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Exposure

COMPARISON BETWEEN MEASUREMENTS AND CALCULATIONS BASED ON DISPERSION MODELLING (EXPOSE)

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Preface

The Expose-project is a co-operation between experts in environmental air quality and in environmental hygiene and medicine. Experts from local environmental authorities of Stockholm, Gothenburg and Malmö have been part of the project. Data from both personal measurements and fixed monitoring stations have been used and compared with air quality dispersion modelling. Gaussian dispersion models have been run for specific time periods corresponding to periods when personal exposure measurements have been carried out. The measured exposure has been compared with the calculated exposure for the three cities. Detailed emission data bases, meteorological data and dispersion models exist already for the cities. Since several years all three cities (Stockholm, Gothenburg and Malmö) have been using Air Quality Management Systems that involve dispersion models and detailed emission inventories. The present report presents the Stockholm exposure activities and results from fixed monitoring stations and dispersion modelling.

The Expose-project is part of the Swedish National Air Pollution and Health Programme (SNAP). It has been financed by the Swedish Environmental Protection Agency. The Stockholm activities have also received financial support from the Stockholm County Council Office of Regional Planning and Urban Transportation (RTK).

Occupational and Environmental Health at the Stockholm County Council has been responsible for the measurements of personal exposure and at some of the fixed stations. SLB analys at Stockholm Environment and Health Protection Administration has been responsible for the dispersion calculations.

The report has been written by Kristina Eneroth and Christer Johansson at SLB analys, and Tom Bellander at Occupational and Environmental Health.

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Sammanfattning (Swedish Summary)

Modellberäkningar av kvävedioxid (NO_2), partiklar (PM2.5 och PM10) och bensen har genomförts och validerats mot exponeringsmätningar vid ett antal fasta mätplatser i Stockholmsområdet under olika delar av året. Detta för att få ett mått på osäkerheter i exponeringsbedömningar, vilket är viktigt för tolkningen av resultat från epidemiologiska studier. Tillgång till tillförlitliga modeller är även nödvändigt för att kunna bedöma vikten av exponering från olika typer av källor.

Jämförelsen av spridningsmodellsberäkningar och exponeringsmätningarna av NO_2-halt visade en relativt god överrensstämmelse med ett beräknat r^2-värde på 0.71. Dock gav modellen något högre NO_2 koncentration än de passiva provtagarna, 25.0 ± 1.0 μg m^{-3} jämfört med 21.4 ± 0.7 μg m^{-3}. Resultaten indikerar att även om NO_2 koncentrationer inte bara beror på NO_x emissioner utan också på fotokemiska processer med bl.a. ozon inblandat, kan gaussiska modeller utan fotokemi användas för att beräkna NO_2-halt i omgivningsluften.

Vad gäller bensen var avvikelsen mellan modellberäkningarna och de uppmätta koncentrationerna relativt stor. Den modellerade medelkoncentrationen var 1.2 ± 0.3 μg m^{-3}, vilket kan jämföras med en uppmätt medelkoncentration på 1.5 ± 0.3 μg m^{-3}. r^2-värdet beräknades till 0.34. Den låga korrelationen mellan modellberäkningar och mätningar beror troligtvis på de stora osäkerheterna i modellens emissionsfaktorer (framförallt vad gäller kallstartseffekter och avdunstning), men låg precision i mätningarna och/eller de få tillgängliga datapunkterna kan också vara en bidragande orsak.

Jämförelsen mellan modellerad och uppmätt partikelkoncentration gav ett r^2-värde på 0.61. Generellt underskattade spridningsmodellen halten av PM2.5 och PM10. Den beräknade medelkoncentrationen var 6.1 ± 0.6 μg m^{-3}, jämfört med en uppmätt medelkoncentration på 9.0 ± 1.4 μg m^{-3}. Den största osäkerheten i modellberäkningarna är de lokala partikelemissionerna av uppvirvlat vägdamm. Dessa emissioner är särskilt höga vid torra vägförhållanden under senvintern och våren. Storleken på emissionerna beror även till stor del på andelen dubbdäck. I Stockholm kan dammpartiklarnas bidrag till PM10-halten vara upp till tio gånger större än bidraget från de direkta avgasemissionerna. I november, när de lokala dammemissionerna är låga, visade modellen generellt bra överrensstämmelse med de uppmätta partikelkoncentrationerna. I april däremot, när inflytandet av uppvirvlat vägdamm är stort, visade samma modell en grov underskattning av luftens PM10-halt.

Executive summary

In this project exposure measurements have been compared with model calculated exposure in order to identify the main uncertainties involved in exposure assessments. This is important for the interpretation of results in epidemiological studies. Reliable models are also necessary to be able to assess the importance of different sources for the exposure.

The focus has been on evaluating the accuracy of model calculations of ambient concentrations of particulate matter (PM2.5 and PM10), nitrogen dioxide (NO_2) and benzene. This has been done by comparing dispersion model calculations with fixed monitoring measurements at a number of different sites in the Stockholm area during different periods of the year.

The comparison between model calculations and fixed point measurements of NO_2 showed a generally good agreement with a calculated r^2-value of 0.71. However, the model gave slightly higher NO_2 concentrations than the passive samplers, 25.0 ± 1.0 μg m^{-3} compared to 21.4 ± 0.7 μg m^{-3}. This result indicates that even though NO_2 concentrations depend not only on NO_x emissions, but also on photochemical processes involving ozone, Gaussian models without photochemistry can be used to calculate NO_2 concentrations in ambient air.

For benzene the model calculations showed relatively poor agreement with stationary benzene measurements. The simulated mean concentration was 1.2 ± 0.3 μg m^{-3}, which should be compared with a measured mean concentration of 1.5 ± 0.3 μg m^{-3}. The r^2-value was 0.34. The relatively low correlation between the model calculations and the measurements is probably due to large uncertainties in the emissions factors (especially the importance of cold start emissions and evaporative losses) but may also be due to low precision of the measurements and/or the few data points available.

The comparison between model simulations and fixed point measurements of particles resulted in a r^2-value of 0.61. The dispersion model generally underestimated the concentration of PM2.5 and PM10 compared to the measurements, cf. 6.1 ± 0.6 μg m^{-3} and 9.0 ± 1.4 μg m^{-3}. The main uncertainty in the model calculations is the local road dust emissions. These emissions are especially important during dry road conditions in spring and late winter. The importance of road dust emissions also vary with the share of studded tyres. In Stockholm, where around 70 % of the cars have studded tyres, road dust emissions is probably more important than in for example Malmö, with around 40 % studded tyres. In addition, there are large variations during the year and between cities in the importance of local sources compared to long distance transport. In November, when the local road dust emissions are low, the performed model simulations generally showed high correlation with the fixed point measurements. In contrast, due deficiencies in the model's parameterization of road dust emissions, the same model calculations for April largely underestimated the ambient PM10 concentrations.

Introduction

Urban air pollution is associated with adverse health effects, including respiratory morbidity, cardiovascular diseases and mortality (WHO, 2005). Exposure assessment of air pollution is necessary to quantify this health impact. The interpretation of epidemiological studies critically depends on assumptions regarding the exposure.

Many different methods have been used for assessing exposure, from more indicative exposure assignments like “living close to roads” to more sophisticated methods using combinations of air quality measurements and air quality dispersion calculations (cf. Harrison et al., 2004; Chow et al., 2002; Colls and Micallef, 1997). The use of dispersion models makes it possible to calculate air pollution concentrations in different microenvironments (e.g. urban background, busy roadside, different street geometries), resulting in better expose assessment than using one single monitoring station to represent the total population exposure (cf. Harrison et al., 2004).

Most people spend a significant portion of their lives indoors, meaning that their exposure to air pollution differs from air quality levels from fixed air monitoring stations and model calculations (Colls and Micallef, 1997). However, ambient particles and pollutants penetrate indoors and may make a significant contribution to the concentration of indoor particles (Hussein et al., 2005; Chow et al., 2002).

Cyrys et al. (2004) measured fine particle mass (PM2.5) and black smoke (BS) and particle number concentration (NC) simultaneously indoors and outdoors at an urban location in Erfurt, Germany. They found a strong correlation between indoors and outdoor levels of PM2.5 and BS (lower for NC), indicating that ambient concentrations of PM2.5 and BS can be used as a good approximation of indoor concentrations in the absence of indoor particles sources.

