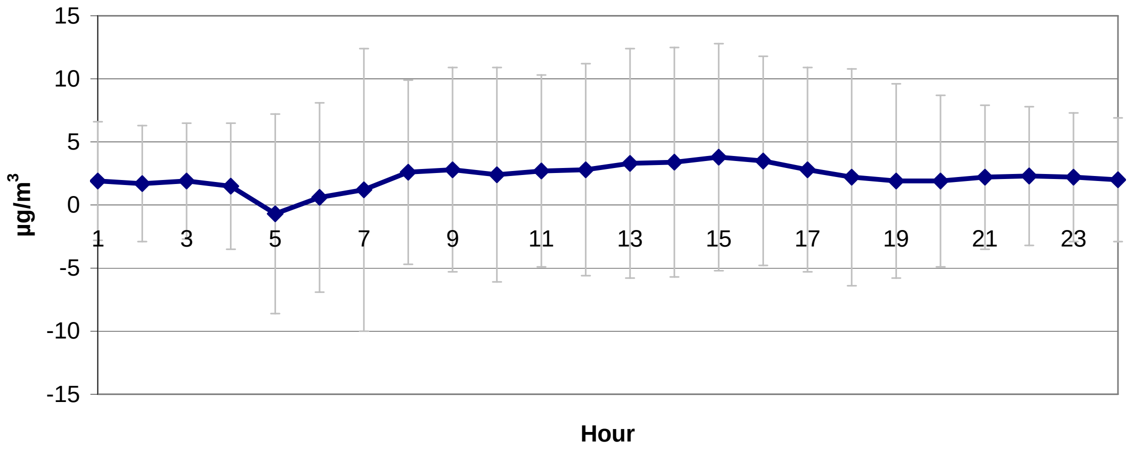


Figure 14. Mean diurnal variation of the difference in PM10 concentration measured at Norr Malma and Aspvreten (June 2005 – Aug 2006). The vertical bars are standard deviations of hourly mean values for each hour (in total 9526 hours).

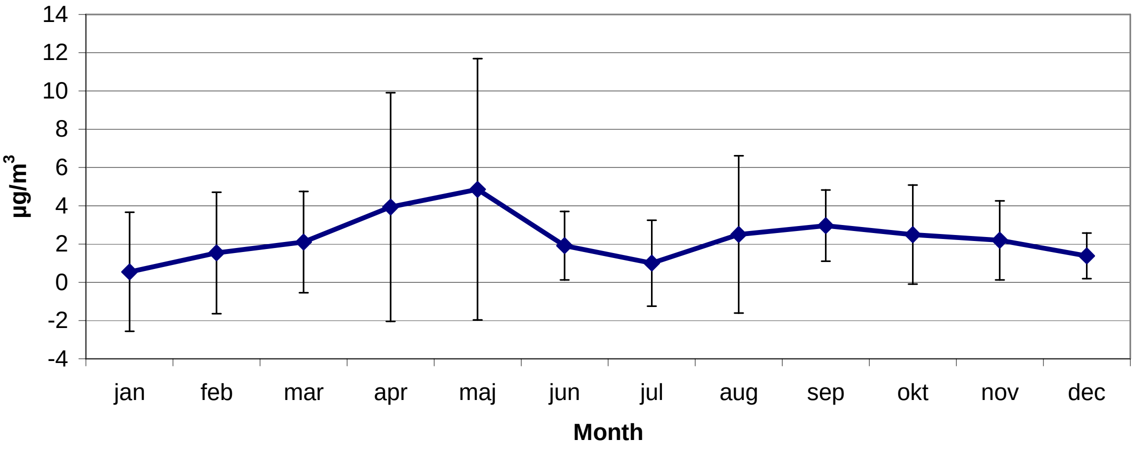


Figure 15. Mean annual variation of the difference in PM10 measured at Norr Malma and Aspvreten (June 2005 – Aug 2006). Vertical bars are standard deviations of the daily mean values.

Using rural measurements to get non-local exposure concentrations assumes that the impact of rural background pollution is the same in Stockholm as at the rural sites located some 70 – 80 km south and north of Stockholm. During most of the time this is true, but there might be regional episodes that are seen in Stockholm, but not transported over Aspvreten and vice versa.

Figure 15 shows the difference in PM10 at Norr Malma and Aspvreten for each month of the year. The smallest differences are seen in summer and winter, and the largest in spring and autumn. Regional episodes are more common in spring and autumn, which might explain some of the deviations seen.

Long range transported PM can sometimes give rise to large horizontal gradients, making estimation of *hourly* mean background concentrations uncertain[4]. An example of this is illustrated in , where estimated PM2.5 concentrations, based on satellite measurements from March 29, 2007, are shown. Unfortunately, there were no PM10 and PM2.5 data at Aspvreten during this period, but a comparison of data from central Stockholm and Norr Malma during the episode is shown in Figure 17.

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[4] For *annual* mean concentrations the gradients of the long-range transport contributions are expected to be evened out making it easier to estimate the yearly average background concentrations than the hourly values.

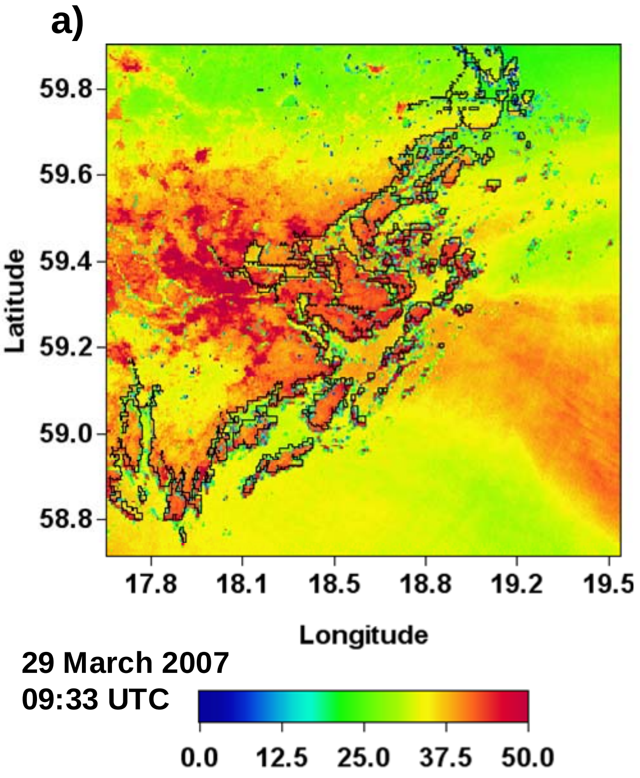

Figure 16. Estimated fields of PM2.5 (300 m horizontal resolution), based on Satellite optical data (MERIS) and an empirical relationship to PM2.5 (from Glantz et al., 2008). Unit: µg/m³.

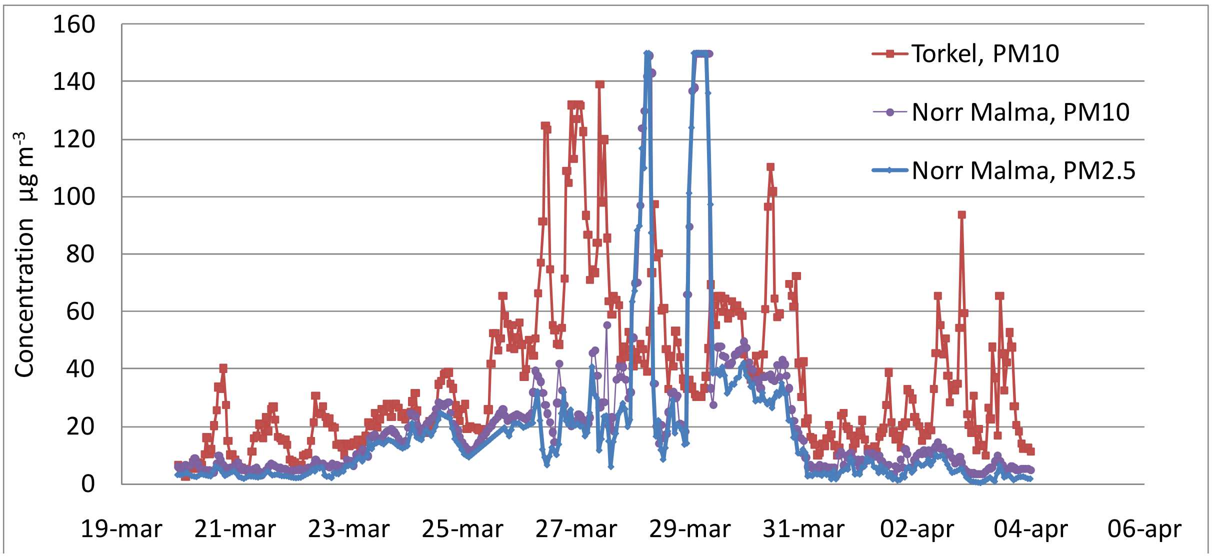

Figure 17. Comparison of PM10 levels at Torkel Knutssonsgatan (Stockholm) and Norr Malma (rural station) during an episode in 2007.

6.2 BC and PM1

There are only very limited data on PM1, but it is clear that the fraction of PM10 that is PM1 varies substantially. During the period April 4 to June 6 (2007) it varied from 10 % to 80 % (Johansson & Hansson, 2007). PM1 is influenced by both non-local and local sources. Local sources are mainly combustion particles, but also mechanically generated particles (like road and brake wear) might be important. 8

shows relative contributions to PM10 of particles with diameters <1 µm, 1-2.5 µm and 2.5-10 µm based on measurements during spring at Aspvreten, Torkel Knutssonsgatan and Hornsgatan. Particles with diameter 1-2.5 µm make a minor contribution (<10%) in the urban area, but quite large contribution (30%) at the rural site Aspvreten. This is due to the increasing importance of local coarse and ultrafine particle sources in the city.

BC is also affected by both local and non-local sources, but, in general, local sources are more important for the BC levels than for PM1. At kerb side sites BC can be 10 to 20 times higher than rural air (see Johansson & Hansson, 2007). Measurements of black smoke (BS) during the winter in many Swedish cities show that the levels can be 2 to 10 times higher than in rural air.

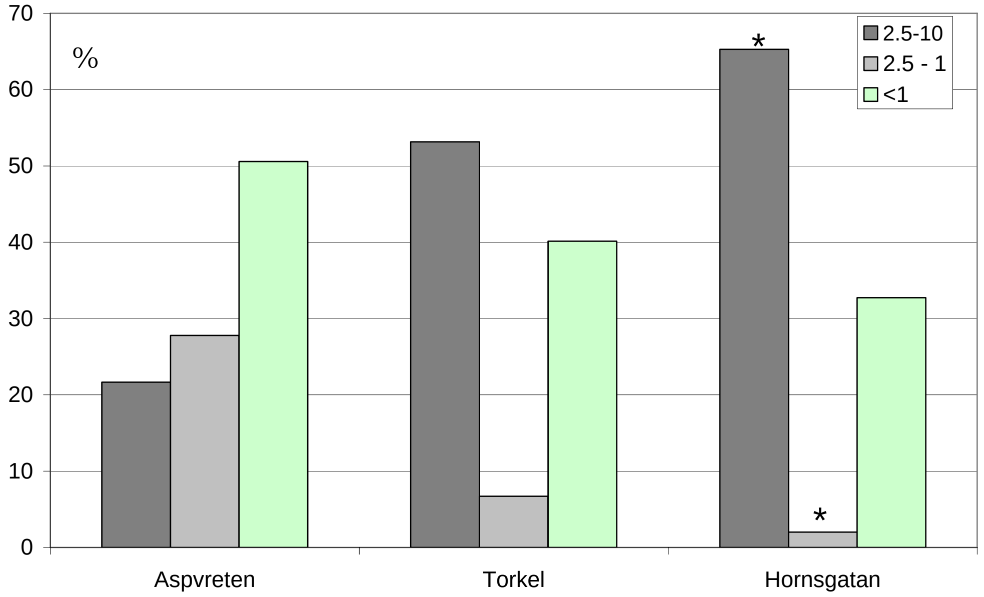

Figure 18. Relative contribution to PM10 of particles with diameters <1 µm, 1-2.5 µm and 2.5-10 µm at Aspvreten, Torkel Knutssonsgatan and Hornsgatan based on measurements during April 3 – May 17, 2006. ”*” = measurements of PM10 and P2.5 during 2003, 2004, 2005 and 2006.

Table 2. Simultaneous measurements of PM1, PM2.5, PM10 and BC at Aspvreten, Torkel Knutssonsgatan and Hornsgatan.

	Aspvreten	Torkel	Hornsgatan	Ratio Torkel/Aspvreten	Ratio Hornsgatan/Torkel
PM1	7.1[1]	10.8[2]	20.0[2]	1.5	1.9
PM2.5	11.0[1]	12.6[2]	-	1.1	-
PM10	14.0[2] (corr.)	26.9[2]	61.1[2]	1.9	2.3
BC	0.4[4]	1.7[3]	5.9[3]	4.3	3.5

[1] IVL filter method (3/4-17/5)
[2] TEOM (3/4-17/5), uncorrected values except for Aspvreten, which has been multiplied by 1.20.
[3] Commercial aethalometer (21/4 – 17/5)
[4] Custom built PSAP, using specific absorption coefficient of 10 m^2/g (21/4 – 17/5)

6.3 Particle number concentrations

Since local exhaust particles comprise a very small fraction of PM10 other measures need to be used to estimate this contribution.

Model calculations in combination with measurements have shown that the total particle number concentration in the city centre is dominated by the contribution from vehicle exhaust (Gidhagen et al., 2003 & 2004). The mean number concentrations in the urban background of Stockholm is 15 000 cm^{-3} and at Aspvreten the level is around 3 000 cm^{-3}.

Figure 19 shows correlation coefficients for different sizes of particle number concentrations at Hornsgatan and Aspvreten. It is seen that particles with diameters 10 – 100 nm at Aspvreten are not strongly correlated with fine particles (20 nm to >200 nm) at Hornsgatan. However, the concentrations of coarser particles (198 nm to >390 nm) at Aspvreten correlate quite well with particles >200 nm at Hornsgatan (>0.70). These particles are of non-local origin, which is also confirmed by looking at the diurnal variations of particles at Hornsgatan. Particles >200 nm show very small or no diurnal variation (Figure 20).

