Chapter 5: Particulate Matter in Urban Air

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5.1 Introduction

Research within SATURN has focussed on improving our understanding of the physical and chemical properties of particulate matter, and our capability for modelling particulate matter concentrations in urban areas. Campaigns have been conducted at various urban centres on PM_{10}, $PM_{2.5}$ and on finer size fractionated samples. Measurements have been made to reveal the chemical composition, and particle number and mass concentrations at various urban locations. Models have been developed, evaluated and applied in order to predict aerosol processes, and the number and mass concentrations, of the coarse and fine fractions in urban areas.

5.1.1 The reasons for investigating urban particulate matter

Studies of long-term exposure to air pollution, especially to particulate matter (PM), suggest an increased mortality, increased risk of chronic respiratory illness, and of developing various types of cancer. World Health Organisation (WHO

1999) has estimated that in Europe air pollution has caused 168.000 (range of estimate 100.000 – 400.000) excess deaths annually; in the United States the corresponding figure has been estimated to be approximately 100.000. The best estimate on the reduction in life expectancy in Central Europe is about 1 year.

Fine PM particularly may be causing a significant burden of disease and excess deaths in Europe and North America. However, it is not known, which chemical and physical characteristics of the PM are responsible for these effects, and which source categories are responsible for the most harmful exposures.

The chemical and physical properties of PM are important for assessing environmental impact and deposition in the lungs as well as adverse health effects (Harrison et al. 1999; Morawska et al. 1999; Kleeman et al. 2000). In addition to size and number concentration, important properties include state (liquid/ solid), volatility, hygroscopicity, chemical composition (content of organics, metals, salts, acids etc.), morphology and density. These properties also need to be taken into account in selecting methods for PM emission regulation and control. A reliable assessment of urban PM pollution and the subsequent adverse health effects is therefore crucial in terms of the promotion of public health (WHO 2000).

5.1.2 Review of experimental field campaigns on particulate matter in SATURN

Several experimental campaigns were carried out within the framework of SATURN during the past years. Different European urban areas were investigated in Denmark, Finland, France, Greece, Hungary, Sweden and the United Kingdom. Field studies aimed mainly at investigating particle size distributions and their chemical properties. Key objective of the studies was to conduct sufficiently detailed measurements in order to enable source apportionment investigations and characterisation of particles, including the fine and ultrafine particle fractions.

Measurement campaigns of particulates were carried out in Copenhagen at streets, urban background and regional background in order to characterise urban PM, including size distributions as well as chemical composition and physical properties. Special measurements of ultra-fine particles were carried out in order to estimate the emission and number size distribution of the actual car fleet, separately for diesel and petrol vehicles (e.g., Wåhlin et al. 2001a).

In the Helsinki metropolitan area, virtual and Berner low-pressure impactors were applied to collect aerosol samples. Size distribution and chemical composition of the particles were analysed (e.g., Pakkanen et al. 2001a, b, c). In Budapest, source profiles of toxic elements were determined and the source-receptor relationships were analysed by the chemical mass balance method (e.g., Bozó et al. 2001).

A field measurement campaign was performed in urban air and in a road tunnel in Stockholm. The tunnel experiment has provided source profiles and emission factors for gasoline and diesel vehicles. Both particulate species such as metals, polycyclic aromatic hydrocarbons (PAH), elemental and organic carbon and gases (NO, NO_2, CO and several volatile hydrocarbons) were measured in the tunnel (e.g., Kristensson et al. 2001). The same set of compounds was also measured in an urban ambient air campaign.

Sampling campaigns were carried out in Hatfield, UK by the University of Hertfordshire in the summer of 2000 and spring 2001. Gravimetric measurements at various size fractions were coupled with detailed elemental analysis. The aim of their research was to better understand the mass and chemical distributions of size fractionated aerosols. Sampling for these campaigns was conducted at a semi-urban roof top site (Sokhi et al. 1999 and et al. 2001).

Research at Imperial College on source apportionment has used modelling of PM_{10} concentrations across London to differentiate contributions from local major roads, emissions from central, inner and outer London, imported secondary particulate contributions from longer range transport, and the coarser fraction between 2.5 and 10 μm. This has been used to assess the benefits of a range of potential future scenarios involving both technological and traffic reduction measures.

5.1.3 Review of modelling particulate matter in SATURN

Mathematical models have proved to be very useful for a range of air quality applications. Examples of these and, in particular those related to particulate matter, can be found in Sokhi et al. (2000b) and Sokhi and Bartzis (2002). Within the framework of SATURN, a range of aerosol process, emission and dispersion models have also been developed in order to evaluate urban PM concentrations.