Wu et al. (2005) used personal nephelometers to characterize asthmatic children’s exposure to PM2.5. They found that the personal exposures were higher than those measured at fixed sites. The subjects only received 45.0% of their exposure indoors at home, even though they spent more than 60.0% of their time there. In contrast, 29.2% of their exposure was received at school where they spent only 16.4% of their time. Thus, exposures in certain microenvironments with high PM levels where less time is spent can still make significant contributions to the total exposure.

To increase confidence in epidemiological associations between health outcomes and ambient concentrations of air pollution, a better understanding of the relationship between ambient concentrations and individual exposures is needed.

Aims of this study

The main aim of this study is to evaluate the uncertainties in quantifying population exposure of particulate matter, nitrogen dioxide and benzene.

In this study we focus on evaluating the accuracy of model calculations of ambient concentrations. This is done by comparing model calculations with fixed monitoring measurements at a number of different sites in the Stockholm area during different periods of the year.

Methods

Measurements

Measurement data of monthly nitrogen dioxide (NO_2) mean concentrations were available for 8 periods 1995-1999 in Järfälla, Sundbyberg, Solna and Stockholm City. The locations of the sampling sites are shown in Fig 1. The NO_2 measurements were performed using diffusive samplers, Palmes Tubes. This measurement technique is associated with several sources of error: short spikes with high concentrations, heightened absorption during periods of high wind speed, temperature dependence of the diffusion constant, oxidation of NO to NO_2, and degeneration of the absorbent (Plaisance, 2004; Ferm and Svanberg, 1995; Gair and Penkett, 1995; Campbell et al., 1994). The sampling times for the NO_2 measurements (n=495) are shown in Fig 2.

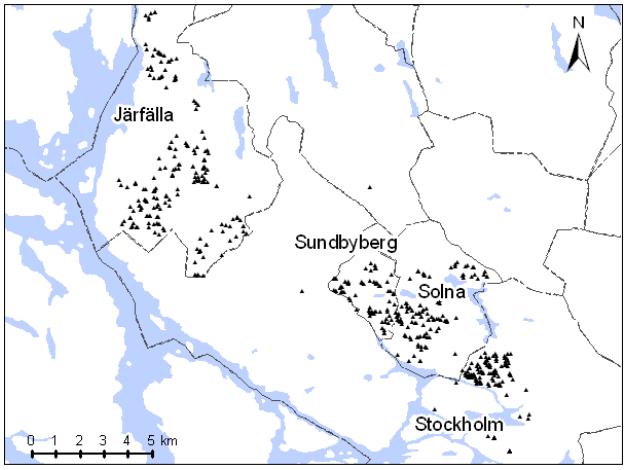

Fig 1. Location of sampling sites of NO_2 in Järfälla, Sundbyberg, Solna and Stockholm City during 1995-1999.

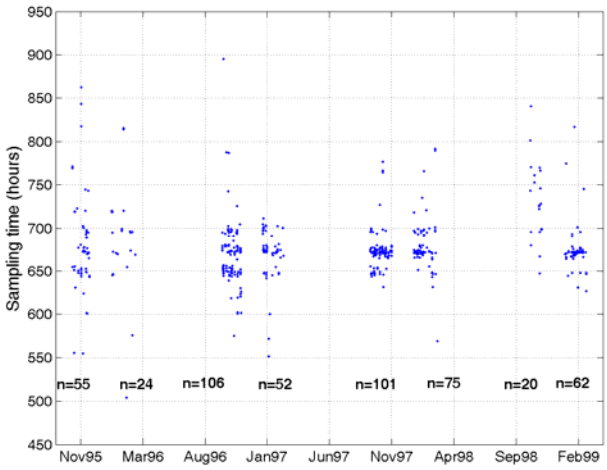

Fig 2. Numbers of NO_2 samples and sampling times for the measurements (n=495) during 1995-1999.

In addition to the monthly NO_2 samples, weekly NO_2 and benzene measurements from the spring 2003 were available. These measurements (n=21) were performed at two sites at Södermalm – one at street level and one at roof level - and at one site in Huddinge. NO_2 was measured using the Willems Badge technique. The benzene measurements were made using diffusive samplers with CarbopackX as adsorbent.

The measurement data of particulate matter consisted of daily mean concentrations of PM2.5 at three fixed stations - Huddinge, Solna and Lidingö. At the Lidingö station PM10 was measured as well. The measurements were performed during two periods: 8-21 November 2004 and 4-17 April 2005. The samples were analyzed using weighing under standard conditions.

Emission database

The emission database, administered by the Stockholm - Uppsala Air Quality Management Association, was used to calculate NO_x, benzene and PM10 concentrations. The database is yearly updated and contains detailed information about emissions from e.g. road and ferry traffic, petrol stations, industrial areas and households (Johansson et al., 1999). In Stockholm, road transport (both vehicle exhaust particles and resuspension of road dust) is the dominating source of primary particulate matter and the combustion of fuel is the main source of NO_x and benzene.

The road traffic is described using emission factors for different vehicles and road types given by the Swedish Road Authority's EVA-model 2.3. Five vehicle types and six purification steps (EURO 0-5, 1987-2008) as well as 45 different road types are characterized. The classification of roads is based on the signed speed limit, the percentage heavy-duty traffic and the temporal (diurnal, weakly and annual) variation of the traffic. Furthermore, for every road link the emission factor varies depending on the vehicle speed (7 different speed intervals are defined).

Table 1 presents total emissions as well as emissions from different source sectors of NO_x, PM10 and benzene in Greater Stockholm in 2003 (LVF, 2005). Road traffic accounts for about 60% of the total NO_x emissions. Corresponding values for PM10 and benzene are about 65% and 40%, respectively. There are large uncertainties in the emissions from working machinery. Since there is large uncertainties in both the total amount emitted and the spatial distribution of working machineries their contribution is not included in the model calculations. Shipping, industrial processes and energy production have only very local impacts on the concentrations. The PM emissions from residential wood burning are associated with very large uncertainties.

Table 1. Total emissions as well as emissions from road traffic, energy production, industrial processes, shipping and working machinery of NO_x, PM10 and benzene in Greater Stockholm in 2003 (tons/year; LVF, 2005).

Substance	Road traffic	Energy	Industry	Shipping	Working machinery	Total
NO_x	5890	2050	6	1440	1	9430
PM10	2070	820	2	90	120	3110
Benzene	140	230	≅0	2	≅0	370

Road traffic emissions of NO_x, benzene and PM10

The NO_X emissions from road traffic in Greater Stockholm have decreased from about 9000 tons per year in 1999 to about 6000 tons per year in 2003. This decrease is mainly due to less emission from passenger cars. The relative share of NO_X emissions from heavy duty vehicles has increased from 43% in 1999 to 47% in 2003. The emissions from the heavy duty vehicles stand for the largest uncertainties estimating the NO_X emissions in Stockholm.

The main source of benzene from road traffic in Stockholm is emissions from passenger cars - only a few percent of the total emissions origins from heavy duty vehicles. The total emissions from passenger cars are estimated to have decreased by about 67%, from 425 tons per year in 2000 to 140 tons per year in 2003. The cold start emissions are estimated to have decreased less, why the relative share of emissions from cold start has increased from 38% to 83% during the same period. There are large uncertainties associated with the benzene emission estimates, for example the effects of cold starts, evaporation processes and the relationship between concentrations of volatile organic compounds (VOCs) and benzene.

Tables 2 and 3 show mean NO_x and benzene emission factors for the whole vehicle fleet in Greater Stockholm used in the model calculations. However, previous studies indicate that the benzene emission factors in Table 3 may be overestimated. Omstedt and Johansson (2004), using measurement data in combination with model calculations, estimated an average emission factor between 21 and 28 mg benzene vkm^{-1} in Greater Stockholm for the year 2000. For the same year, Kristensson et al. (2004) estimated an emission factor of 17.3 mg benzene vkm^{-1} in Söderledstunneln in Stockholm.

Table 2. Average NO_x emission factors for the years 1995-2003 in Greater Stockholm.

Year	Light duty goods vehicles (NO_x g vkm^{-1})	Heavy duty vehicles (NO_x g vkm^{-1})	All vehicles (NO_x g vkm^{-1})
1995	-	-	1.87
1997	-	-	1.59
1999	0.83	8.77	1.33
2000	0.73	8.29	1.20
2001	0.62	7.76	1.06
2002	0.55	7.11	0.96
2003	0.50	6.62	0.88

Table 3. Average benzene emission factors for the years 1999-2003 in Greater Stockholm.