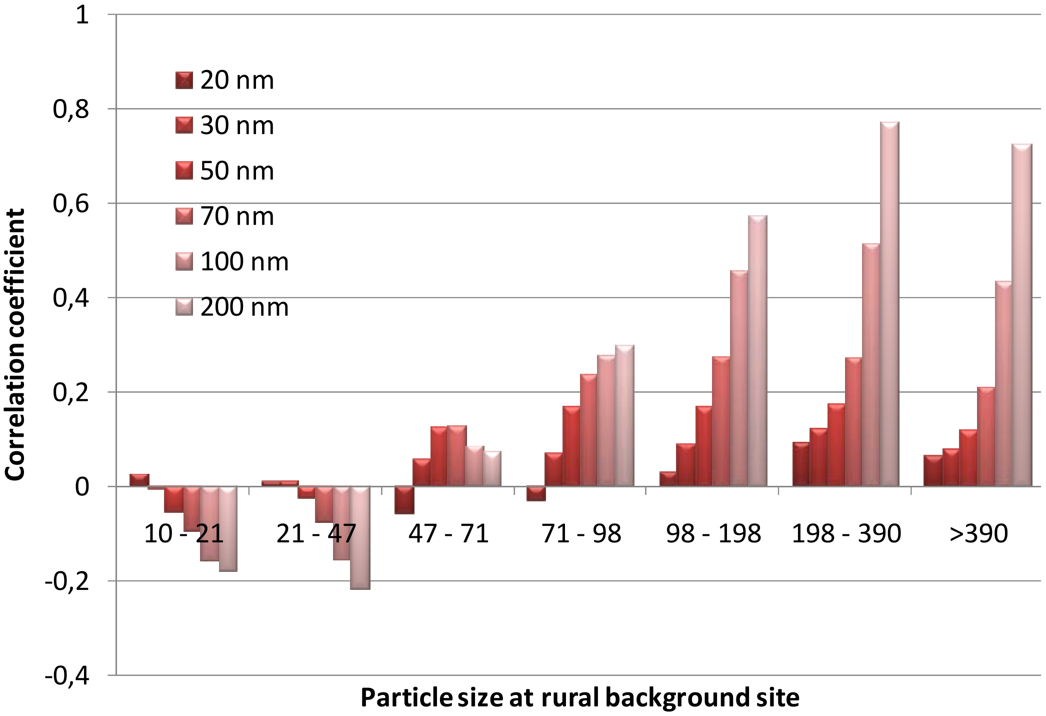

Figure 19. Correlation coefficients for different sizes of particle number concentrations at an urban site in Stockholm (Hornsgatan) and a background site outside the city (Aspvreten). The vertical bars represent the different size fractions (particle diameters) measured at Hornsgatan.

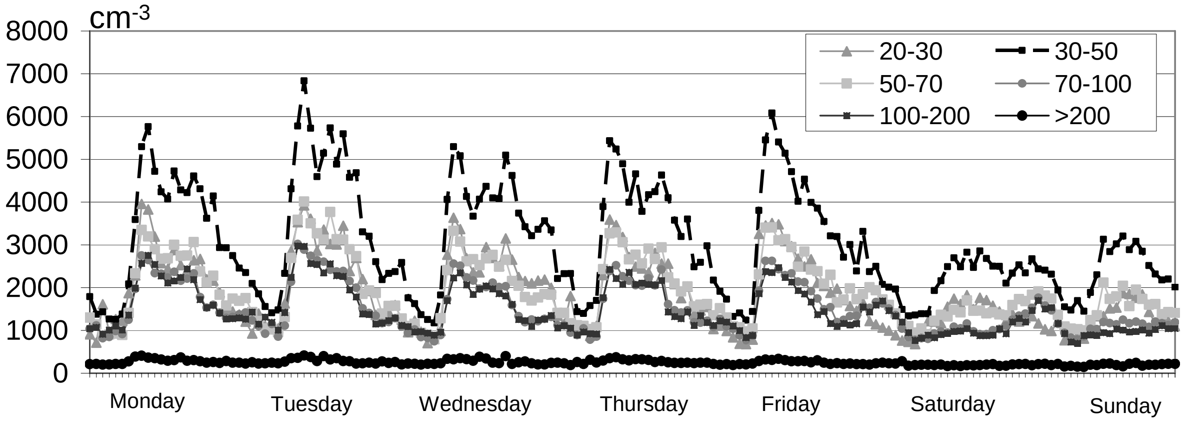

Figure 20. Mean diurnal variations of particle number concentrations of different sizes at Hornsgatan.

Unfortunately, there are no particle number measurements before 2001 in Stockholm. At the rural background site Aspvreten the measurements started in 2000. NOx has been found to be an excellent tracer for vehicle exhaust particles (e.g. Gidhagen et al., 2003), see Figure 21.

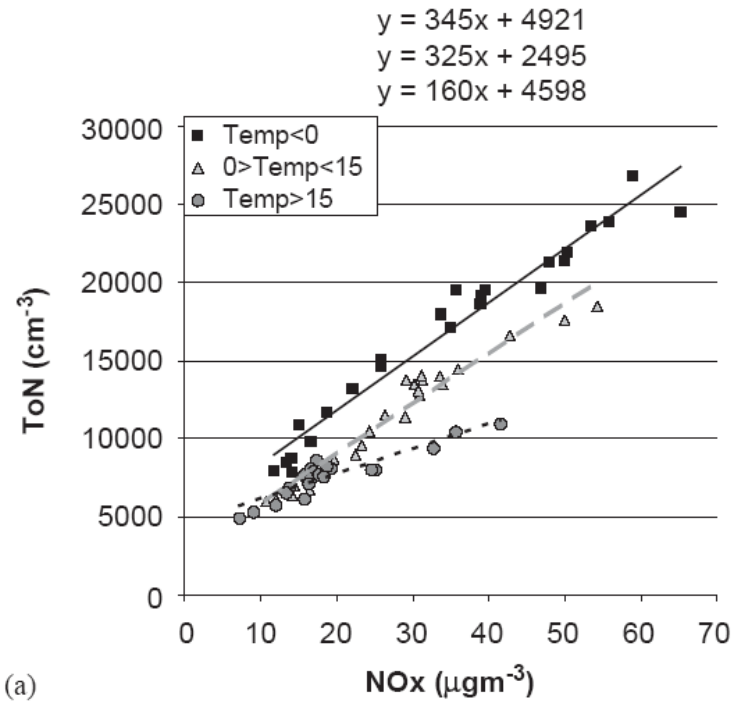

Figure 21. Relation between particle number and NOx concentrations at Hornsgatan, Stockholm (from Gidhagen et al., 2003).

7. Emissions

7.1 Emission factors of PM and NOx

7.1.1 According to the EVA model

According to the EVA model the PM emission factors for the light duty vehicle fleet in the Greater Stockholm area have decreased from 21 mg/vkm to 10 mg/vkm in 1998 to 2006 (Table 3 & Figure 22). For NOx the emission factors have dropped from 941 to 330 mg/vkm during this period. This means that the ratio of PM/NOx has increased from 22 to 31 mg/g for the LDV fleet.

Both the NOx and PM emission factors are much larger for heavy duty vehicles, but the emission factors for PM decreases more rapidly than for NOx; from 217 to 65 mg/vkm for PM and from 9,2 to 5,4 g/vkm for NOx (Figure 24). This means that the ratio for the HDV:s has decreased from 24 to 12 mg/g during 1998 - 2006. The overall exhaust vehicle ratio of PM/NOx has been almost stable or slightly decreased from 23 to 21 mg/g during this period.

There are, however, large uncertainties in the emission factors for PM. Based on measurements in the Söderleden road tunnel (Stockholm) during winter in 1998/1999 the overall emission factor for PM0.6 was estimated to be 40 mg/vkm (Kristensson et al., 2004). For the Söderleden road tunnel EVA gives an emission factor of 18 mg/vkm and 255 mg/vkm for LDV and HDV respectively. In the

Söderleden road tunnel 4,4 % of the vehicles were HDV, then EVA would give an overall emission factor of 29 mg/vkm, which is almost 30 % lower than the measured value. Unfortunately, it is impossible to say whether the main discrepancy is due to HDV or LDV emission factors.

In addition, the LDV fleet consist of around 5% diesel vehicles which have much higher emissions of PM compared to gasoline vehicles. According to EVA light duty diesel vehicles emitted almost 25 times more than gasoline vehicles in 1998. In the ExternE (Johansson & Ek, 2003) calculations for year 2000, the emission factors were higher by more than a factor of 2 for LDV gasoline vehicles than EVA emissions but 20 % higher for diesel LDV. For HDV, ExternE used a mean emission factor of around 500 mg/vkm for PM, whereas EVA gives 164 mg/vkm for Greater Stockholm (that is, a factor of 3 difference).

Thus, it seems as if the EVA model gives quite low emission factors of PM for diesel vehicles. Since diesel vehicles are only a relatively small fraction of all vehicles in Stockholm this may not be very important presently. As the vehicle fleet composition is changing it may, however, becomes more important, and it introduces uncertainties in the time trend of the ratio of PM/NOx.

Table 3. NOx and PM(exhaust) emission factors for Greater Stockholm area 1998 – 2004 according to the EVA emission model.

Year	Parameter	LDV	HDV	Total
1998	NOx (mg/veh km)	941	9177	1454
	PMexhaust (mg/veh km)	21	217	33
	PMexhaust/NOx (mg/g)	*22*	*24*	*23*
1999	NOx (mg/veh km)	833	8686	1322
	PMexhaust (mg/veh km)	19	191	30
	PMexhaust/NOx (mg/g)	*23*	*22*	*23*
2000	NOx (mg/veh km)	725	8194	1191
	PMexhaust (mg/veh km)	18	164	27
	PMexhaust/NOx (mg/g)	*24*	*20*	*23*
2002	NOx (mg/veh km)	547	7022	950
	PMexhaust (mg/veh km)	14	118	21
	PMexhaust/NOx (mg/g)	*26*	*17*	*22*
2003	NOx (mg/veh km)	500	6545	877
	PMexhaust (mg/veh km)	13	99	19
	PMexhaust/NOx (mg/g)	27	15	21
2004	NOx (mg/veh km)	462	6290	825
	PMexhaust (mg/veh km)	12	88	17
	PMexhaust/NOx (mg/g)	*27*	*14*	*21*
2005	NOx (mg/veh km)	403	5736	735
	PMexhaust (mg/veh km)	11	76	15
	PMexhaust/NOx (mg/g)	*28*	*13*	*21*
2006	NOx (mg/veh km)	330	5400	646
	PMexhaust (mg/veh km)	10	65	14
	PMexhaust/NOx (mg/g)	*31*	*12*	*21*

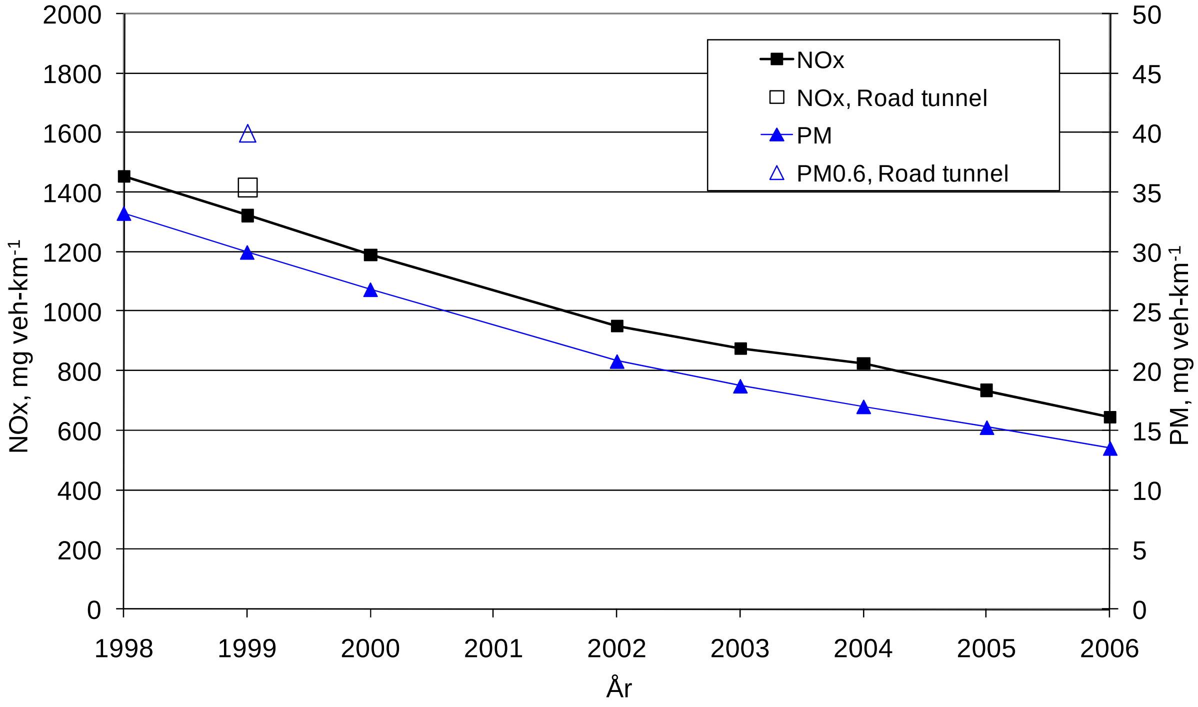

Figure 22. Total exhaust road traffic emission factors of NOx and PM for Greater Stockholm according to the EVA-model. Also shown are emission factors estimated from road tunnel measurements in Stockholm (Kristensson et al. (2004).