The Danish street pollution model and the urban background model were refined to include treatment of particulate matter. The application of inverse modelling has led to estimates of emission factors of particles from the Danish car fleet under real life driving conditions. The emission factors comprising size distributions and particle composition are important as input to air quality and exposure models.

In Finland, the aerosol dynamical model MONO32 was applied; this model has been originally developed at the University of Helsinki by Pirjola and Kulmala (2000). The model takes into account gas-phase chemistry and aerosol dynamics, i.e., the processes of nucleation, coagulation, condensation, evaporation and deposition. The particles are classified into four different size modes, which are assumed to be monodispersive. The first objective was to evaluate quantitatively the impact of various chemistry and aerosol processes on aerosol evolution (Pohjola et al. 2003).

Based on road tunnel measurements of particle size distributions in Stockholm, an aerosol dynamical module has been developed to study the dynamics of different particle sizes in traffic tunnels and other heavily trafficked urban environments, such as, e.g., street canyons (Gidhagen et al. 2002). The road tunnel represents a well-controlled environment in order to produce good-quality data for model evaluation. In addition, tunnel data may be used to determine vehicle emission factors and to investigate aerosol dynamics and dispersion.

A fairly simple model was developed for predicting the concentrations of $PM_{2.5}$ in urban areas (Tiitta et al. 2002). This model also includes a method for evaluating the regionally and long-range transported fine particle fraction (Karppinen et al. 2002). The model was tested against the results from a measurement campaign in a suburban environment near a major road in Kuopio, Central Finland. The mass concentrations of fine particles ($PM_{2.5}$) were measured simultaneously at four distances from a major road, together with traffic flows and relevant meteorological parameters.

The US EPA Gaussian plume (CALINE 4) and ADMS-Urban models were employed to calculate PM_{10} concentrations near to a major motorway (M25) encircling the City of London. The model was used to calculate 24 hour and annual means of PM_{10}; a comparison with measurements for 1996 showed a close agreement (Sokhi et al. 1998 and 2000a). The study showed that roughly half of the modelled PM10 concentrations could be attributed to the road traffic contribution.

A semiempirical model has been developed for evaluating the PM_{10} concentrations in urban areas (Kukkonen et al. 2001a). The basic model assumption is that local vehicular traffic is responsible for a substantial fraction of the street-level concentrations of both PM_{10} and NO_x, either due to primary emissions or vehicle driven material from street surfaces. The model performance was evaluated against the measured PM_{10} data from five air quality stations in the Helsinki area in 1999.

5.1.4 Review of investigations on exposure to particulate matter in SATURN

The exposure of road-users to $PM_{2.5}$ was studied both experimentally and by modelling in London (e.g., Adams et al. 2001a, b). One of the main results was that people moving along city streets are exposed to levels of pollution that are not only substantially higher than urban background fixed-point measurements, but also higher than roadside and kerbside measurements. In Helsinki, a detailed population exposure model was developed that can be utilised for evaluation of the whole urban population to PM (Kousa et al. 2002).

5.2 Sources and emissions of particulate matter

5.2.1 Review of various emission sources of urban particulate matter

Road traffic is a major source of particles in urban air in most European cities. Traffic related particulate matter is not only emitted directly from the vehicle exhaust but also includes particles from wear on road, tires and brakes, and airborne dust from road surfaces.

As an example, one can consider the contributions to measured concentrations of $PM_{2.5}$ at a roadside measurement location originating from various source categories. The total measured concentration of $PM_{2.5}$ can be written as (modified from Tiitta et al. 2001; Kukkonen et al. 2001a):

$$PM_{2.5} = PM_{2.5}^{tr,e} + PM_{2.5}^{tr,n\text{-}e} + PM_{2.5}^{st} + PM_{2.5}^{bg,urb} + PM_{2.5}^{bg,lrt} + PM_{2.5}^{wind} \quad (5.1)$$

where the superscripts 'tr,e' and 'tr,n-e' refer to the primary (exhaust) and non-exhaust contributions of vehicular traffic from the nearest roads and streets, respectively. The superscript 'st' refers to stationary sources, and the superscripts 'bg,urb' and 'bg,lrt' refer to the urban, and regionally and long-range transported (LRT) background, respectively. The superscript 'wind' refers to wind-driven PM from various surfaces (excluding non-exhaust vehicular traffic emissions). Equation (5.1) is useful for illustrating the multiple source types of urban airborne PM. Clearly, this is not the only option in order to categorise various emission sources.