Year	Light duty goods vehicles (benzene mg vkm^{-1})	Heavy duty vehicles (benzene mg vkm^{-1})	All vehicles (benzene mg vkm^{-1})
1999	77.14	0.62	72.39
2000	66.19	0.55	62.11
2001	54.42	0.48	51.07
2002	47.54	0.42	44.61
2003	42.46	0.57	39.86

The most important local source of PM10 in Stockholm is mechanically generated road dust due to the wear of roads (Omstedt et al., 2005). The road dust consists of particles from tyre, brake and pave wear, anti-skid material (sand and salt) as well as deposited airborne particles and is accumulated during the winter season, in pace with anti-skid treatment and the use of studded tyres. The contribution from the studded tyres to the dust depot can be substantial depending on the street moisture (the wear is larger if the roadway is wet) and the amount of sand and salt on the roadway. The re-suspension of road dust is limited by road surface moisture. When the roadways are dry the re-suspension may be larger than 10 times the direct emissions from the exhaust pipe. Table 4 presents estimated values for emissions factors of re-suspension and direct emission of particles.

Table 4. Estimated emissions factors for the direct and re-suspension part of PM10 emissions in Stockholm (Omstedt et al., 2005).

Process	Emission factor (mg vkm^{-1})
Exhaust (diesel and gasoline combustion)	20-30 [a]
Brake wear	5.9 [b] (light-duty vehicles)
	29 [b] (heavy-duty vehicles)
Tyre wear	0 [c]
	1.2 [d]
Road abrasion	10
Resuspension	205 [e] (range: 100-1000)

[a] According to the Swedish Road Authority's EVA-model.
[b] Based on studies by Westerlund and Johansson (2002) and Garg et al. (2000).
[c] Based on studies by Abu-Allaban et al. (2003).
[d] Based on studies by EPA (1985).
[e] Annual mean value based on street canyon measurements (Johansson et al., 2005).

Table 5 shows mean PM10 emission factors in Greater Stockholm used in the model calculations. The emission factors for the total PM10 emissions are based on traffic number and particle concentration measurements at Hornsgatan in Stockholm.

Table 5. PM10 emission factors for the years 1999-2003.

Year	Light duty goods vehicles (Exhaust PM10 mg vkm^{-1})	Heavy duty vehicles (Exhaust PM10 mg vkm^{-1})	All vehicles (Exhaust PM10 mg vkm^{-1})	All vehicles (Total PM10 mg vkm^{-1})
1999	19.79	192.77	30.51	322.53
2000	17.80	165.27	27.11	319.60
2001	15.57	140.72	23.39	315.63
2002	14.42	119.14	21.02	313.51
2003	13.49	99.69	18.94	311.43

Dispersion modelling

To calculate the temporal and spatial distribution of NO_x, benzene and PM10 concentrations a Gaussian dispersion model was used. The wind field for the whole model domain was calculated based on the concept described by Danard (1977). 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. 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). When the topography is relatively smooth, without dominating ridges or valleys, the free wind is assumed to be horizontally uniform. In the present study, input data to the wind model is meteorological measurements at a 50 meter tall tower in Högdalen south of Södermalm, Stockholm. The measurements include horizontal and vertical wind speeds, wind direction, temperature, temperature difference between 5, 20 and 50 meters as well as global radiation. The wind model

takes into account land use variations and local topographic conditions at a resolution of 500 meters. The concentration of the pollutants was calculated at 2 meters above roof or ground.

The dispersion model calculates the NO_x concentration, i.e. the sum of NO and NO_2. Most of the NO_x is emitted as NO, which is rapidly oxidized in the atmosphere by ozone to NO_2. The relative amounts of NO and NO_2 are mainly controlled by the concentration of ozone (Johansson and Forsberg, 2005). To obtain NO_2 from NO_x a statistical relationship was used, $NO_2 = f(NO_x)$[1]. This relationship is based on measurements from stations at street and roof levels as well as background stations in Stockholm and its surroundings during the 1990s (Johansson et al., 1999). The empirical equation is non-linear, which means that at high concentrations, NO_2 is less sensitive to errors in NO_x emission data and dispersion calculations. Since NO_2 concentrations are highly dependent on photochemistry, the modelling of hourly NO_2 concentrations may need to consider variations in ozone levels. An alternative approach calculating NO_2 has therefore been suggested by Johansson and Forsberg (2005) where both NO_x and ozone levels are considered. Figs 3 and 4 show calculated hourly mean NO_2 concentrations – using the empirical and the photochemical equation, respectively - as a function of measured hourly mean NO_2 concentrations at Torkel Knutssonsgatan at Södermalm, Stockholm (n=14210). For the calculations hourly measurements of NO_x, ozone and temperature at Torkel Knutssonsgatan as well as global radiation at Högdalen were used. Both methods show good correlation between calculated and measured NO_2 concentrations with r^2-values higher than 0.9. However, at high concentrations the photochemical equation seems to give better agreement between calculated and measured NO_2 compared to the empirical relationship, which appears to underestimate the NO_2 concentration.

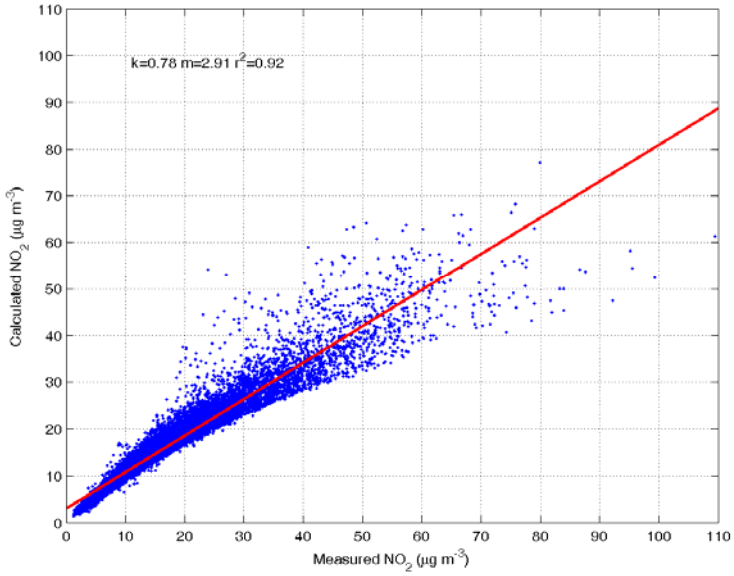

Fig 3. The relation between of calculated and measured hourly NO_2 mean concentrations at Torkel Knutssonsgatan during May 2001 to December 2002 (n=14210). For the calculations a statistical relationship, $NO_2=f(NO_x)$, was used. k=slope, and m=y-intercept of the regression line.

[1] $NO_2 = NO_x \wedge \left(A + \frac{B}{C+NO_x}\right)$ where A, B and C are empirical constants. A=0.66, B=34 and C=100.

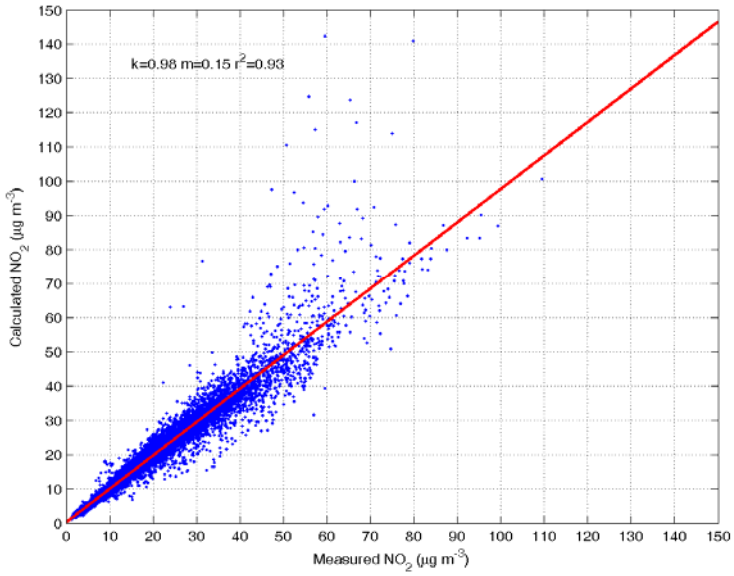

Fig 4. The relation between of calculated and measured hourly NO_2 mean concentrations at Torkel Knutssonsgatan during May 2001 to December 2002 (n=14210). For the calculations a photochemical equation was used. k=slope, and m=y-intercept of the regression line.