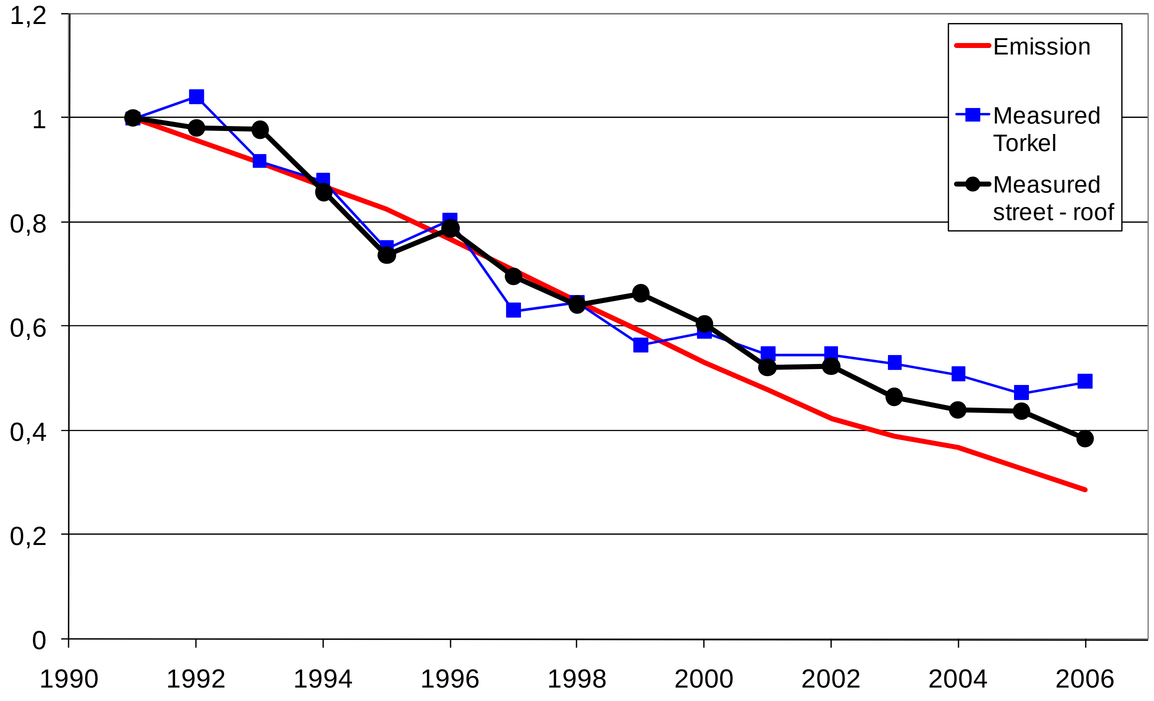

Figure 23. Comparison of the trends in emissions and the trends in the concentrations. Emissions are for total road traffic in Greater Stockholm area. Measured values are concentrations of NOx at Torkel Knutssonsgatan (urban background) in Stockholm minus Norr Malma (rural background). Also shown is the change in the concentration difference between Hornsgatan (street) and Torkel Knutssonsgatan (roof = urban background).

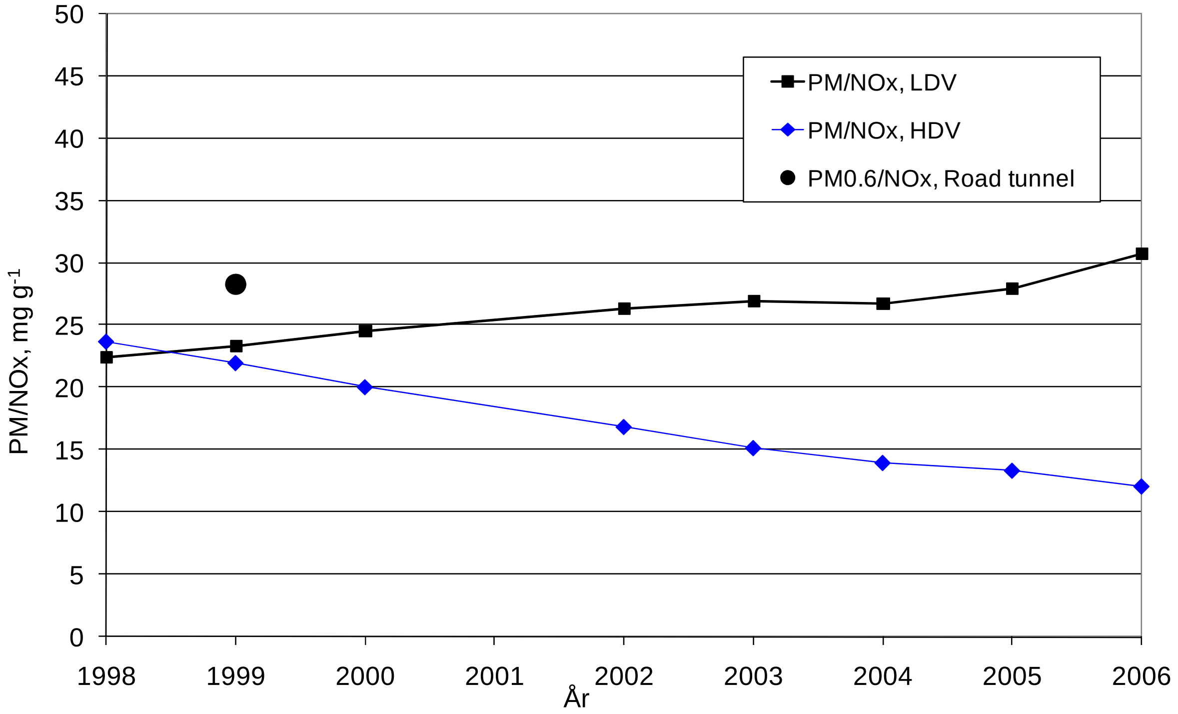

Figure 24. Relation between PM and NOx according to the EVA-model for the Greater Stockholm area. Also shown is the ratio of PM0.6 to NOx according to measurements in a road tunnel in Stockholm (from Kristensson et a., 2004).

7.1.2 PM exhaust to NOx according to measurements

For NOx, simultaneous measurements in a street canyon, urban background and rural background gives information on the importance of the local sources. The difference between the street and the roof top concentrations can be regarded as proportional to the emissions. The ratio of the concentrations differences of any substance to NOx is a measure of the emission factor if the emission factor for NOx is known.

For exhaust PM there are not so many measurements that can be used. PM10 and PM2.5 include a large contribution from non-exhaust particles. Even if the urban background is subtracted, neither PM10, PM2.5 or PM1 can be used as measures of PM exhaust, since these PM measures include particulate material from the wear of the roads, brakes and tires. Instead we can use black carbon or particle number concentrations. Black carbon is a measure of the blackness of the particles (light absorption properties).

7.2 Emission factors for BC

Light and heavy duty vehicle exhaust emissions is the most important source of submicron particles in urban areas. Generally, emission factors for diesel vehicles are much higher than for gasoline vehicles. The mean emission factors for the vehicles have decreased as exhaust after treatment has been introduced (Kupiainen & Klimont, 2004 och 2007).

Diesel particles consist mainly of organic and elemental carbon. Generally, the BC/OC ratio is higher for diesel exhaust than for gasoline. Catalysts are more efficient in reducing OC than BC. As shown in Johansson et al., 2006, the emission factors for BC from HDV, without exhaust after treatment, are in the range 45 - 150 mg/vkm. With after treatment the emission factors are reduced to 9-18 mg/vkm. For light duty diesel vehicles without after treatment emission factors are in the range 60-300 mg/vkm and for light duty gasoline vehicles with three way catalyst they are in the range 2-5 mg/fkm. Since 2006 black carbon (BC) has been measured at Hornsgatan and Torkel Knutssonsgatan using continuous Aethalometers[5] connected to the PM10 and PM2.5 measurements. The difference in concentrations between street and roof is an indication of the exhaust emission of BC. Figure 25 shows the ratio of the BC ad NOx concentration due to traffic emissions at Hornsgatan (measured as the street minus roof concentration). On average the ratio has decreased from ca 45 mg/g to 15 mg/g, which indicates that the exhaust particle emissions have been reduced more than NOx emissions during the last years.

Measurements along the highway from Stockholm to Arlanda has shown that BC is highly correlated with NOx (Johansson et al., 2004). In these measurements (from 2003) the mean ratio of BC to NOx was around 12 mg/g, somewhat lower than for Hornsgatan, which may be due to different driving conditions on the highway. The BC/NOx ratios may be compared with the ratios of PM/NOx according to the EVA model. The PM exhaust/NOx ratio for 2006 is 30 mg/g for Hornsgatan, which is slightly lower compared to the measured BC/NOx as seen in Figure 25. But since BC is not exactly the same as PM this is not surprising. It is also interesting to note that the BC/NOx ratio has been decreasing; from 45 mg/g in April 2006 to 15 mg/g in February 2008.

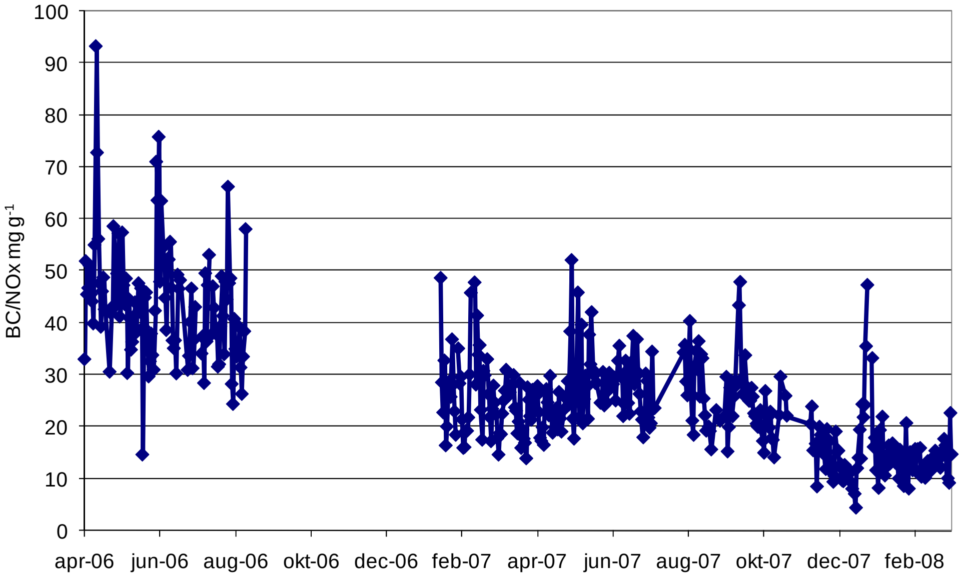

Figure 25. Measured ratio of BC (street-roof) to NOx (street-roof) concentrations for Hornsgatan 2006-2008 (mg/g). Daily mean values.

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[5] The Aethalometer measures the absorption of visible light through a filter onto which particles has been sampled. The mass concentration is obtained by assuming a constant specific absorption coefficient of 16.6 m^2/g.

7.3 Particle number emission factors

A review of field and laboratory measurements of emission factors for particle number is in preparation (Johansson et al., 2008). Emission factors based on roadside studies are available from Sweden (Gidhagen et al. 2004a, 2004b; Mårtensson et al., 2006), USA (Zhang et al. 2005; Kittelson et al. 2004), Great Britain (Corsmeier et al. 2005; Jones & Harrison, 2006), Switzerland (Imhof et al. 2005), Australia (Morawska. et al 2005, 2001; Gramotnev et al. 2003; Jamriska & Morawska 2001), and Denmark (Ketzel et al. 2003). All these studies measured or estimated the EFs for vehicle fleets in the range of 0.57-12.6×10^{14} particles veh^{-1} km^{-1}, with a median average value 2.8×10^{14} particles veh^{-1} km^{-1}. It is also found that the median EFs of HDVs are more than an order of magnitude higher than EFs of LDVs. However, the emission values from the individual studies depend on the vehicle fleet composition (share of diesel vehicles), traffic speed, measured particle size range and measurement conditions.

It is also important to consider when the measurements were made since the vehicle fleet is changing quite rapidly. In Stockholm, measurements show that the ratio of particle number to NOx is decreasing (Figure 26). In 2001 the ratio was around 500 particles/cm^3 per µg/m^3 and in 2007 the ratio was less than 200 particles/cm3 per µg/m^3. So the measurements of both BC and particle number concentrations at Hornsgatan indicate that vehicle exhaust emissions of particles are decreasing, but this is not consistent with emission factors according to the EVA model as shown earlier in Figure 24. For LDV the PM/NOx emission ratio is increasing and for HDV it is decreasing but overall the emission ratio has been about the same since 1998 (see Table 10). At least for long term estimates of exposure due to vehicle exhaust particles using NOx as indicator the trends in the ratio of exhaust particle/NOx need to be considered. For short term exposure (day to day variations within a year or two) NOx is a good indicator for vehicle exhaust particle exposure.

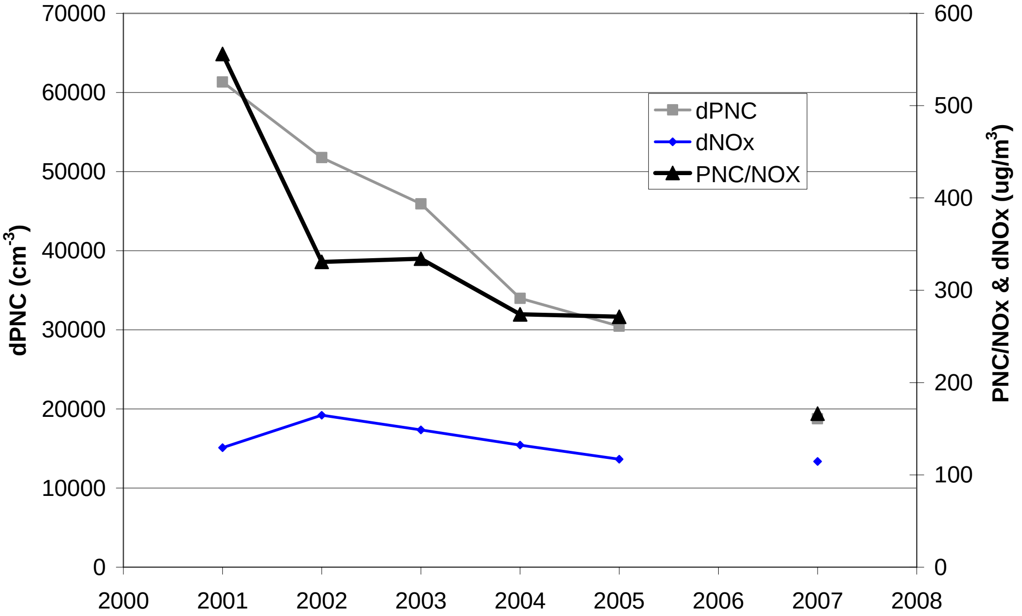


Figure 26. Annual mean values of NOx (street-roof), PNC (street-roof) and the ratio of PNC/NOx at Hornsgatan. Roof data are from Rosenlundsgatan and Torkel Knutssongatan.