5.2.2 The relative importance of various particulate matter emission sources

The particles may grow by coagulation of primary particles, and by condensation of gases on particles. The fine particles (accumulation mode in the range from 0.1 to 2 μm) are typically formed by chemical reactions (e.g., SO_2 and NO_x to form sulphate and nitrate), or other relatively slow processes in the atmosphere; the fine PM are therefore commonly aged particles. The coarse particle mode can be defined as the particles with diameters larger than 2.5 μm, which in urban areas are formed typically mechanically by abrasion of road material, tyres and brake linings, construction works, soil dust raised by wind and traffic turbulence.

In the UK, there are numerous sources of fine particles that are recognised in urban centres (QUARG 96, APEG 1999); these include contributions from natural sources, road transport, stationary combustion and industrial processes. Road transport nationally can contribute about 25 % of PM_{10}, whereas in urban centres the contribution rises to approximately 80-90 % (QUARG 96, APEG 1999). The coarse fraction (particles with aerodynamic diameters from 2.5 to 10 μm) tend to consist mainly of soil, sea salt, biogenic and airborne dust components, whereas

the fine fraction (diameters below 2.5 μm) consists of components from road vehicles (mainly diesel) and stationary combustion processes, and secondary aerosols (such as sulphates, nitrates and ammonium) as well as transboundary contributions.

In Nordic countries, atmospheric long-range transport constitutes an important part of the total urban background $PM_{2.5}$ concentration (e.g., Johansson et al. 1999; Pakkanen et al. 2001b, c; Karppinen et al. 2002). In urban areas in Sweden, it even constitutes the most important single contribution to both urban background $PM_{2.5}$ and PM_{10}, which is clearly illustrated by the spatially uniform urban background concentrations of these particulate fractions (Areskoug et al. 2000).

The long-range transported aerosol dominates the accumulation mode of the aerosol size distribution even in urban areas, whereas its contribution to the ultrafine and coarse mode is small, in comparison to that of the local particle emissions. For cities in Nordic countries, the coarse particle fraction is important for the elevated PM_{10} concentrations, especially in spring (e.g., Johansson 1999; Pohjola et al. 2000 and 2002).

Like in other urban areas, in Budapest, Hungary, the highest PM_{10} concentrations tend to occur in the vicinity of major urban roads (Bozó et al. 2001), indicating that local traffic is mainly responsible for the elevated coarse particle concentrations. The amount of major industrial pollution sources has substantially decreased in urban areas in Central Eastern Europe during the last decade. In Budapest, heat energy production is based mainly on natural gas, regarding both thermal power plants and domestic heating.

As stated earlier, a major contribution to particulate pollution in urban areas is believed to be attributed especially to emissions from diesel-powered vehicles (Palmgren 2001). Ultra-fine particles emitted from petrol as well as diesel engines are formed at high temperature in the engines, in the exhaust pipe, or immediately after emission to the atmosphere. Some of these particles may be in the so-called nucleation mode (nanoparticles < 50 nm). The dominating ultrafine particle mode has a number concentration peak in the range of 20-50 nm. The ultrafine particles in urban air are mainly emitted from diesel and petrol fuelled vehicles.

In Copenhagen, hourly elemental and organic carbon (EC/OC) measurements of PM_{10} were carried out in a busy street, related to measurements of other traffic-originated pollutants. The statistical correlations between EC and CO and also OC and NO_x /CO were relatively low. However, a clear correlation between EC and NO_x was observed, indicating a significant contribution to EC from diesel traffic (Palmgren et al. 2001).

Measurement campaigns carried out in two cities in Denmark included monitoring of the ultrafine particles from traffic under normal driving conditions and in ambient air, in order to be able to establish the relationship between the sources and the exposure of the population. Measurements of ultrafine particles were carried out by a SMPS (Scanning Mobility Particle Sizer) with a high time resolution that corresponds to the variation in traffic and meteorological variables.

The particles were separated in 29 size fractions, ranging from 0.01 to 0.7 µm. It was shown that particles originating from diesel engines include a large nanoparticle fraction (< 30 nm). These very small particles are probably mainly droplets, which are formed in or immediately after the tailpipe. The sulphur content plays a key role as condensation nuclei for condensation of fuel, lubrication oil and volatile combustion products. The particle number concentration may therefore depend strongly on the sulphur content of the diesel oil consumed (Fig. 5.1).