The model calculations represent only PM10 concentrations. Some information about the relative amounts of PM2.5 and PM10 can be obtained by comparisons of measurements. However, since these two particle fractions have different removal rates and partly different sources – that vary differently in space and time, the correlation between fine and coarse particles may be quite variable from site to site. Measurements by Wilson and Suh (1997) indicate that PM2.5 and PM10 concentrations are highly correlated at sites where fine particles dominate the PM10 concentration, whereas at sites with high levels of coarse particles the correlation is low. Fig 5 shows hourly mean PM2.5 concentrations as a function of hourly mean PM10 concentrations for April 2005 and November 2004. The PM concentrations are calculated as the difference between roof (Rosenlundsgatan) and street level (Hornsgatan) measurements, and thus reflect the correlation between PM2.5 and PM10 in local emissions, i.e. road traffic. The local emissions mainly consist of larger particles, with low concentrations of PM2.5 compared to PM10. This is particular true in spring when the contribution from re-suspension is higher than during other seasons.

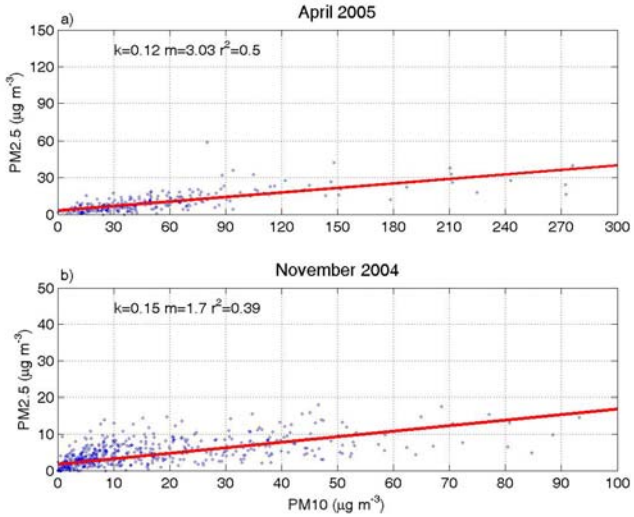

Fig 5. The relation between of PM2.5 and PM10 from local emissions (street – roof level concentrations) in April 2005 (a) and in November 2004 (b). k=slope, and m=y-intercept of the regression line. Note the scale on the axis differ between the two panels.

Table 6 presents statistics of the comparison between PM2.5 and PM10 observations. The amount of coarse particles is higher at street level compared to roof level due to re-suspension of road dust (Wallin, 1998). The percentage of PM2.5 of PM10 in April is about 24% and 41% at street and roof level respectively. In November, the percentage amount of PM2.5 is about 45% at street level whereas 67% of the particles at roof level comprise PM2.5. These values are in agreement with results by Johansson et al. (1999), making a similar comparison between PM2.5 and PM10 measurements at street and roof level at Södermalm, Stockholm. They found that at street level the PM2.5 concentration was about 43% of the PM10 concentration (June 1997 and April to June 1998), whereas at roof level PM2.5 concentrations were about 70% of the PM10 concentrations (April to September 1998). In the present study, the highest correlation between the PM2.5 and PM10 measurements was found during periods with high percentage PM2.5 of PM10 i.e. at roof level in November. This is due to relatively low road dust emissions of coarse particles in November and large contribution of long-range transported fine particles. In contrast, in March and April road dust emissions maximise, resulting in high concentrations of coarse particles (Omstedt et al., 2005; Bringfeldt et al., 1997).

Table 6. Comparison of hourly mean PM2.5 and PM10 measurement values at street level (Hornsgatan) and roof level (Rosenlundsgatan) for April 2005 and November 2004.

	April			November		
	Roof	Street	Street-Roof	Roof	Street	Street-Roof
PM2.5 ($\mu g\ m^{-3}$)	12.3	21.3	9.3	6.0	9.7	3.7
PM10 ($\mu g\ m^{-3}$)	29.9	89.6	51.2	8.9	21.5	12.9
PM2.5/PM10 (%)	41	24	18	67	45	29

The dispersion model calculations were performed for six different areas: one area which includes the whole study region, i.e. Greater Stockholm, and five smaller areas which only include the immediate surroundings of the measurement points, i.e. Järfälla, Solna, Huddinge, Lidingö and Stockholm City. The size of the calculation areas was defined to include the measurement points as well as large nearby pollution sources. The choice of number of grid squares was a compromise between feasible simulation times and high resolution. The calculations for the Stockholm region had a horizontal resolution of either 500 x 500 m or a 100 x 100 m. The resolution of the smaller areas varied between 25 x 25 m and 100 x 100 m depending on their size. Tables 7 and 8 show the size and the resolution for the different model calculation areas.

Table 7. Horizontal resolution and size for the NO_x and benzene model calculation areas.

Area	Horizontal resolution	Number of grid squares
Stockholm region	500 x 500 m	70x70
Järfälla	100 x 100 m	140x170
Solna	100 x 100m	100x100
Huddinge	100 x 100 m	230x190
Stockholm City	50 x 50 m	140x140

Table 8. Horizontal resolution and size for the PM10 model calculation areas.

Area	Horizontal resolution	Number of grid squares
Stockholm region	100 x 100 m	350x350
Lidingö	25 x 25 m	254 x 266
Huddinge	25 x 25 m	252 x 265
Solna	25 x 25 m	180 x 188

The NO_x and benzene concentration calculations were carried out for nine time periods whereas the PM10 concentration was calculated for two time periods. For each period three simulations were made:

1. The pollution concentration in Greater Stockholm with emissions from the whole simulation area.
2. The pollution concentration in Greater Stockholm with emissions from one of the smaller defined simulation areas, e.g. Lidingö.
3. The pollution concentration in a smaller defined area with emissions from the whole simulation area.

The contribution of emissions from Greater Stockholm to the smaller specific areas was calculated as the difference between the concentrations from simulations 1 and 2. This contribution was added to the calculated concentrations in each specific area, i.e. simulation 3. Finally, to obtain the total concentrations of NO_x, benzene and PM10, the contribution from long-range transport of pollutants was added to the modelled concentrations. Background measurements at Norr Malma, 15 km northwest of Norrtälje, was used for the long-range contribution of NO_2 and NO_x. For benzene, 0.5 µg/m^3 was used as background concentration (Johansson et al., 2001). The background concentrations of PM10 were obtained from the Aspvreten station, which is operated by the Department of Applied Environmental Science, Stockholm University and is situated about 70 km south of Stockholm.

Results

Monthly and weekly time-series of NO_2 and benzene

Fig 6 shows comparison between model calculations and measured NO_2 concentrations from passive samplers – both weekly and monthly samples are included. There is a good correlation between the model and the measurements, with a calculated r^2-value of 0.71. However, the model gives slightly higher NO_2 concentrations than the passive samplers, 25.0 ± 1.0 µg m^{-3} compared to 21.4 ± 0.7 µg m^{-3} (95% confidence interval).

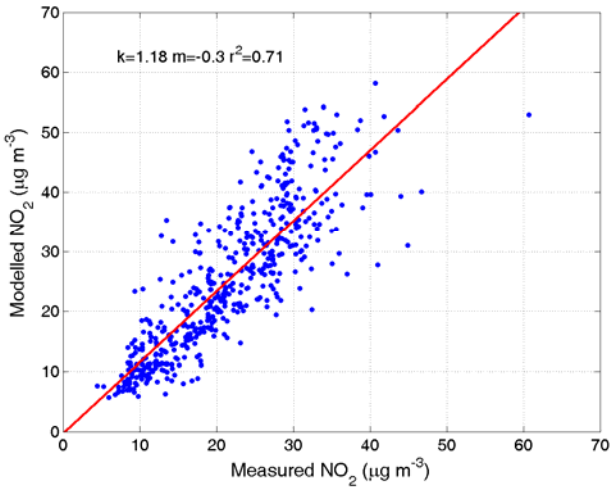

Fig 6. Comparison between NO_2 model calculations and passive samplers in Järfälla, Solna, Huddinge and Stockholm City during 1995-1999 and 2003. Samplers placed at street level have been excluded. The standard deviations of the slope (k) and the y-intercept (m) of the regression line are 0.03 and 0.76 µg m^{-3}, respectively.