7.4 Total emissions in Greater Stockholm

Total PM emissions from LDV:s are about twice as large as from HDV:s in the Greater Stockholm area. Light duty diesel cars account for around 40% of the total LDV emissions. As shown in Table 5 the emission factor for HDV:s is more than 7 times higher than for LDV:s.

Other (than vehicle exhaust) large sources of combustion particles are power plant emissions and residential heating due to combustion of biomass and fossil fuels. The main part (70%) of the emission of combustion particles from residential heating is due to biomass combustion.

For off-road machinery only combustion particles are included. Road and building construction and maintenance work may generate large amounts of non-combustion particles, but this has not been included.

The total NOx concentration in Stockholm is due to several sources:

- Vehicle exhaust (mainly light duty vehicles running on gasoline)
- Off road machinery (diesel engines)
- Ferries and other sea traffic
- Heat and energy production

In addition some NOx is due to non-local sources. The background concentration of NOx is about 5 times lower than the urban background level. This is similar to the relation for particle number concentration.

Table 4 shows the total emissions in 2003 of NOx and particles from road and sea traffic, power plants, residential heating and off-road (working) machinery in the greater Stockholm area. For NOx LDV and HDV emissions from road traffic are the largest sources. For combustion particles residential heating due to biomass combustion is higher than vehicle exhaust emissions, but there is a very large uncertainty in this estimate. The most recent estimates of residential biomass combustion emissions in Stockholm in the frame of the national SMED consortium gives a factor 5 lower emissions.

Table 4. Total emissions (tonnes/year) of NOx and particles from road traffic and other sources in Greater Stockholm (35x35 km^2) area during 2003. Vehicle exhaust emissions are based ion emission factors from the EVA model. Road, brake and tire wear emission factors are based on local measurements in Stockholm according to Johansson et al. (2004).

	Road traffic	**Sea traffic**	**Power plants**	**Residential heating**	**Off-road vehicles**	**Sum**
NOx LDV	*3133*					
NOx HDV	*2749*					
NOx Total	**5882**	**872**	**2002**	**485**	**1903**	**11144**
Exhaust LDV	*84*					
Exhaust HDV	*41*					
Total emission of combustion particles	126	33	249	486	110[b]	1003
Road, brake and tyre wear[a]	1941					
PM10 Total	**2067**	**33**	**249**	**486**	**110**	**2944**

[a] Only non-combustion particles with a diameter <10 um. The size is mainly larger than 1 um.
[b] Only combustion particles due to the use of diesel fuel.

Table 5 shows that the ratio of PM to NOx varies from 15 to 1000 mg PM per g NOx, depending on the source considered. For residential biomass combustion much more PM relative to NOx is emitted (1000 mg PM/g NOx) as compared to vehicle exhaust (around 20 mg/g). According to this database the ratio is higher for light duty vehicles (27 mg/g) as compared to heavy duty vehicles (15 mg/g). The overall ratio for all sources is 90 mg PM/g NOx. This will be further discussed below.

Table 5. Total annual emissions of NOx and combustion particles (PM) and the ratio of NOx to PM for the whole greater Stockholm area.

	NOx (tonnes/y)	PM (tonnes/y)	Ratio PM/NOx (mg/g)
Vehicle exhaust total	5882	126	21
Only LDV	*3133*	*84*	*27*
Only HDV	*2750*	*41*	*15*
Sea traffic	872	33	38
Power plants	2002	249	124
Residential heating	485	486[a]	1000
Off-road machinery	1903	110	58
SUM of alla	11 144	1 003	90

[a] Wood 340 t/y; Oil 146 t/y.

Figure 27 shows that road traffic dominate the emissions of particles measured as PM10 and NOx, whereas for combustion particles residential heating using biomass is responsible for the dominating emission. As pointed out before there are large uncertainties in the emissions from biomass burning. Combustion particles are part of PM10 but PM10 also include non-combustion particles. In this case non-combustion particles come from road traffic due to mechanical wear of road, brakes and tyres. Road wear is the dominating contributor, but the contributions from brakes and tyres are somewhat uncertain (Johansson et al., 2004).

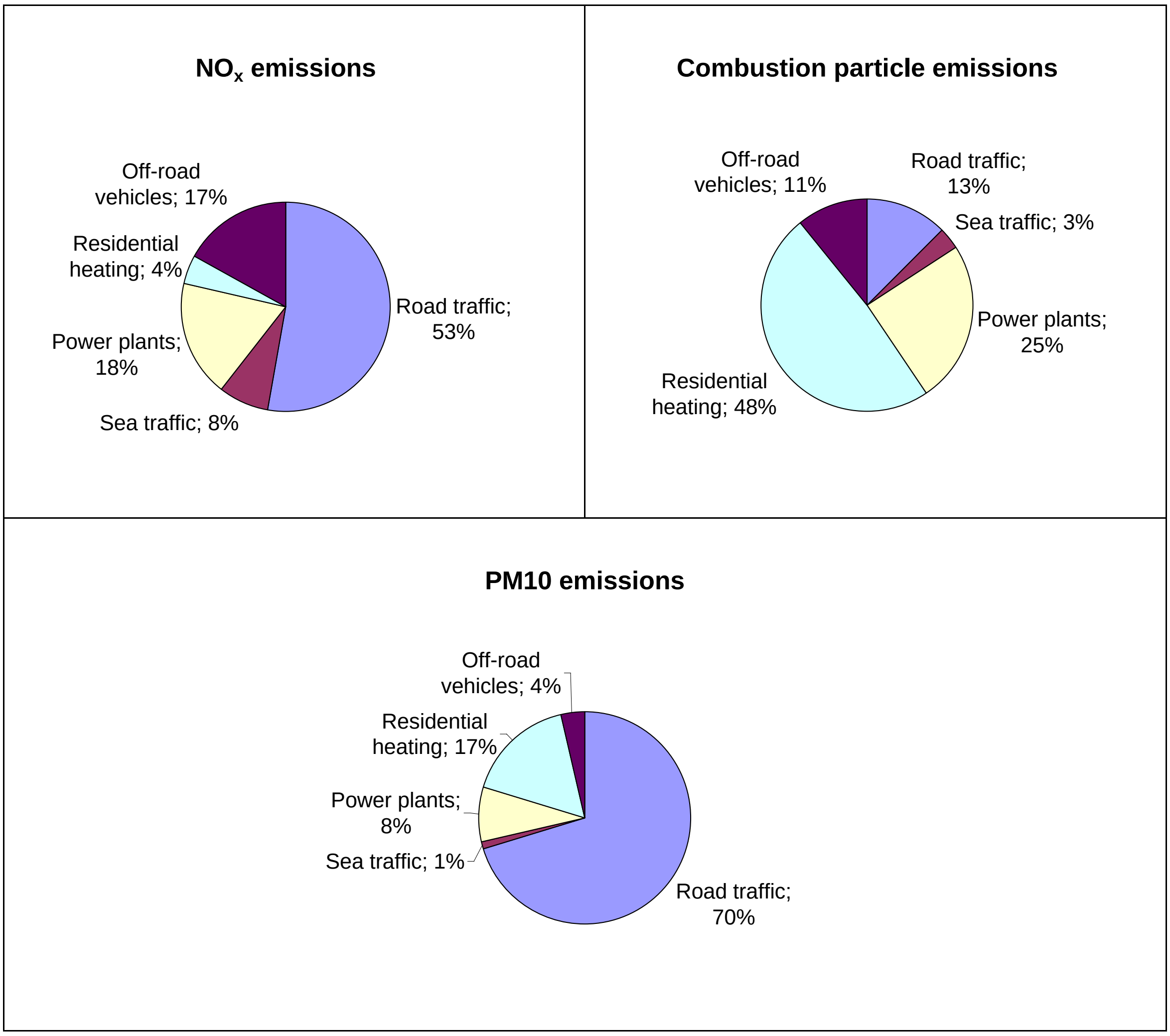


Figure 27. Percentage contribution of emissions of NO_x, combustion particles and PM10 from different source sectors in the Greater Stockholm area.

7.5 Total Emissions in Europe

In Table 6 the total emissions in Europe are displayed for different sectors. These are based on emission inventories collected by EMEP (Vestreng, 2003) and were used (in gridded form) in the model simulations. The division of PPM2.5 into EC, OM and IM2.5 is also detailed in Table 6. Table 7 shows the total emissions and the distribution of the emissions between the different source-regions.

Table 6. Emissions from different sectors in the model domain. Values are based on EMEP expert emissions for the year 2001. Emissions of NO_x are given in equivalents of NO_2, in the model distributed at the ratio 5% NO to 95% NO_2. Emissions of SO_x are given in equivalents of SO_2, in the model distributed between 95% SO_2 and 5% sulphate (the first 10 sectors: CIS to AGR, for the last sector 88% is SO_2). The original primary PM2.5 emissions are apportioned among EC, OM and $IM_{2.5}$ (see text). Unit: Tg yr^{-1}.

	EC	**OM**	**$IM_{2.5}$**	**PPM** coarse	**CO**	**NH_3**	**NM-VOC**	**NO_x**	**SO_x**
CIS	0.0325	0.0650	0.173	0.33	0.75	0.01	0.12	3.32	9.89
CNS	0.161	0.321	0.321	0.28	10.85	0.01	1.13	1.01	1.46
CMS	0.0376	0.0752	0.176	0.19	4.82	0.01	0.18	2.04	2.35
PPS	0	0	0.539	0.32	3.45	0.11	1.42	0.36	0.78
EDE	0.0224	0.00596	0	0.06	0.18	0.01	1.12	0.03	0.07
SOU	0	0	0.0218	0.01	0.02	0.01	4.41	0	0
RTR	0.176	0.199	0	0.09	26.55	0.08	4.57	6.66	0.38
OMM	0.215	0.223	0	0.05	3.03	0.00	0.75	5.52	2.62
WTS	3.33E-4	6.66E-4	0.0890	0.02	1.77	0.13	0.15	0.05	0.05
AGR	0.0215	0.0429	0.101	0.32	0.41	5.45	0.51	0.25	0
OSS	0	0	0	0	0	0	0	0	2.49

CIS = Combustion industry (energy & transformation), CNS = Non-industrial combustion plants, CMS = Combustion in manufacturing industry, PPS = Production processes, EDE = Extraction and distribution of fossil fuels and geothermal energy, SOU = Solvent use & other product use, RTR = Road transport, OMM = Other mobile sources and machinery (including ship emissions), WTS = Waste treatment and disposal, AGR = Agriculture, OSS = Other sources and sinks.

For the particulate matter it may be noted that:

- EC and OM originate mainly from non-industrial combustion plants, road transport and shipping (termed other mobile sources and machinery in Table 6),
- For the non-transport sectors the OM part of the $PPM_{2.5}$ emissions is twice as large as the EC part; for the transport sectors RTR and OMM the fractions are approximately equal,
- $IM_{2.5}$ is mainly due to stationary sources; the largest contribution comes from the production processes (PPS) and the second largest from non-industrial combustion plants (CNS),
- for $PPM_{2.5}$ as a whole the main sources are stationary and transport sectors,
- Coarse primary particulate matter (PPM_{coarse}) also originate mainly from stationary sources, but agriculture is also an important source.

The total emission of $PPM_{2.5}$ in the model domain was 3.02 Tg yr^{-1} for the year 2001. The emission inventory and the sector distribution of emissions used should be regarded with some care, especially when comparing different source sectors for particulate matter, since there may be deficiencies in how and what various countries report.

For CO the largest source is road transport, but the contributions from stationary sources (mainly the CNS, CMS and PPS sectors) are also considerable. The total CO emission in the model domain is 51.8 Tg yr^{-1}.

NMVOC mainly originates from solvent and other product use (SOU) and from road transport (RTR). The total emissions add up to 14.4 Tg carbon yr^{-1}.

Agriculture is the largest contributor by far to NH_3. The total emission in the model domain is 5.8 Tg yr^{-1}.

The largest sources of NO_x are road transport and other mobile sources and machinery (sectors RTR and OMM). However, there are significant contributions from combustion at stationary sources (CIS, CNS and CMS) also. The total NO_x emission in the model domain is 19.2 Tg yr^{-1}.

Sulphur emissions mainly originates from combustion industry according to this inventory. Other stationary combustion processes (CNS, CMS), and other sources and machinery (OMM, consisting mainly of sea traffic emissions) are also large contributors. The total SO_x emissions in Table 7 include the natural emissions from DMS (from seas and oceans) and volcanoes (OSS, other sources and sinks). These are included in the given total, which is 20.1 Tg yr^{-1}.

Table 7. Total emissions in the model domain and distribution between the regions. Values are based on EMEP emissions (see text) for the year 2001. Emissions of NO_x are given in equivalents of NO_2. Emissions of SO_x are given in equivalents of SO_2. SE stands for Swedish contribution, DNF for contribution from Denmark-Norway-Finland, WEU for western European contribution and EEU for eastern European contribution. The definitions of the regions are illustrated in Figure 10.

	Total (Tg yr^{-1})	SE (%)	DNF (%)	WEU (%)	EEU (%)	SEA (%)	Other (%)
PPM2.5	3.02	1.5	3.9	37.4	49.1	8.0	0.0
PPM_{coarse}	1.67	1.3	1.8	36.3	59.7	0.8	0.0
CO	51.8	1.3	3.4	52.2	41.8	0.6	0.7
NH3	5.80	1.0	2.8	53.0	41.2	0.0	2.1
NOx	19.2	1.1	3.2	47.5	31.1	16.5	0.6
NMVOC	14.4	1.9	4.6	55.9	36.1	0.7	0.7
SOx	20.1	0.3	0.7	27.7	45.8	11.0	14.5

The Swedish contribution to the European emissions is between 1 and 2% of the anthropogenic emissions (see Table 7) depending on emitted compound. The emissions in eastern Europe exceed those of western Europe for primary particles and sulphur emissions.