Particles from diesel as well as petrol engines comprise also solid, mainly carbonaceous, particles, which may have a crucial role regarding the adverse health effects. The measurements of ultrafine particles in Copenhagen have indicated this mode of the particle size distribution (Wåhlin et al. 2001b).

The results from Stockholm (Johansson *et al.* 2001) and Nantes (Despiau *et al.* 2001)suggest that local traffic is the main source of ultra-fine particles, and most of the particles close to traffic are in the range from 3 to 30 nm. The size distributions change rapidly with distance from the traffic sources. At the rooftop location, the size distributions shift towards larger sizes compared with those in the street level.

In residential areas of North European cities, biomass burning for domestic heating may be an important source of fine particles during winter. For instance in Sweden wood is most frequently burned in boilers constructed for multiple energy sources (oil, wood and electricity). Recently, low emission boilers have been introduced into the market, but stoves have come into use as an additional heating device, commonly also in urban areas.

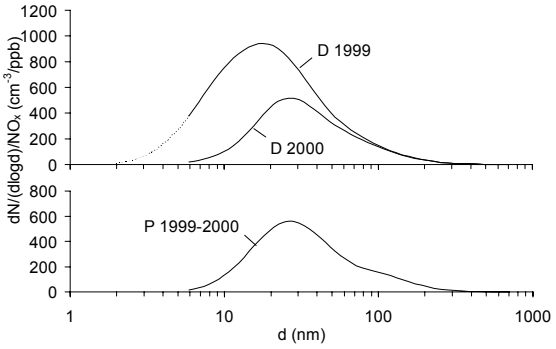

Fig. 5.1. The number size distributions in the winters 1999 and 2000, respectively, at Jagtvej in Copenhagen before and after the reduction of the sulphur content in diesel fuel in Denmark in July 1999 (Wåhlin et al. 2001b). The upper graphs are diesel distributions, which showed a pronounced shift (probably mostly due to diesel cars with oxidising converters), and the lower graph is a distribution which did not change (probably due to a combination of petrol and diesel traffic)

Emissions from wood combustion may be important for PM, but also for many particle-bound hydrocarbons, such as polycyclic aromatic hydrocarbons. The aerosol originated from biomass combustion may substantially differ from that originating from vehicle exhausts, both regarding chemical composition and particle size distribution. For instance, PM from biomass burning may contain a larger fraction of hygroscopic particles (Hedberg et al. 2002).

5.2.3 Evaluation of the vehicular emissions of particulate matter

The assessment of the emission of pollutants is normally based on dynamometer studies with different driving cycles for a few vehicles. Dynamometer cycles are essential in order to establish uniform emission standards for regulatory purposes and for testing of new technologies. However, such measurements do not reflect the real driving conditions and the level of maintenance of the actual vehicle fleet.

The control parameter is normally the mass of the particles that will be dominated by the coarse particulate fraction. The mass includes often only the solid part of the particles, which are measured after heating of the exhaust gas to, e.g., 300°C; this will remove semi-volatile compounds, e.g., organic condensates, which could nevertheless be present in the urban air. There is therefore a need for on-road emission estimates of air pollutants from the actual fleet.

The emission factors for particle distributions from different vehicle categories can also be estimated by inverse modelling. These estimates are based on time series of particle spectra from streets and urban background locations, as well as detailed traffic data and local meteorology at rooftop level.

The model CAR-FMI (e.g., Kukkonen et al. 2001b) was extended to allow for the emissions and dry deposition of PM. The vehicular $PM_{2.5}$ emissions were modelled to be dependent on vehicle travel velocity (ranging from 0 to 120 km h^{-1}), separately for the main vehicle categories. Light-duty vehicles were classified into three categories: (i) gasoline-powered cars and vans without a catalytic converter; (ii) gasoline-powered cars and vans equipped with a catalytic converter; and (iii) diesel-powered cars and vans. Similarly, heavy-duty vehicles were classified into four categories: (i) diesel-powered trucks with a trailer; (ii) diesel-powered trucks without a trailer; (iii) diesel-powered buses; and (iv) buses powered by natural gas.

Numerical correlations of PM emissions were fitted in terms of vehicle travel velocity, separately for each of the above-mentioned seven vehicle categories. These correlations are based on nationally conducted vehicle emission measurements (Laurikko 1998).

5.3 Spatial and temporal variation – temporal trends

5.3.1 Spatial and temporal variation of particulate matter in urban areas

To estimate the extent of impact at various urban centres, we need to understand the temporal and spatial variations of PM concentrations.