Fig 7 shows comparison between model calculations and weekly mean benzene values at Södermalm and Huddinge during spring 2003. The discrepancy between measured and calculated concentrations is relatively large. The slope of the regression line is 0.69 and the r^2-value is 0.34. The simulated mean concentration is 1.2 ±

0.3 µg m^{-3}, which can be compared with a measured mean concentration of 1.5 ± 0.3 µg m^{-3} (95% confidence interval). The relatively low correlation between model calculations and measurements is probably mostly due to the large uncertainties in the emissions factors but may also be due to low precision of the measurements or the few data points available.

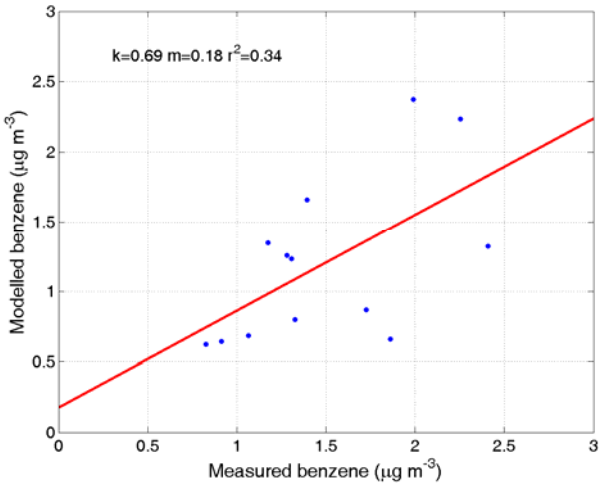

Fig 7. Comparison between benzene model calculations and passive samplers at Södermalm and Huddinge during spring 2003. Samplers placed at street level have been excluded. The standard deviations of the slope (k) and the y-intercept (m) of the regression line are 0.29 and 0.45 µg m^{-3}, respectively.

Daily time-series of stationary PM2.5 and PM10

Fig 8 shows comparison between model calculations and daily measured particle concentrations at Lidingö, in Huddinge and Solna. The correlation between the model and the measurements is fairly good with a r-square value of 0.61. The simulated mean concentration is 6.1 ± 0.6 µg m^{-3}, which can be compared with a measured mean concentration of 9.0 ± 1.4 µg m^{-3} (95% confidence interval). Table 9 presents measured and modelled mean particle concentrations as well as their correlation for the different sampling points and periods. In November there is a good agreement between the model and the measurements with similar concentration levels and high correlation (not Huddinge). In April, the comparison shows that the calculated concentrations are much lower than the measured ones. In particular, the model underestimated the PM10 concentrations. As in November, the measured and modelled PM2.5 concentrations in Huddinge show poor correlation.

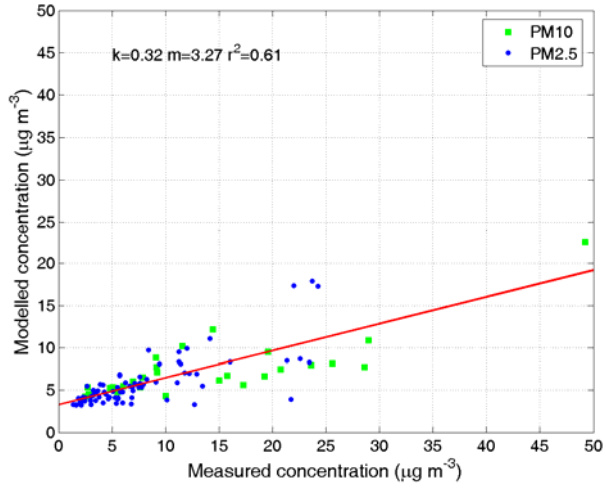

Fig 8. Comparison between modelled and measured PM2.5 and PM10 concentrations at Lidingö, in Solna and Huddinge during November 2004 and April 2005. The standard deviations of the slope (k) and the y-intercept (m) of the regression line are 0.02 and 0.29 µg m^{-3}, respectively.

Table 9. Measured and modelled PM2.5 and PM10 mean concentrations, standard deviations and r-square values at Lidingö, in Solna and Huddinge in November 2004 and April 2005.

		November 2004					April 2005				
		obs (μg m^{-3})		model (μg m^{-3})			obs (μg m^{-3})		model (μg m^{-3})		
		mean	std	mean	std	r^2	mean	std	mean	std	r^2
PM2.5	Lidingö	4.4	2.9	4.9	2.0	0.70	8.6	6.4	6.2	3.4	0.68
	Solna	4.7	3.9	5.5	2.5	0.88	9.3	6.4	6.6	3.6	0.75
	Huddinge	6.0	4.4	4.9	2.0	0.40	11.3	6.8	6.3	3.4	0.41
PM10	Lidingö	7.2	3.5	6.5	2.4	0.66	20.3	11.2	8.3	4.4	0.78

The two sampling sites at Lidingö and in Huddinge, respectively, are both situated in quiet residential areas far from busy roads. In contrast, the sampling site in Solna is located at the roof of one of the buildings at the Karolinska University Hospital area near the heavily trafficked road Norra Länken, with around 80000 vehicles per day. Fig 9 shows daily PM2.5 concentrations at Lidingö, in Huddinge and Solna in November 2004 and April 2005, respectively. In view of the locations of the measurement sites, the high PM2.5 concentrations obtained the $17^{th}/18^{th}$ of November (Wednesday) and the $17^{th}/18^{th}$ of April (Sunday) in Huddinge are probably due to some local non-typical process. Excluding these two deviating samples, the r-square value between measured and modelled PM2.5 in Huddinge is increased to 0.70 and 0.68 in November 2004 and April 2005, respectively.

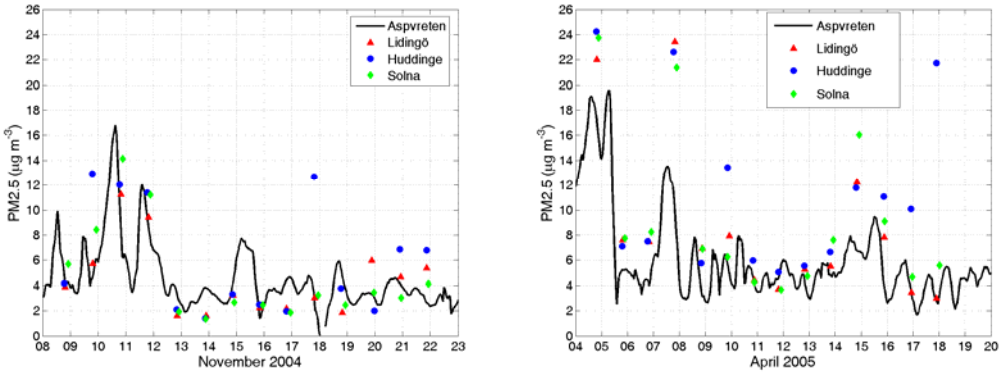

Fig 9. The solid line denotes time-series of hourly PM2.5 concentrations at the background station Aspvreten in November 2004 and April 2005. The triangles, circles and diamonds denote measured PM2.5 concentrations at Lidingö, in Huddinge and in Solna, respectively.

Hourly time-series of NO_x, NO_2 and PM10

To validate the dispersion model, the simulation results were also evaluated against hourly measurements at monitoring stations at Torkel Knutssonsgatan and Rosenlundsgatan at Södermalm, operated by SLB analys.

NO_x and NO_2

Fig 10 shows time-series of measured and modelled NO_x concentration at Torkel Knutssonsgatan during May 2001 to December 2002. The calculation area is Greater Stockholm with a resolution of 500 x 500 m. On average there is a good agreement between model calculations and observations, but for some hours the two times-series show large differences. The modelled mean NO_x concentration is 22.8 ± 0.4 μg m^{-3}, which can be compared with an observed concentration of 22.9 ± 0.4 μg m^{-3} (95% confidence interval). Fig 11 shows modelled NO_x as function of measured concentrations. The correlation calculation gives a r^2-value of 0.55.