For nitrogen emissions (NO_x and NH_3) and carbon species (CO, NMVOC) the emissions in western Europe exceed those of eastern Europe. This is probably due to the difference in activity in the two regions, having more industrial activity and a more efficient agriculture industry in western Europe, whereas eastern Europe still uses fuel not as clean and the cleaning of flue-gases is not as efficient. Note the contribution of international sea traffic to the total European (model domain) emissions, which for some components is quite large: 8% for fine particulate matter, 16.5% for NO_x and 11% for SO_x.

Note also that since different countries may have different quality of the information on which they base their emission estimates and also to which sector they report it, this might bias the comparison between eastern and western European emissions. This makes it difficult to compare the sector contributions of the different regions. The emissions are to be regarded as preliminary, and only give an indication of possible differences between different source regions. Especially for particulate matter there may be large uncertainties in emissions for some countries. The magnitudes of the sea traffic emissions are also uncertain.

The emission of natural carbonaceous emissions as isoprene and natural sea salt is calculated by the model, whereas diurnal and seasonal distribution of the anthropogenic emissions are provided by EMEP.

8. Model calculations

8.1 Local exposure

8.1.1 Comparison between modelled and measured NOx

In this section modelled and measured NOx levels are compared. Calculations have been done for 8 years 1998 -2005. Since the calculated concentrations are due only to local emissions the rural background concentration of NOx (from Norr Malma) has been added to the modelled values. Figure 28 shows yearly mean values of measured NOx at roof level and calculated local contributions plus rural background. The model overestimates the NOx concentrations during 1998 – 2003 and underestimates the levels for 2004 and 2005. Best absolute agreement is seen for 2003 – 2004. Discrepancies are mainly due to wrong emissions (emission factors for vehicle exhaust).

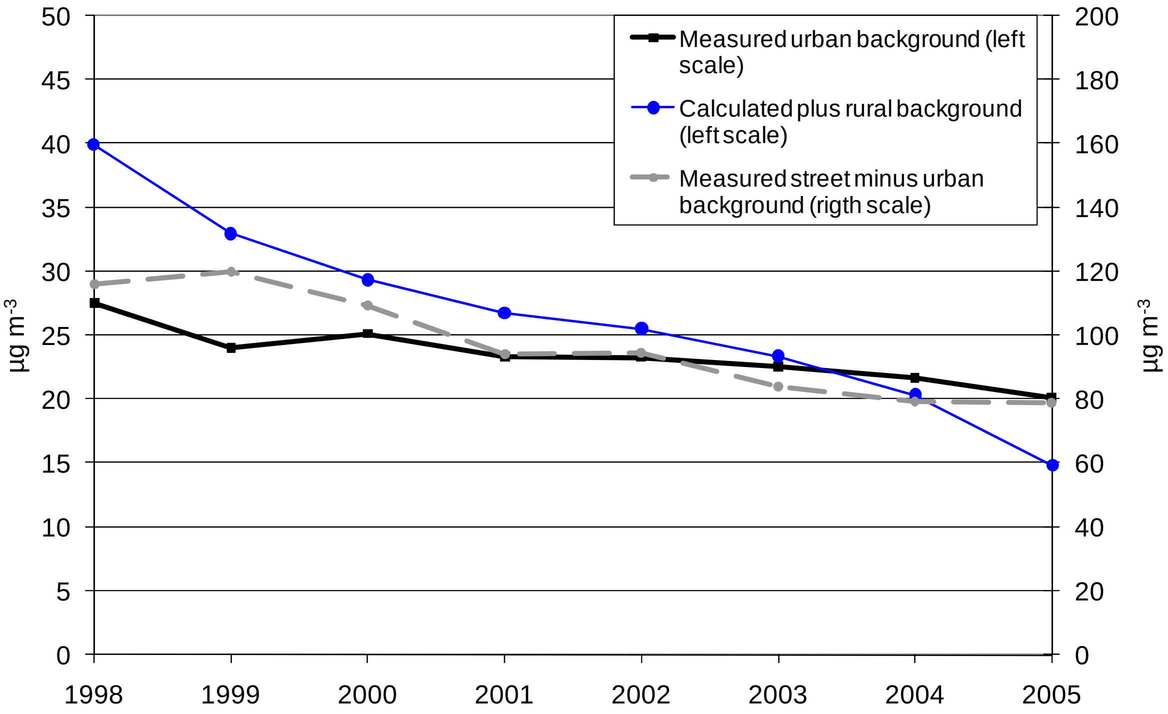


Figure 28. Comparison of yearly average modelled and measured NOx levels (at roof top) in central Stockholm. Grey is street level minus roof top.

For health assessment studies, where daily variations in air pollutants are correlated with different health outcomes, it is not so much the absolute accuracy in exposure, but the temporal variations that are important. Figure 29 shows the calculated and measured time series of daily mean concentrations of NOx for 2004. Measurements are urban background from Torkel Knutssonsgatan in central Stockholm. The model is able to capture most of the variations; the correlation for this year (2004) is 0.73. Table 8 summarizes the comparisons of modelled and measured daily mean concentrations of NOx for all years. The correlation coefficients varies between 0.73 and 0.80.

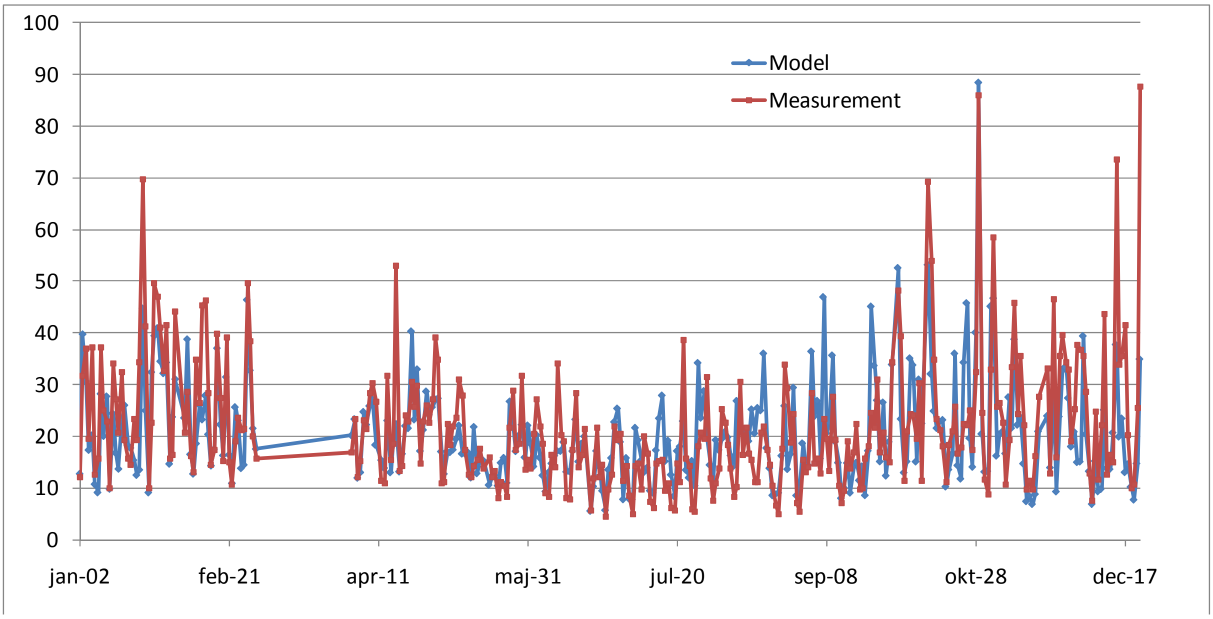


Figure 29. Comparison between modelled and measured NOx at an urban background site in Stockholm (Torkel Knutssonsgatan). Daily mean values for 2004. Unit: µg/m^3

Table 8. Summary table of modeled and measured NOx.

	Measured[a] at roof-top in central Stockholm (µg/m^3)	Calculated[a] (µg/m^3)	Number of days	Corre-lation	Slope[b]	Intercept[b] (µg/m^3)
2005	20.12 ± 1.43	16.27 ± 1.00	341	0.77	0.54 ± 0.02	5.5 ± 0.6
2004	21.74 ± 1.38	20.37 ± 1.07	316	0.73	0.57 ± 0.03	8.0 ± 0.7
2003	22.63 ± 2.03	22.22 ± 1.24	323	0.80	0.49 ± 0.02	11.2 ± 0.6
2002	23.37 ± 1.40	22.67 ± 1.28	343	0.74	0.67 ± 0.03	6.9 ± 0.9
2001	23.35 ± 1.35	21.44 ± 1.39	334	0.79	0.81 ± 0.03	2.5 ± 0.9
2000	25.19 ± 1.59	21.36 ± 1.18	317	0.80	0.60 ± 0.02	6.3 ± 0.7
1999	24.09 ± 1.57	22.14 ± 1.20	276	0.79	0.60 ± 0.03	7.6 ± 0.8
1998	27.49 ± 1.88	23.71 ± 1.62	257	0.74	0.64 ± 0.04	6.2 ± 1.1

[a] Average ± 95% confidence interval.
[b] (Modelled) = Slope± standard error *(Measured values) + Intercept± standard error

8.1.2 Relation between PM and NOx in exposure concentrations

The population exposure of NOx and PM exhaust varies depending on the distance to the roads and depending on the fleet composition. In central Stockholm, where most people live, gasoline exhaust may be somewhat more important for NOx and PM exposure than in residential areas, along the highways, outside the city centre.

The fraction of HDV:s varies with road type. In the inner city, there is an environmental zone, where only low emission HDV:s are allowed. The emission database used in our simulations should describe all these differences, but of course there are uncertainties since the traffic composition is not known in detail everywhere. In addition the emission database used in the simulations for all the years does not consider any changes in fleet compositions for different road types (only emission factors for different vehicle types change).

For this project we have not modelled the contributions to population exposure due to different vehicle types. We have only determined the exposure due to total traffic exhaust for NOx and PM. However, in the EMFO-TESS project population weighted concentrations of NOx and PM for LDV and HDV were made separately (Table 9). In this case the emission factors are from 2003 and the meteorology is a climatology based on 15 years of meteorological data. As can be seen the population weighted ratio of PM/NOx is 15 mg PM/g NOx for HDV's and 32 mg/g for LDV's, with an overall mean of 24 mg/g for all vehicle exhaust. As indicated above the emission factors are uncertain, contributing to making the calculated population weighted concentrations of PM very uncertain.

Table 9. Populated weighted mean concentrations of NO_x and particles ($\mu g/m^3$) for the Greater Stockholm area during 2003 (from the EMFO-TESS project; Johansson & Eneroth, 2007).

Substance	Road traffic	Road traffic, ldv[1]	Road traffic, hdv[2]	Sea traffic	Power plants	Residential heating
NO_x	5.86	3.14	2.44	0.18	0.36	0.68
PM Combustion particles	0.14	0.10	0.036	0.0063	0.051	0.59
PM/NOx (mg/g)	24	32	15	35	142	868

[1] ldv = light duty vehicles

[2] hdv = heavy duty vehicles

8.1.3 Local road dust

The concentration of coarse particles in the urban background (excluding the non-local particles) is due to local emissions from road wear, tire wear, brake wear and possibly other mechanically generated particles. Especially during winter and spring (when studded tires are used) road surface wear dominates (Johansson et al., 2006). During summer and autumn brake wear and possibly tire wear may make important contributions. Figure 30 shows the estimated annual contributions from road traffic to PM10 levels in Stockholm.

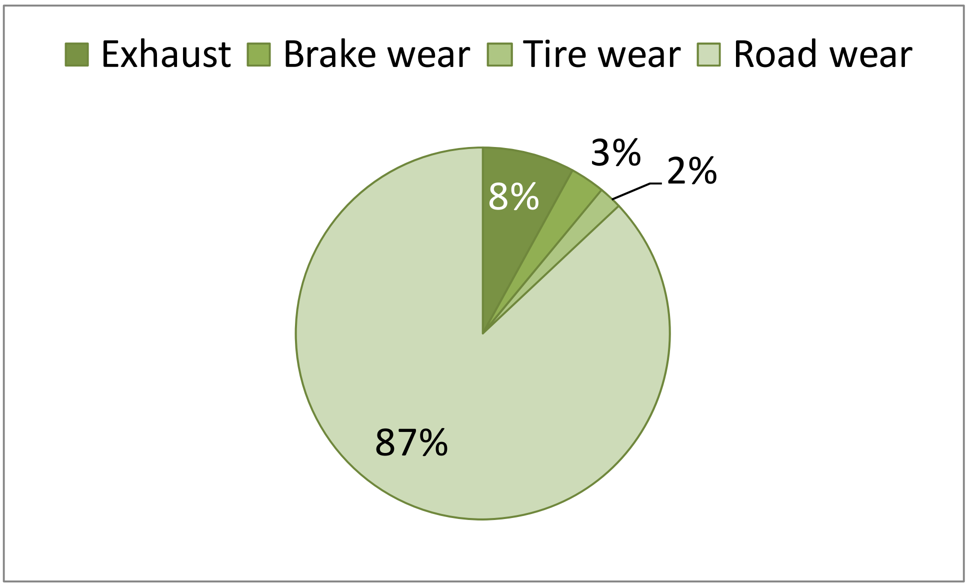


Figure 30. Annual mean contributions from local road traffic to PM10 in Stockholm (from Johansson et al., 2004).

Table 10. Comparison of emission factors for PM10 in Stockholm 2003. (mg per vehicle-kilometer).

Source	**Light duty vehicles**	**Heavy duty vehicles**	**Total**	**Notes**
Exhaust	15.3	94.3	19.5	According to the emission database of Stockholm and Uppsala county
Brakes	5.1	25.3	6.2	Based on Westerlund & Johansson (2002) assuming that 35 % becomes airborne (Garg et al., 2000)
Tyres	3.5	18.6	4.3	CEPMEIP (2002)
Total emission factor excluding road wear/suspension	23.9	138.2	30.1	Sum of exhaust, tyre- and brake wear
Annual mean in the emission database			244	Based on invers modelling at Hornsgatan extrapolatrd to whole Stockholm
Suspension and direct emission due to road, tyre and brake wear			214	Calculated as difference between the total estimated from measurements (Omstedt et al., 2005)

8.2 Exposure due to long range transport

8.2.1 Comparison between modelled and measured concentrations

In this section we take a look at the model performance in Europe and Sweden.