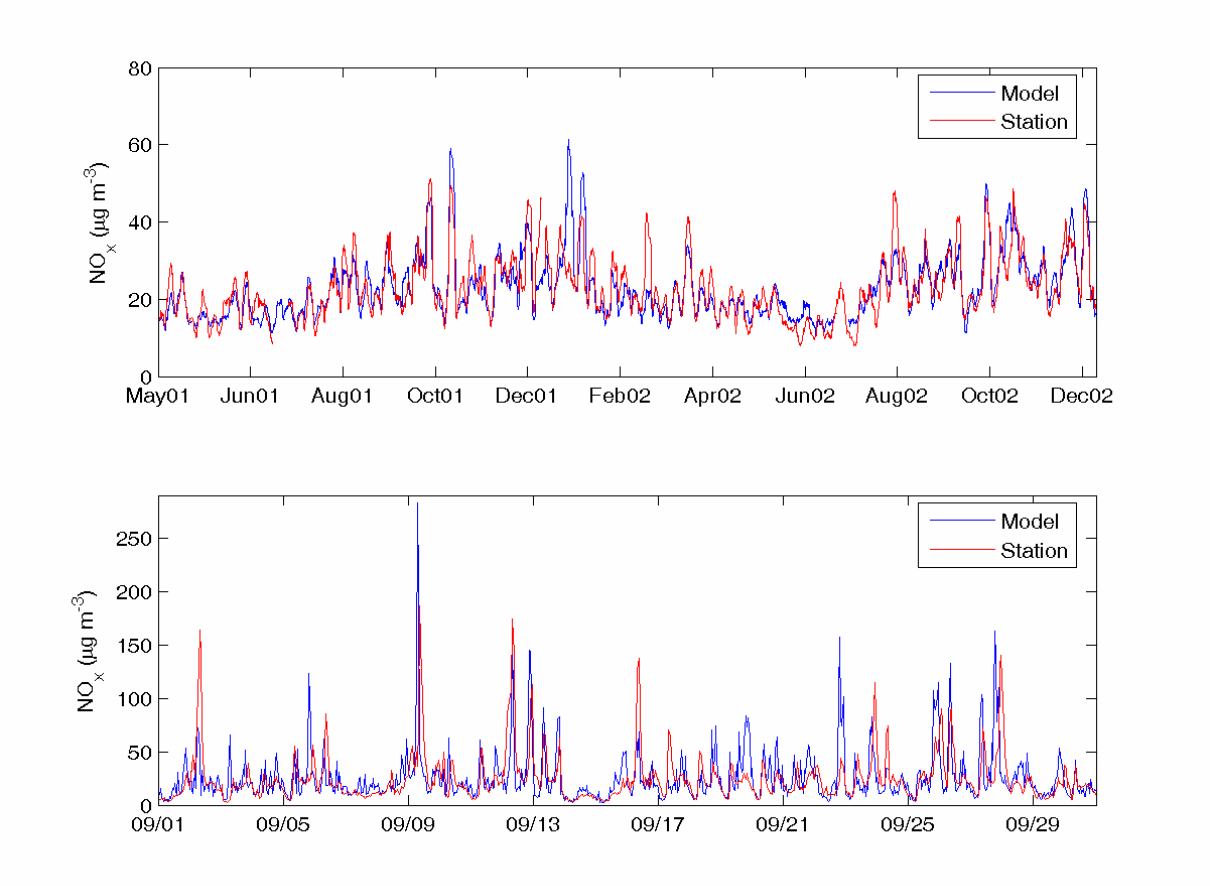

Fig 10. Time-series of modelled and observed NO_x at Torkel Knutssonsgatan. The upper panel shows 96-hour moving averages of hourly NO_x concentrations during May 2001 to December 2002. The lower panel shows hourly NO_x concentrations during September 2002.

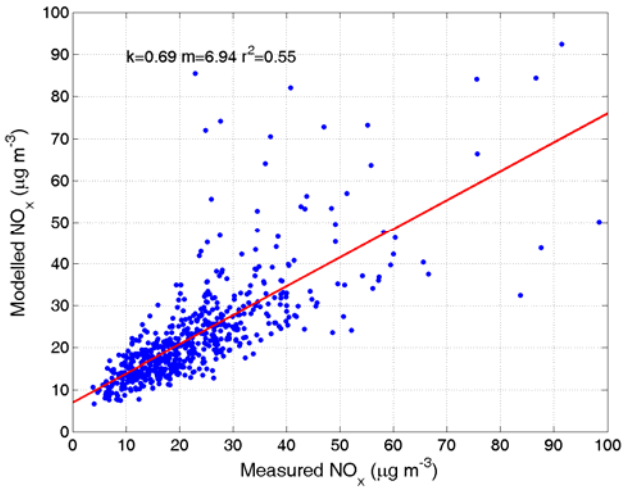

Fig 11. Comparison between modelled and observed NO_x at Torkel Knutssonsgatan during May 2001 to December 2002. The dots denote diurnal averages. The standard deviations of the slope (k) and the y-intercept (m) of the regression line are 0.03 and 0.67 µg m^{-3}, respectively.

Fig 12 shows measured and modelled NO_2 concentrations at Torkel Knutssonsgatan for the same period as in Fig. 10. The modelled and observed mean NO_2 concentrations are 17.2 ± 0.2 µg m^{-3} and 17.7 ± 0.2 µg m^{-3}, respectively (95% confidence interval). Fig 13 shows modelled NO_2 concentration as function of diurnal mean NO_2 levels at Torkel Knutssonsgatan. There is an even better agreement between simulations and observations of NO_2 compared to NO_x - the slope of the regression line is 0.60 and the r^2-value is 0.65. Using the method of Johansson and Forsberg (2005; based on photolytic NO_x and ozone reactions) to calculate the NO_2 concentration,

the modelled mean NO_2 concentration is 15.3 ± 0.3 μg m^{-3} (95% confidence interval) and the r^2-value of the observed and modelled diurnal averages is 0.47.

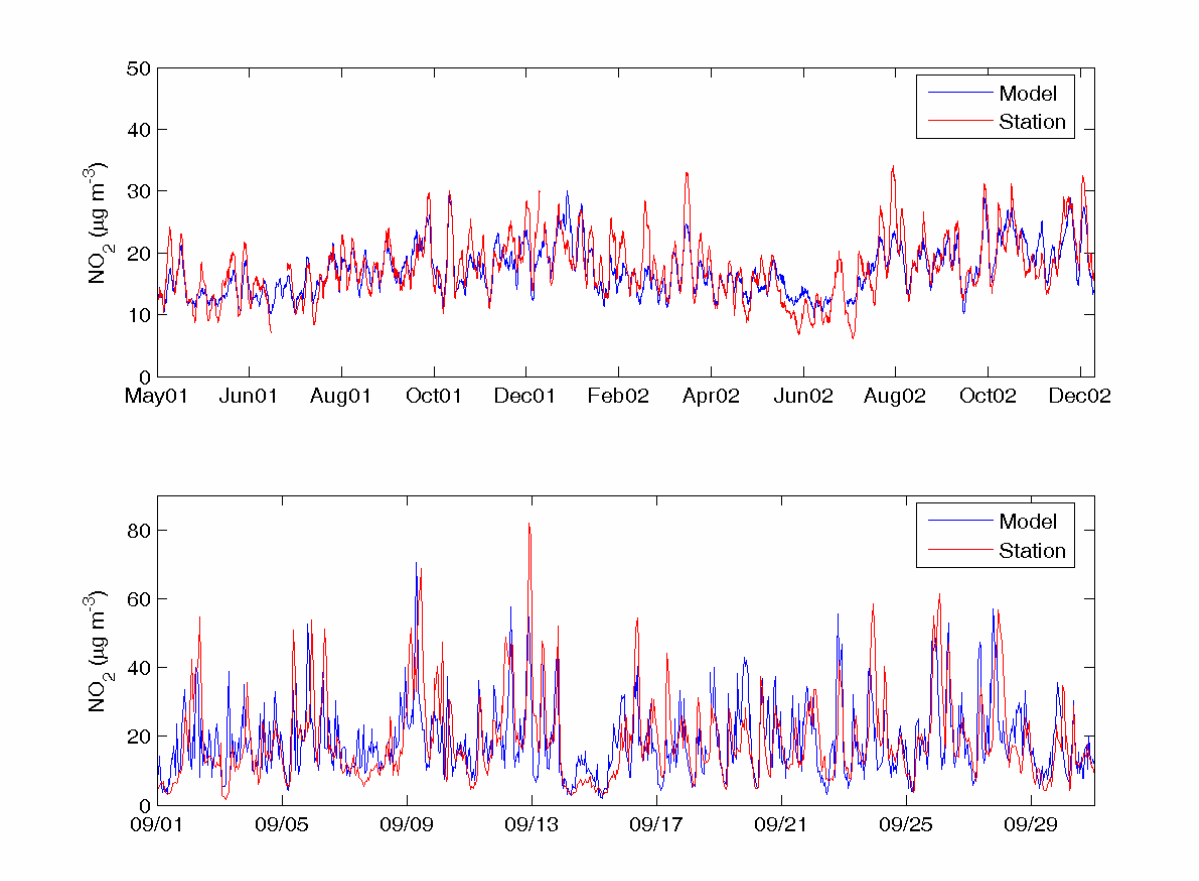

Fig 12. Same as Fig 10 but for NO_2.

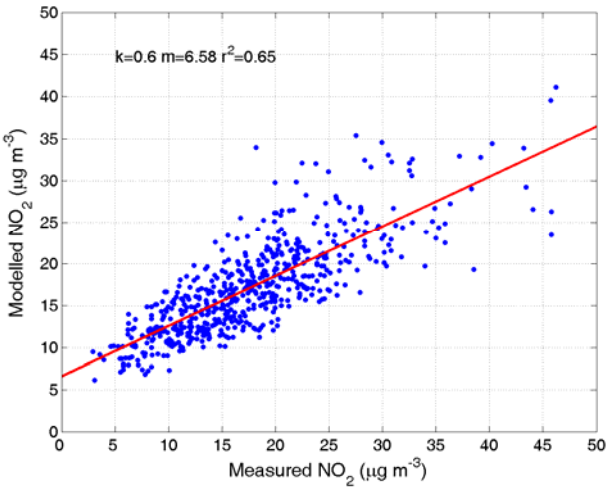

Fig 13. Same as Fig 11 but for NO_2. The standard deviations of the slope (k) and the y-intercept (m) of the regression line are 0.02 and 0.35 μg m^{-3}, respectively.

To validate the model further, the diurnal variation of simulated and measured concentrations were compared. In Fig 14 the diurnal variation of NO_x and NO_2 at Torkel Knutssonsgatan is presented. The model systematically overestimates the NO_x concentration during afternoon rush hour traffic. The same deviation (but smaller) is seen for the NO_2 simulations. One explanation of this deviation may be that the variation of the traffic volume is not correctly described in the emission database, i.e. the traffic load in the afternoon is overestimated. However, comparing the database with traffic measurements made at Hornsgatan a good agreement was found. Another explanation may be that there are deficiencies in the model's parameterization of mixing processes in the planetary boundary layer. For example, if the mixing is underestimated in the afternoon the diffusion of the emissions will be underestimated, resulting high concentration of pollutants.

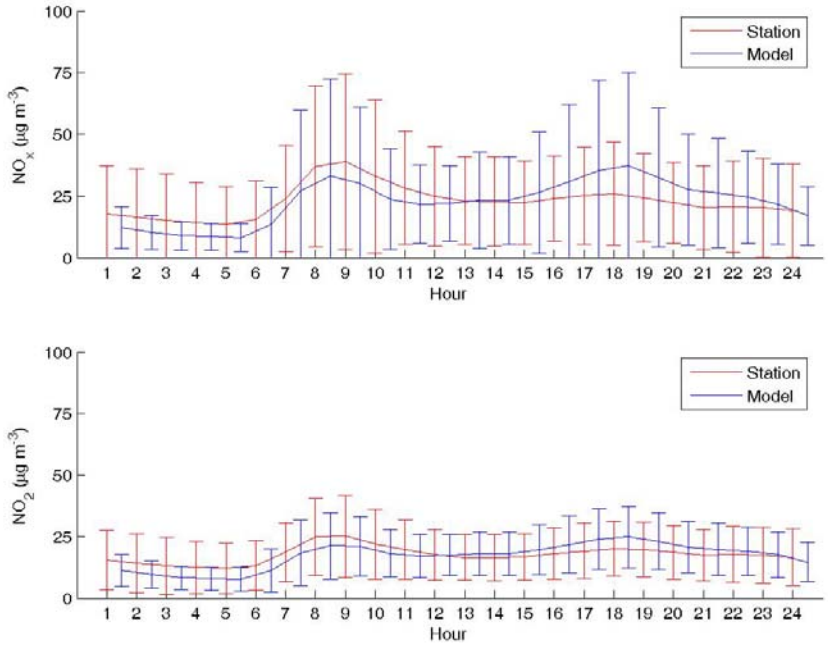

Fig 14. Diurnal variation of modelled and measured NO_x (top panel) and NO_2 (lower panel) concentrations at Torkel Knutssonsgatan during May 2001 to December 2002.

PM10

Fig 15 shows modelled and measured time-series of PM10 at Rosenlundsgatan during October 2004 to April 2005. The calculation area is Greater Stockholm with a resolution of 500 x 500 m. During autumn and winter there is generally a good agreement between the model calculations and the observations, whereas in April-May the model largely underestimates the PM10 concentrations. This poor correlation between measured and modelled PM10 in spring is due to difficulties describing the re-suspension process of road wear particles in the dispersions model. The modelled mean PM10 concentration is 10.6 ± 0.1 µg m^{-3}, which can be compared with an observed concentration of 15.0 ± 0.2 µg m^{-3} (95% confidence interval). The calculated r^2-value between modelled and observed diurnal PM10 averages is 0.29. Excluding data for periods when the road dust emissions are high, i.e. April and May, a better agreement between modelled and measured PM10 is obtained; with mean concentrations of 10.3 ± 0.1 µg m^{-3} and 11.7 ± 0.2 µg m^{-3}, respectively (95% confidence interval) and a r^2-value of 0.62. Fig 16 shows comparison between modelled and observed PM10 at Rosenlundsgatan during October 2004 to April 2005.

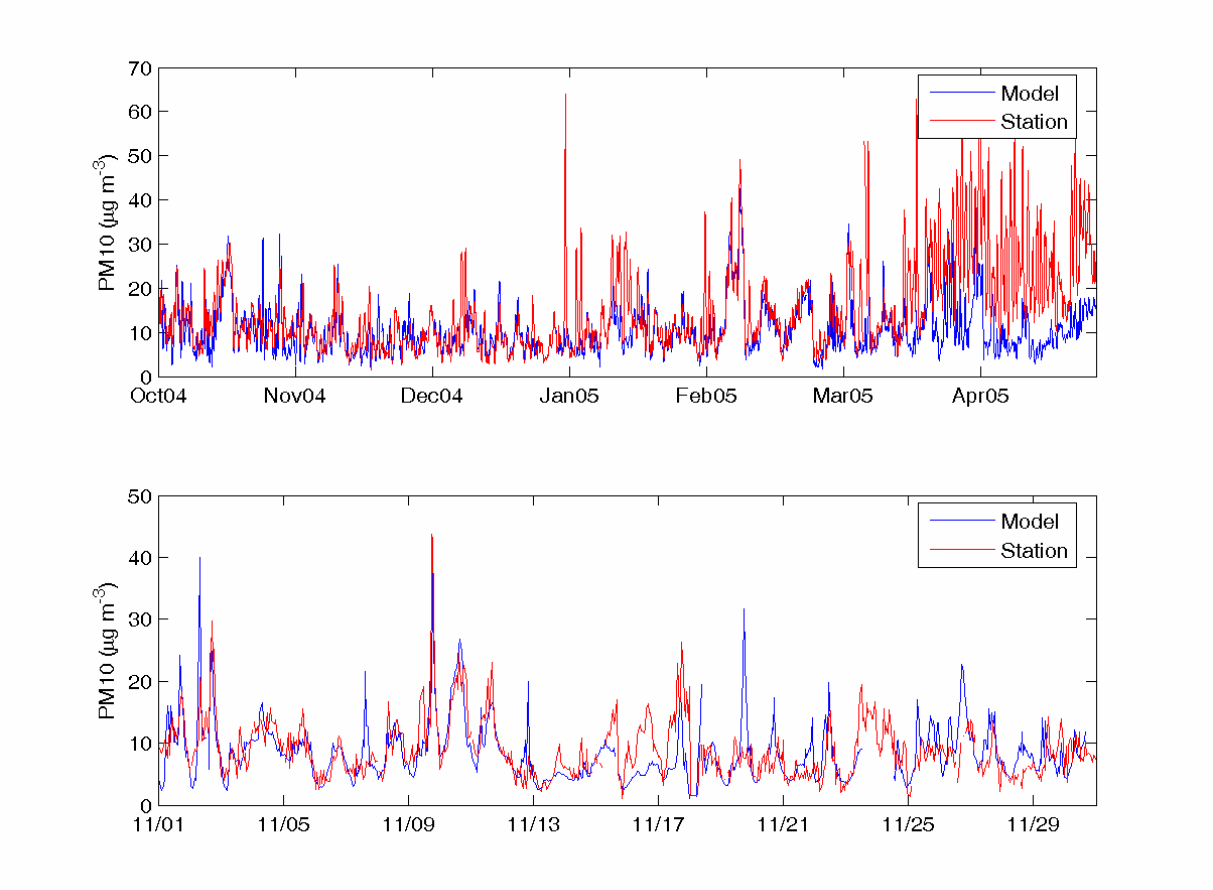

Fig 15. Time-series of modelled and observed PM10 at Rosenlundsgatan. The upper panel shows 6-hour moving averages of hourly PM10 concentrations during October 2004 to April 2005. The lower panel shows hourly PM10 concentrations during November 2004.