Modelled EC and OM were compared to measurements from a Europe wide campaign that took place during one year (July 2002 to July 2003) (extracted from http://www.emep.int, 2007-03-01, Yttri et al., 2007).

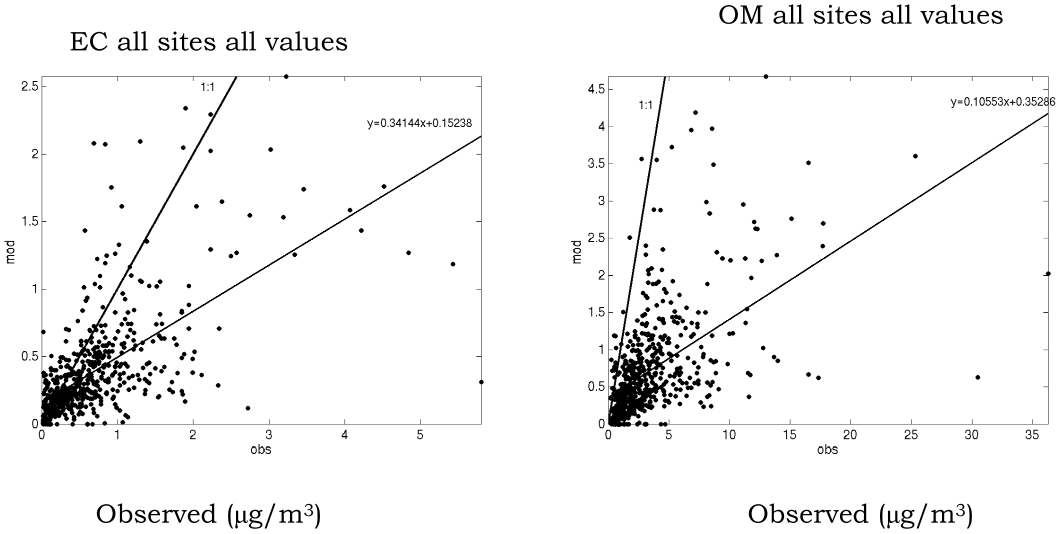
For OM there is a difference between what is modelled and what is measured. In the model we have only included directly emitted (primary) OM, whereas the measurement cannot distinguish primary and secondary components of OM. Furthermore, the measurements give total organic *carbon* (OC) rather than OM. OM contains a variable fraction of other atoms than carbon (mainly hydrogen, oxygen, nitrogen and sulphur). This means that the measured OC needs to be multiplied by a factor to correspond to modelled OM. We multiplied measured OC with the factor 1.4 to translate it to OM. The correct factor is not known and depend on location and age of the organic aerosol. 1.4 is within the range of possible values mentioned in for example Kupianen and Klimont (2007).

The measured *total* OC multiplied by 1.4 was compared to the modelled *primary* OM. This ratio (OM/OC) depends on the origin of the organic matter and the value varies depending on sampling site. Russell, (2003), Saxena & Hildemann (1996) and Turpin et al. (2000) suggested it to be 1.4 and Turpin and Lim (2001) suggested 1.6 for an urban aerosol. Due to the oxidation of primary organic compounds in the atmosphere, the factor has been suggested to be as high as 2.1 (1.9-2.3) for aged (nonurban) aerosol and 2.2-2.6 for an aerosol heavily impacted by wood (Saxena and Hildemann, 1996; Turpin et al., 2000).

In our model calculations, a large underestimation of OM is expected since the *secondary* OM is not included.. Exactly how much that is secondary and how much is primary at different sites is very difficult to quantify by measurements. The model calculations are uncertain both because of incomplete data and understanding about source contributions (both natural and anthropogenic). In many cities total OM constitute a very large fraction of PM2.5. Sillanpää et al. (2005) found 21% of PM2.5 being due to OM at an urban background site in Barcelona and 54% of PM2.5 in Prague. For urban background in Helsinki 50% of PM2.5 and 36% of PM10 (annual mean values) was due to OM (Viidanoja et al., 2002). Generally, secondary OM is more important during summer compared to winter.

In

Figure 31 we display scatter-plots for EC and OM, showing the correlation and bias.


EC all sites all values

OM all sites all values

Observed (µg/m³)

Observed (µg/m³)

Figure 31. Scatterplot displaying comparison of modelled (y-axis) and measured (x-axis) EC and OM concentrations. Daily mean values (N=603) for all regional stations (N=12). Note that measured OM but not modelled OM include secondary organic compounds, which to a large degree explain the bias. Unit: µg m^3.

For OM the underestimation is 79 % and the correlation is 0.53. For EC the corresponding values are 43% and 0.63. When comparing the spatial average (the average for each station for the whole campaign) the correlation for EC is 0.93 and for OM it is 0.91. Thus, the model is better at predicting the spatial distribution in Europe than to predict the daily variations at individual stations.

For other particulate components we see an underestimation in Europe. The model results for the year 2001 are summarized in Table 11. The bias for aerosol nitrate is -10 % and the correlation (for daily average concentrations) is rather low (0.33). The sum of nitrate and nitric acid, which is easier to measure correctly, is overestimated by 15 %. Nitric acid is also overestimated which may be an indication that formation of particulate nitrate from HNO_3 is underestimated in the model.

Particulate ammonium is underestimated on the average in Europe by 40 %. Many stations measure the sum of gaseous NH_3 and aerosol NH_4^+ and for this “total ammonium” the average model bias is low (-1%)

Sulphate is underestimated on the average in Europe by 32 % and the correlation of all available daily averages for 2001 is 0.51.

The general underestimation of particulate compounds needs to be remembered when using the results in exposure calculations. There is an even greater underestimation far from the sources indicating a too short life-time in the model (or too low emissions and/or large-scale background concentrations) of sulphate and ammonium (the underestimation of nitrate is smaller).

Table 11. Comparison of measured and modelled concentrations of air pollutants in Europe. For nitrogen and sulphur containing compounds daily average concentrations are compared, for ozone hourly average and daily maximum (1-h) concentrations, and for EC and OM, daily averages (measured once per week). All available values were compared for all stations for the year 2001,

except for EC and OM for which the time period was July 2002 to July 2003. Units: µg m^{-3} (S/N/EC/OC) or ppb(v) (O_3).

	mean observed	mean model[a]	model bias (%)	Correlation (global)	#cases
NO_2	1.84	1.87	1.6	0.48	16719
NO_3^-	0.39	0.35	-10.3	0.33	7673
HNO_3	0.1	0.12	20.0	0.27	4086
$NO_3^- + HNO_3$	0.46	0.53	15.2	0.4	13692
NH_3	2.02	0.99	-51.0	-0.05	2487
NH_4^+	0.83	0.5	-39.8	0.46	5916
$NH_4^+ + NH_3$	1.27	1.26	-0.8	0.37	13762
SO_2	0.79	0.81	2.5	0.45	23958
SO_4^{2-}	0.72	0.49	-31.9	0.51	25147
O_3	29.54	28	-5.2	0.65	824334
O_3 daily max	39.58	37.37	-5.6	0.78	35213
EC	0.66	0.38	-43	0.63	603
OM	3.35	0.71	-79	0.53	603

[a] The model concentrations (except for ozone) are given in the unit µg m^{-3} at a standard atmospheric density (1.2 kg/m^3).

For sulphate, a validation has also been made for the whole simulation period (1997-2003). In Table 12 some results from the European comparison are displayed. We also show results for the (4 - 6) Swedish measurement stations within the EMEP network. For the whole time period the model behaves fairly similarly to the one-year comparison in Table 11. In total the underestimation is slightly larger and the correlation slightly better. From the comparison it is also evident that the underestimation of sulphate is larger in the northern parts of Europe than in central and southern regions. The results for Sweden also indicate this. There is likely an underestimation of the long-range transport in the model, which should be remembered when using the results. It is also possible that the emissions are underestimated for some regions in Europe.

Table 12. Comparison of modelled and measured sulphate concentration for the whole time period 1997-2003 for the whole of Europe and for Swedish sites only. Units: µg S m^{-3}.

	Mean observed	Mean model[a]	Model bias (%)	Correlation (global)	#cases
Europe	0.758	0.491	-35.3	0.555	178981
Sweden	0.592	0.309	-47.9	0.627	10989

[a] The model concentrations are given at standard atmospheric density (1.204 kg/m^3).

8.2.2 Population exposure and background concentrations

First we will discuss the population exposure in Sweden as a whole, after this we will discuss the situation, as modelled, for regional background in the Greater Stockholm region.

8.2.2.1 Swedish population exposure

The population exposure is calculated as the concentration multiplied by the number of people in each 44 km × 44 km grid square, accumulated over the whole of Sweden according to the equation

$$E_r^p = \sum_{i=1}^{N} n_i \cdot c_{i,r}{}^p \ .$$

Here $E_r{}^p$ stands for the exposure in Sweden due to particulate component p from region r. n_i is the number of people in grid point i and $c_{i,r}{}^p$ is the concentration of particulate component p in grid point i due to region r.

In Figure 32 the population weighted concentration for the summed primary PM below 2.5 micrometers are displayed for different contributions. The spatial distribution and the impact of the population exposure in Sweden will be different for different source regions. The continental emissions have impacts mainly in southern Sweden. Swedish sources and emissions from Denmark, Norway and Finland also contribute in northern Sweden. We suspect that the long-range transport of continental emissions to the northern parts of Sweden may be somewhat underestimated.

Population exposure is a useful measure since it gives a possibility to estimate the expected health outcome in a population. The average or accumulated concentration in a region or country does not take into account where people are. This method estimates exposure under the assumption that people spend most of their time in the grid point where they live. This is not always valid of course but it is a better estimate for comparison with health outcomes than simply using the concentration. Population weighted concentrations are calculated according to the following formula

$$C_r^p = \frac{\sum_{i=1}^{N} n_i \cdot c_{i,r}{}^p}{\sum_{i=1}^{N} n_i} \ .$$

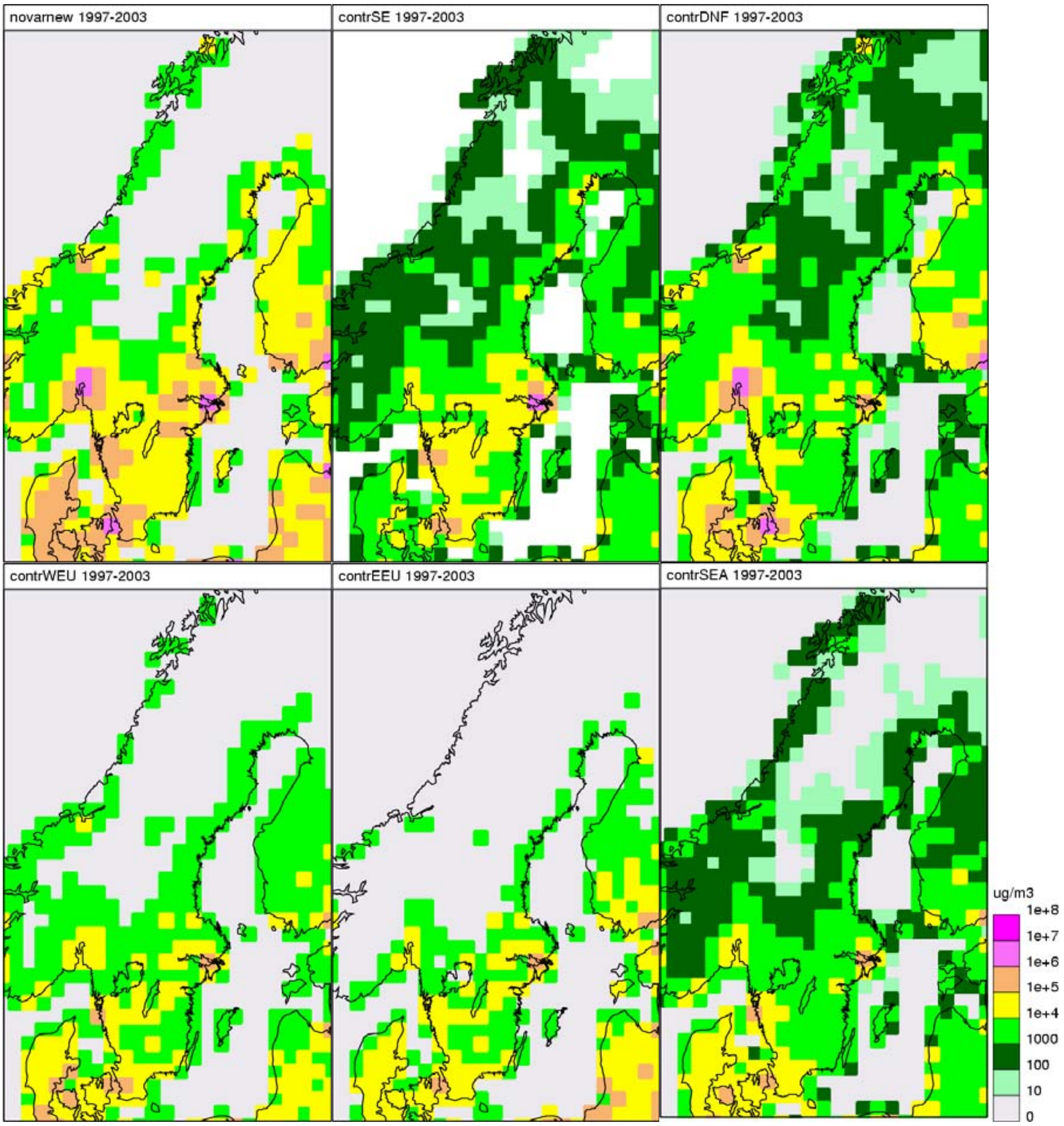


Figure 32. Population exposure in and around Sweden from directly emitted (primary) fine particles ($PPM_{2.5}$). Total population exposure and contributions from different regions. Upper panels: Left: Total exposure; Middle: Swedish sources; Right: Sources in Denmark, Norway and Finland. Lower panels: Left: Western European sources; Middle: Eastern European sources; Right: $PPM_{2.5}$ from international sea traffic. Unit: person × $\mu g/m^3$.