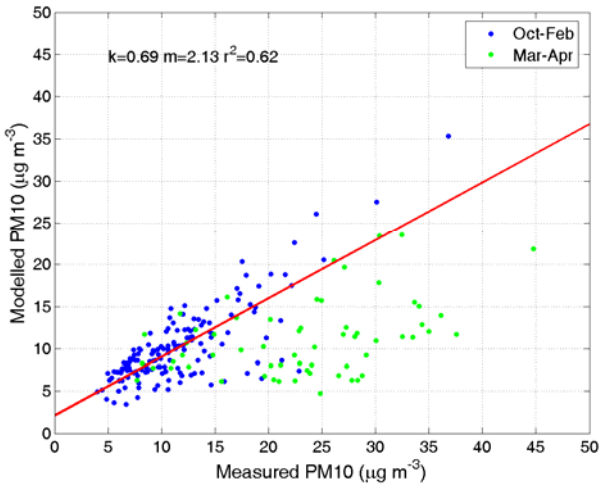

Fig 16. Comparison between modelled and observed PM10 concentrations at Rosenlundsgatan during October 2004 to April 2005. The blue dots denote diurnal averages in October to February. The green dots denote diurnal averages in March to April. The regression calculation only includes October-February data. The standard deviations of the slope (k) and the y-intercept (m) of the regression line are 0.04 and 0.58 µg m^{-3}, respectively.

In November the background concentration of particles represent most of the observed PM10 concentration in Stockholm (see Table 10). In contrast, in spring the background concentrations of PM10 are much lower than those observed in Stockholm, i.e. most of the particles are locally produced. It is clear that without including road dust generation, the model is unable to reproduce the high springtime concentrations of PM10. For NO_x, the local emissions dominate over background concentrations all year round (see Table 10).

Table 10. Mean concentrations of PM10 and NO_x from measurements at Rosenlundsgatan/Torkel Knutssonsgatan (station), Aspvreten/Norra Malma (background) and simulations at Rosenlundsgatan/Torkel Knutssonsgatan (model) in November 2004 and April 2005.

	Nov 2004 PM10 ($\mu g\ m^{-3}$)	Nov 2004 NO_x ($\mu g\ m^{-3}$)	Apr 2005 PM10 ($\mu g\ m^{-3}$)	Apr 2005 NO_x ($\mu g\ m^{-3}$)
Station	8.8	27.3	26.9	19.4
Background	5.3	4.2	8.7	2.6
Model (excl. background)	4.4	24.3	3.4	19.0
Model (incl. background)	9.8	27.9	12.1	21.7

Conclusions

From the comparison between dispersion model calculations and fixed monitoring measurements of particulate matter, NO_2 and benzene at a number of different sites in the Stockholm area we arrive at the following conclusions:

There was generally a good correlation between modelled and measured NO_2, with a calculated r^2-value of 0.71. However, the model gave slightly higher NO_2 concentrations than the passive samplers, $25.0 \pm 1.0\ \mu g\ m^{-3}$ compared to $21.4 \pm 0.7\ \mu g\ m^{-3}$. This result indicates that even though NO_2 concentrations depend not only on NO_x emissions, but also on photochemical processes involving ozone, Gaussian models without photochemistry can be used to calculate NO_2 concentrations.

For benzene, the discrepancy between measured and calculated concentrations was relatively large. The r^2-value was 0.34 and the simulated and measured mean concentrations were $1.2 \pm 0.3\ \mu g\ m^{-3}$ and $1.5 \pm 0.3\ \mu g\ m^{-3}$, respectively. Both the model calculations and the measurements have significant uncertainties. The emission factors used in the model include poor knowledge of the importance of cold starts emissions and evaporative losses. Diffusive sampling of benzene is connected with uncertainties depending on the type of absorbent that is used.

The correlation between modelled and measured PM2.5 and PM10 was fairly good with a r^2- value of 0.61. The simulated mean concentration was $6.1 \pm 0.6\ \mu g\ m^{-3}$, which can be compared with a measured concentration of $9.0 \pm 1.4\ \mu g\ m^{-3}$. The main uncertainty in the model calculations is the local road dust emissions. These emissions are especially important during dry road conditions in late winter and spring. The importance of road dust emissions also vary with the share of studded tyres. In Stockholm, where around 70% of the cars have studded tyres, road dust emissions is probably more important than in for example Malmö, with around 40% studded tyres. In addition, there are large variations during the year and between cities in the importance of local sources compared to long distance transport. In Stockholm, the contribution from the road dust emissions to the PM10 concentrations can be ten times higher than the contribution from exhaust particle emissions. In November, when the local road dust emissions are low, our model simulations generally showed high correlation with the measurements. In contrast, due deficiencies in the model's parameterization of road dust emissions, the same model calculations for April largely underestimated the ambient PM10 concentrations.

Acknowledgements

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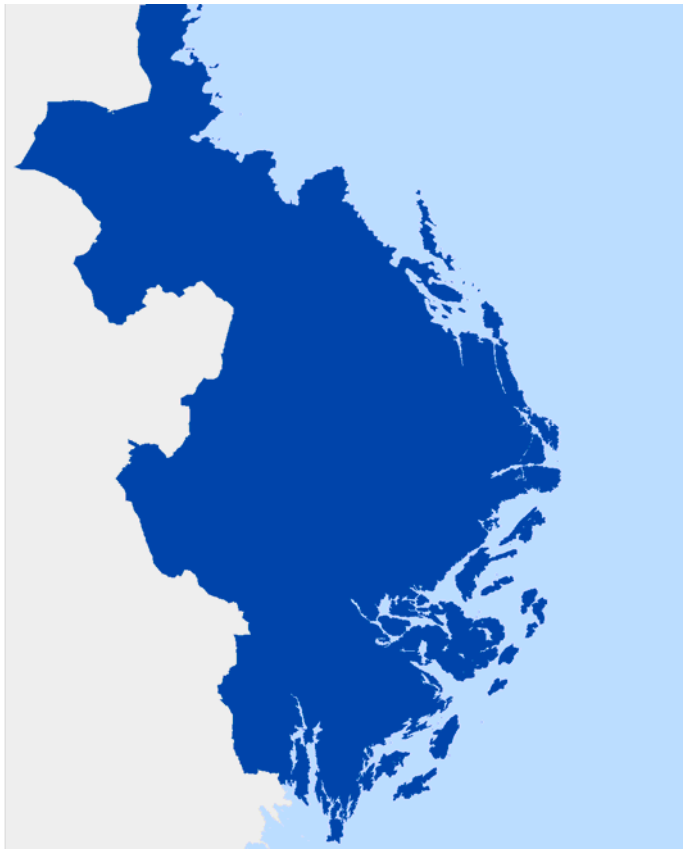

Stockholms- och Uppsala Läns Luftvårdsförbund är en ideell förening. Medlemmar är 33 kommuner, länens två landsting samt institutioner, företag och statliga verk. Samarbete sker med länsstyrelserna i länen. Även Gävle och Sandvikens kommuner är medlemmar. Målet med verksamheten är att samordna arbetet vad gäller luftmiljö i länen med hjälp av ett system för luftmiljöövervakning, bestående av bl a mätningar, emissionsdatabaser och spridningsmodeller. SLB-analys driver systemet på uppdrag av Luftvårdsförbundet.

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