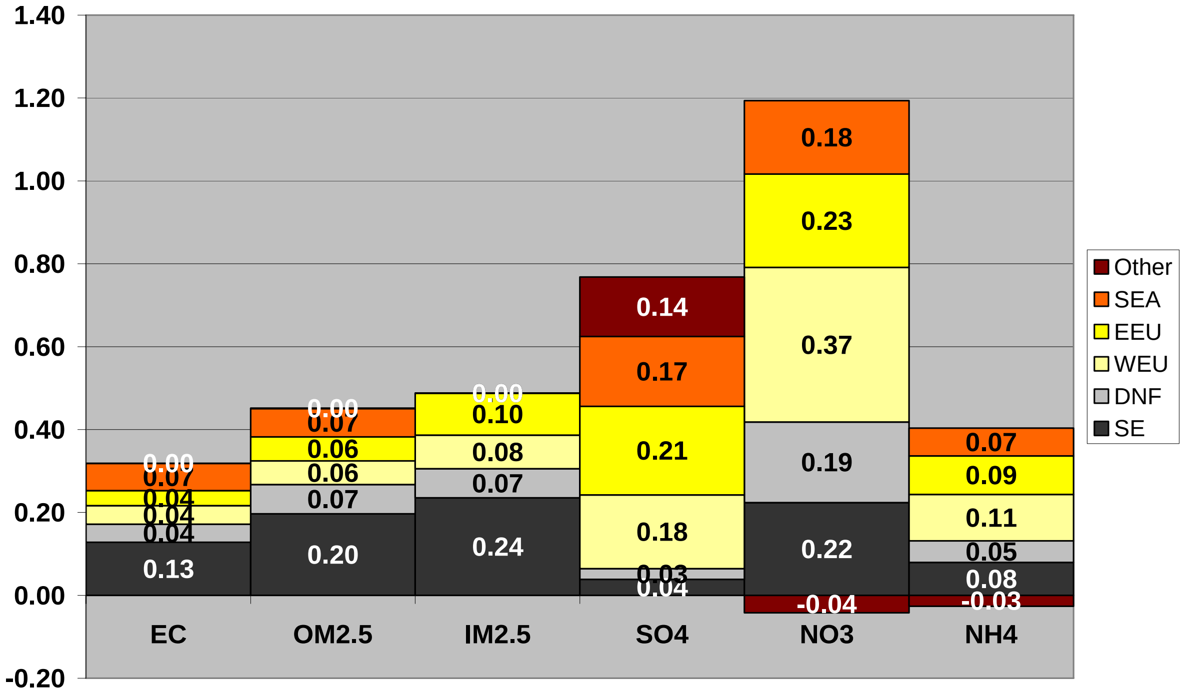


Figure 33. Population weighted average contributions to concentration in Sweden due to different regions for different particulate components. Unit: µg m^{-3}.

Figure 33 demonstrates the modelled population exposure in Sweden in terms of the population averaged concentration for different particulate components (presented as columns). For each component we also display the contribution from different regions to the exposure of that particulate component. Other sources than those incorporated into the emission scenarios are displayed as “other”, which means the emission scenario results subtracted from the modelled situation with total emissions included. This means that the “other” contribution includes non-linear effects and concentrations resulting from emissions which are not included in any scenario, e.g. volcanic and oceanic emissions of sulphur. For the secondary components the non-linear effects are visible through the occurrence of negative population weighted average; the sum of the source regions contribution is larger than exposure in the computation with all emissions included.

The corresponding exposures (population weighted averages) are displayed in Table 13. The largest contribution to population exposure in Sweden is from the secondary inorganic aerosol components: sulphate, nitrate and ammonium. The largest of these is nitrate.

The Swedish contribution to Swedish population exposure is largest for the primary emitted compounds: EC, OM and $IM_{2.5}$, whereas the Swedish contribution to sulphate is particularly small.

For the primary emitted compounds the distribution between the rest of the emission scenarios are of equal size, except for international sea traffic (SEA), which is a larger contributor for EC than the other scenarios (DNF, WEU, EEU). SEA make no contribution at all to $IM_{2.5}$ since it is not emitted in that sector.

Table 13. Swedish population exposure, averaged over the years 1997-2003, for particulate components and the relative distribution from different source regions. Population weighted average concentration of particulate components and the distribution of origin between source regions. Unit: weighted average: μg m^{-3} and contributions: %.

	EC	OM2.5	IM2.5	SO4	NO3	NH4
Weighted average (μg m^{-3})	0.32	0.45	0.49	0.77	1.15	0.38
SE (%)	40	44	48	5	19	21
DNF (%)	14	16	14	3	17	14
WEU (%)	14	13	17	23	32	30
EEU (%)	11	13	21	28	20	25
SEA (%)	21	15	0	22	15	18
Other (%)	0	0	0	19	-4	-7

To compare the origin of the population exposure to the distribution of emissions we recapitulate that the emissions were greater in western Europe than in eastern Europe for NOx, NH_3, CO and VOC, whereas the emissions in eastern Europe were greater for SOx and primary PM. For sulphate, nitrate and ammonium the origin of the population exposure follows the same pattern; the contribution is greater from that source region from which they are emitted in larger amounts. However, for the primary PM components this is not the fact, except for $IM_{2.5}$. The reason for this is probably due to the shorter lifetime of these pollutants, and the fact that prevailing transport patterns (the wind is more commonly south-western than any other wind direction in northern Europe) and other meteorological factors are important.

To illustrate the difference between using the population distribution or not in the estimation of exposure concentrations we compare Figure 33; displaying the population averaged concentration in Sweden, to Figure 34 displaying the average concentration in Sweden due to the different source regions and international shipping. For calculating the average Swedish concentration the following equation was used

$$C_r^p = \frac{1}{N^{Sweden}} \sum_{j=1}^{N^{Sweden}} c_{j,r}{}^p ,$$

where i is an index running over all Swedish grid boxes, $c_{j,r}{}^p$ is the concentration in grid box j due to region r for particulate component p and N^{Sweden} is the total number of grid boxes in Sweden.

From the two figures we draw the conclusion that the population weighted average concentration in Sweden is greater for all modelled particulate components. The interpretation of this could be that the Swedish contribution to the average is greater since the Swedish emissions correlate with the population density, but this is not the only reason. For the other regional contributions there is also an increase, which probably can be explained by the gradient in Sweden in both population density and concentration decreases from south to north. The decrease in concentration is due to large emissions on the European continent.

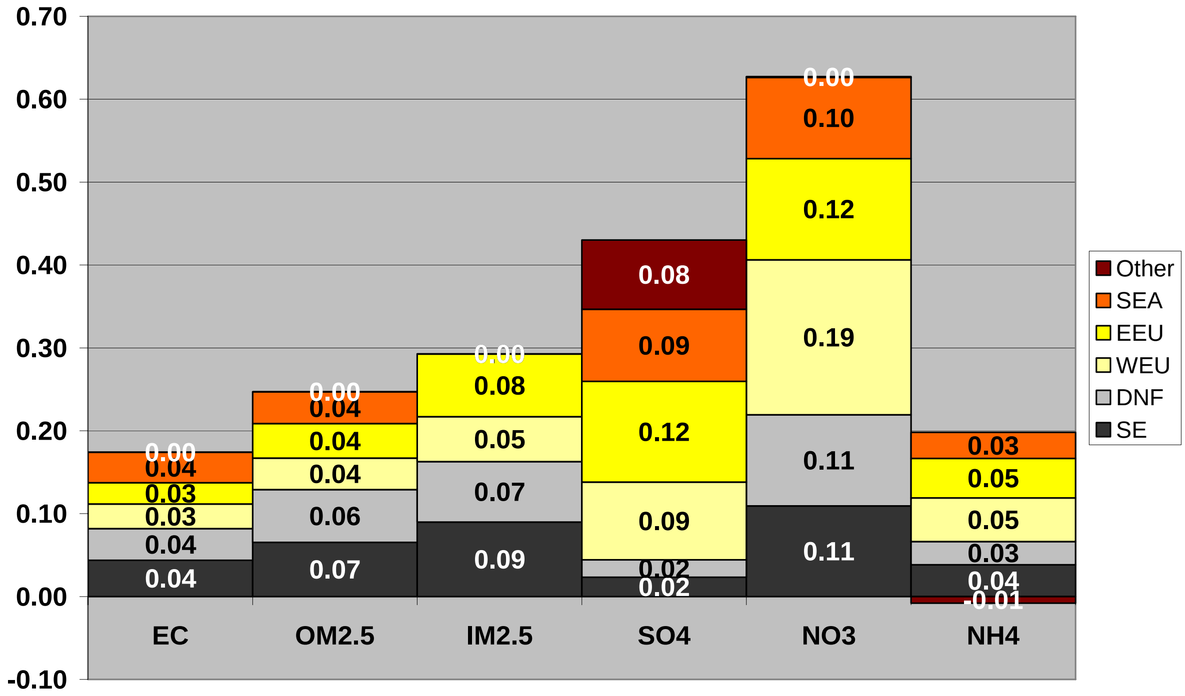


Figure 34. Average concentration in Sweden due to different regions for different particulate components (the average is not population weighted). Unit: µg m^{-3}.

Table 14 demonstrates the inter-annual variability in Swedish population exposure for different modelled particulate component and for different regional contributions. Here, the inter-annual variability is defined as the relative standard deviation in annual averages according to the following equation:

$$IAV(\%) = 100 * \frac{std^{y}(c_y)}{avg^{z}(c_z)} = 100 * \frac{\sqrt{\frac{\sum_{y=1997}^{2003}(c_y - \bar{c})}{N-1}}}{\bar{c}},$$

Where

$$\bar{c} = avg^{z}(c_z) = \frac{1}{N}\sum_{y=1997}^{2003} c_y .$$

where IAV is the inter-annual variability, $std^y(c_y)$ is the standard deviation of the annual averages c_y and $avg^z(c_z)$ the total average, $\bar{c}$, of annual averages. N (=7) is the total number of years.

We see that for the contributions from western Europe all components have rather similar inter-annual variability (16%-25%). For the contribution from eastern Europe the SIA components (SO4, NO3 and NH4) show much larger inter-annual variability (24%-27%) than the primary emitted components (6%-8%).

The large inter-annual variability, especially for secondary components which contribute most to the exposure, indicates that it is important to include more than one year in studies of the health impact since inclusion of one year or less does not give an accurate estimate of the exposure. It also means that it is very important with long-term records of air pollutants, both measured and modelled to keep track on variability and long-term evolution of exposure in Sweden. A combination of modelling and measurement can give an estimate on the exposure and the relative contribution of different components and different source regions to the Swedish population exposure.

Table 14. Inter-annual variability calculated as relative standard deviation of annual averages in regional contributions to Swedish population exposure for the different particulate components. Unit: %.

	EC	OM2.5	IM2.5	SO4	NO3	NH4
SE	13	13	9	5	12	11
DNF	5	6	6	15	13	11
WEU	23	23	21	25	16	20
EEU	6	6	8	26	24	27
SEA	8	8	X	6	12	6

8.2.2.2 Greater Stockholm region population exposure

In this section we will discuss the contribution of long-range transport to the exposure of people in the Greater Stockholm region. The regional background in the Greater Stockholm area is interpreted here as being a grid point close to Stockholm, where the influence from the Stockholm region emissions is considered small enough. Therefore when we talk about the exposure in this section we imply the average background concentration at the chosen point (Aspvreten). The scenario excluding Swedish emissions provides the possibility to distinguish the contribution from other regions than Sweden to the exposure in Stockholm due to non-local emissions of particles.

Due to the general underestimation of the particulate components that have been validated throughout Europe it is important to remember that there may be errors in the exposure amount. There may also be errors in the distribution between source regions for the population exposure or for average concentrations. It is less probable that the distribution between west and east differs due to too fast sink processes and therefore it is better to compare eastern and western contribution distribution than the contributions from Sweden and eastern Europe.

Table 15 displays the 7 year average of the rural background concentration for different particulate components. According to this model study more than 50% of the primary PM components is due to emissions in Sweden. For SIA the largest contribution is from eastern (27%) and western (23%) Europe, whereas the Swedish contribution is smaller and of similar size as the international shipping contribution. The contribution from emissions in Denmark, Norway and Finland, makes a small contribution to the PM2.5 exposure in Greater Stockholm region.

Table 15. Rural background concentration in the Greater Stockholm region averaged over the 7-year time period (1997-2003) for modelled particulate components. Distribution of contributions from different source regions. Units: average: μg m^{-3}, contributions: %.

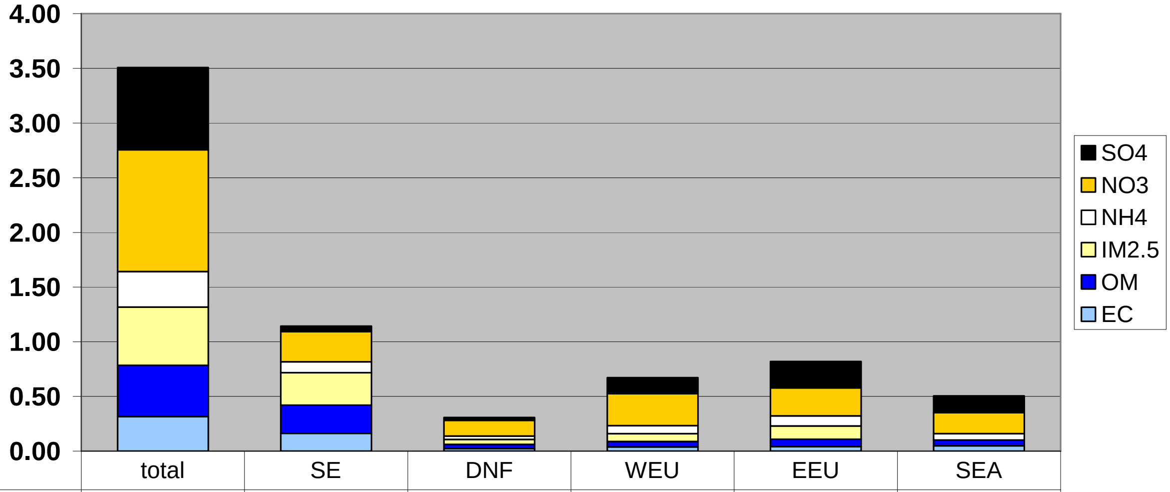
	EC	OM	$IM_{2.5}$	SO_4^{2-}	NO_3^-	NH_4^+	*Total $PPM_{2.5}$*	*Total SIA*
1997-2003 average background conc. (μg m^{-3})	0.32	0.47	0.53	0.75	1.11	0.32	*1.32*	*2.19*
SE (%)	51.6	55.2	55.7	6.8	24.8	30.3	*54.5*	*19.4*
DNF (%)	7.4	8.3	8.3	3.6	12.9	9.4	*8.1*	*9.2*
WEU (%)	12.1	10.7	13.3	19.6	26.1	23.1	*12.1*	*23.4*
EEU (%)	13.1	14.5	22.6	32.2	22.9	28.7	*17.4*	*27.0*
SEA (%)	15.6	11.1	0.0	20.4	17.3	17.6	*7.7*	*18.4*
Other (%)	0.2	0.2	0.1	17.4	-4.0	-9.1	*0.1*	*2.6*

Figure 35 shows the calculated contributions to exposure in Stockholm due to emissions in different regions for different components of PM2.5. The largest contribution to total modelled particulate exposure is due to emissions in Sweden. The eastern European contribution is larger than that of the western Europe, international shipping is also a large contributor.

Nitrate make the largest contribution to modelled fine particles.

It should be mentioned again that the relatively large contribution from Swedish sources may partly be due to a bias in the model; e.g. too fast wash-out of particles in the model of especially sulphate and ammonium making the contributions from eastern and western Europe too small.

As noted before, the calculated exposure does not include secondary organic particulate matter and inorganic compounds from soil suspension. Secondary organic compounds are due to both natural and anthropogenic emissions.



	total	SE	DNF	WEU	EEU	SEA
SO4	0.75	0.05	0.03	0.15	0.24	0.15
NO3	1.11	0.28	0.14	0.29	0.26	0.19
NH4	0.32	0.10	0.03	0.08	0.09	0.06
IM2.5	0.53	0.30	0.04	0.07	0.12	0.00
OM	0.47	0.26	0.04	0.05	0.07	0.05
EC	0.32	0.16	0.02	0.04	0.04	0.05

Figure 35. Modelled contributions to the average regional background concentrations of different PM2.5 components in the Stockholm region from different emission regions. Unit: µg m^{-3}.

Figure 36 shows the calculated annual average contributions to different PM-components ($PPM_{2.5}$, PPM_{10}, SIA) from the different source regions for the years 1997-2003. Note that the contribution from natural sources and boundaries are not included in the figure, thus there is a discrepancy between the total in the columns and the modelled scenario without emission decreases.

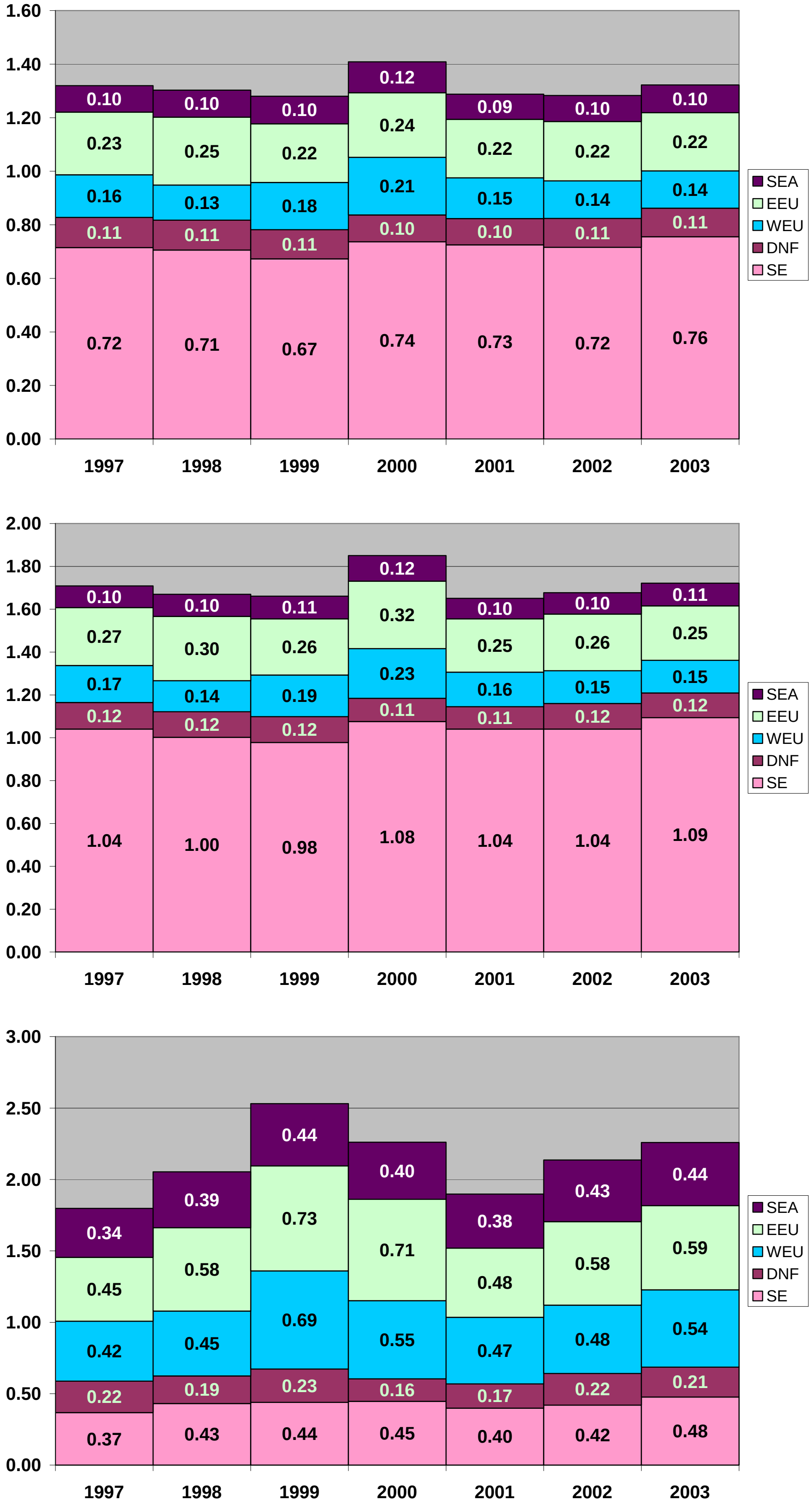


Figure 36. Annual average of PPM2.5 (top panel), PPM10 (middle panel) and SIA (bottom panel) and contribution from different regions to the background average concentration in the Stockholm region.

The year-to-year variation is similar for the different primary components and for the SIA components (not shown) but there are differences between the primary components and the SIA components. For SIA components the annual average is greatest for 1998 and 2001 displays the smallest annual average, whereas the greatest annual average for primary components is the year 2000. The reason for this difference seems to be that the long-range transport is greater during 2000 than during 1999, whereas the Swedish contribution is larger for 2000 for the primary components.

Table 16. Inter-annual variability calculated as relative standard deviation of annual averages in regional contributions to Greater Stockholm region background concentration of particulate components. Unit: %.

	EC	OM	IM2.5	SO4	NO3	NH4	PPM2.5	SIA
Total	4.1	3.5	3.2	12.0	9.4	10.6	3.4	9.7
SE	3.9	4.1	3.0	7.2	8.8	8.3	3.6	8.2
DNF	5.4	4.6	5.0	19.3	13.8	14.4	4.9	14.3
WEU	18.5	18.7	17.4	23.9	15.8	16.6	18.1	17.4
EEU	5.2	5.3	7.5	18.9	21.0	22.0	6.1	17.9
SEA	6.9	6.9	X	6.5	13.8	8.3	6.9	9.0

In Table 16 the inter-annual variability of the modelled particulate components is presented. The inter-annual variability of the SIA components lies between 9.4 and 12.0%. It is 9.7 % for total SIA. For the primary PM2.5 components the inter-annual variability is lower, between 3.2 and 4.1 %. It is 3.4 % for total PPM2.5.

For the primary particulate components the inter-annual variability is greatest for the WEU contribution, being 18.1 % for total PPM2.5. The SIA components have the greatest inter-annual variabilities for the contributions from EEU for nitrate and ammonia and from WEU for sulphate. The inter-annual variabilities for total SIA are large for the contributions from the neighbouring countries(DNF), WEU and EEU, ranging from 14.3% for DNF to 17.9 % for the EEU contribution.

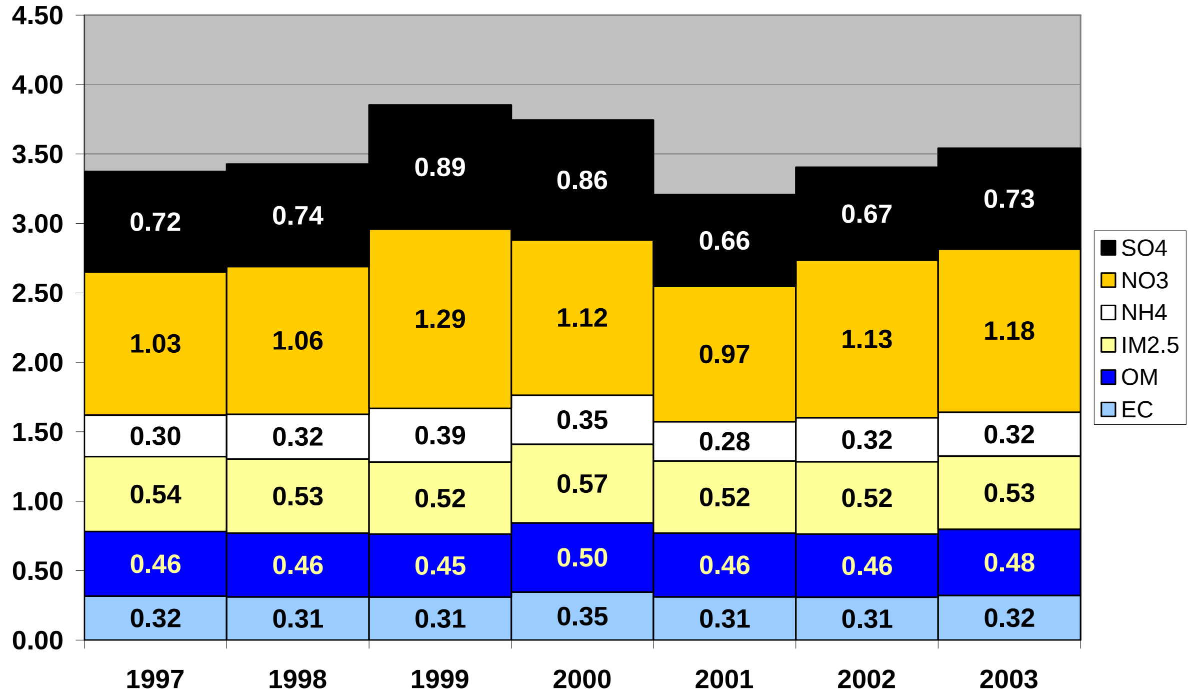


Figure 37. Modelled contributions from different components to PM2.5 to Stockholm regional background air. The different chemical components modelled are indicated by different colours; Black: Sulphate (SO4), Orange: Nitrate (NO3), White: Ammonium (NH4), Yellow: Directly emitted Inorganic Material (IM2.5), Dark Blue: Directly emitted Organic Matter (OM), Light Blue: Directly emitted elemental carbon (EC). Unit µg m^{-3}.

Figure 37 displays the modelled PM2.5 for regional background air in the Stockholm region for all the years included in the study. Note that not all particulate components are included in the model, hence the model should underestimate the measured concentration. As a total the largest average background concentration is for 1999. In the model, the largest contributor to the particulate mass is nitrate, but as discussed earlier this could be an effect of the model underestimating sulphate in this study.

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Appendix

Linearity check

For the SIA scenarios a 100% reduction in emissions in a certain source region may lead to large non-linear effects. By linear we here mean that if we reduce the emissions in one region by 10 % or by 20 % the concentration decrease of the 20 % reduction should be twice as large as the concentration decrease of the 10 % reduction. A 100 % reduction would not decrease the concentrations 10 times that of a 10% reduction because of the chemical reactions involved (e.g. reversible nitrogen reactions and coupling to ozone production which is non-linear most of the time).

For SIA we therefore performed two different emission reductions (by 20 % and by 10 %) to check for non-linearities. The result was that for most regions in Europe the long-term average was to a large extent linear.

However, non-linearities do occur under certain premises. In Table A1 the distribution between positive and negative values is displayed for the components contributing to SIA. The values are hourly averages at a regional background station in the south of Stockholm close to Aspvreten for the selection of years 2001-2003. Negative concentrations are due to non-linearities where an emission decrease results in an increase in concentration for short periods of time. For ammonium there is hardly no non-linear effect at all except for one night out of the three years, this was due to ammonium nitrate increasing for a few hours. The largest non-linear effects occurred for nitrate. This was mainly due to nitrate formed with other ions than ammonium, which could be due to non-linearities in the photo-chemistry scheme (e.g. due to ozone titration effects) affecting the formation and destruction of nitrate either close to the receptor point Aspvreten or earlier acting to change the lifetime of nitrate. The exact reason has not been investigated further, and is left for future studies.

Table A1. The distribution between positive and negative hourly averages for the period 2001-2003 for the contribution of ions to SIA. The distribution below and above 0.1 and 0.01 μg m^{-3} are also included in the table. Unit: % of total number of hourly averages.

	SO4	NO3	NH4
% positive values	62.6	75.8	65.3
% negative values	8.1	10.3	9.8
% values > 0.1 μg m^{-3}	22.5	28.7	16.0
% values < -0.1 μg m^{-3}	0.1	0.5	0.0
% values > 1 μg m^{-3}	3.2	8.6	1.3
% values < -1 μg m^{-3}	0.0	0.1	0.0

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