The main objective of the UK study was to investigate the temporal variation of PM_{10} and $PM_{2.5}$ concentrations at two London sites. Data includes measured PM concentrations (PM_{10} and $PM_{2.5}$) from Marylebone Road (roadside site) and Bloomsbury (urban centre site) for the period 1997-1999. Data was obtained from 'The UK National Air Quality Archive'.

Monthly variation of particulate concentrations at the two sites has been investigated, and correlation with other air quality and selected meteorological parameters has been analysed. Fig. 5.2 shows monthly variations of PM_{10} and $PM_{2.5}$ at these two sites. The correlation analysis suggests that PM_{10} and $PM_{2.5}$ have the highest correlation amongst each other and a highly significant correlation with other air quality parameters, such as CO and NO_x. This obviously confirms road traffic as one of the main sources. On average the PM concentrations were found to be about 40 % higher at the roadside site compared to the urban centre site, confirming the importance of particles from surrounding areas.

These two sites also show a moderate monthly variation for both years, except for the coarse fraction at the roadside site (1999), which shows a peak during the late summer and early autumn months. The peak is caused by high hourly concentrations (> 50 $\mu g/m^3$) of PM_{10} which were observed at the roadside site for about 10% of the time. A possible reason for the high coarse fraction concentrations could be the occurrence of generally dry conditions coupled with wind speed of around 4-5 m/s increasing the dust content in the local atmosphere. The coarse fraction is higher at the roadside site compared to the background site, indicating the importance of vehicle turbulence as a possible mechanism for in introducing this fraction into the local atmosphere.

Seasonal variation of the PM_{10} and $PM_{2.5}$ concentrations at some stations in the Helsinki Metropolitan Area during one selected year (1998) are presented in Fig. 5.3 (Pohjola et al. 2000, see also Pohjola et al. 2002).

The seasonal variation of the PM_{10} concentrations in Fig. 5.3 is very pronounced. The PM_{10} concentrations have maximum values in spring at all measurement stations; these are mostly caused by vehicle induced airborne material from street surfaces. The fine particulate matter ($PM_{2.5}$) concentrations also show a maximum in spring that may be caused by vehicle induced airborne material or unfavourable meteorological conditions, or both reasons. However, there is a substantial year-to-year variation; for instance during the previous year (1997), there was no clear seasonal variation of the monthly averaged $PM_{2.5}$ concentrations.

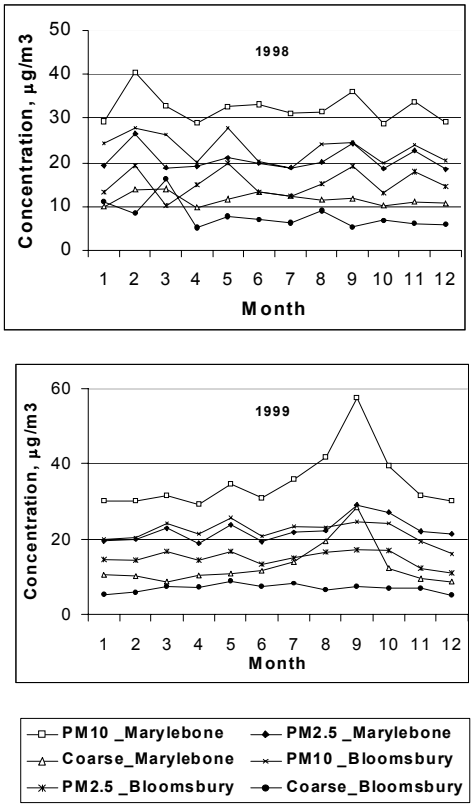

Fig.5.2. Monthly variation of PM_{10} and $PM_{2.5}$ at two London sites

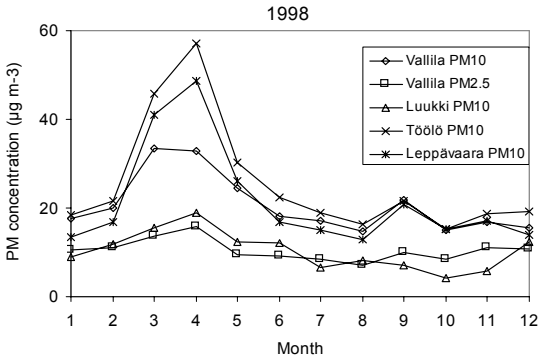

Fig. 5.3. Seasonal variation of PM_{10} concentrations (monthly averages) in Helsinki, at the stations in Vallila and Töölö (urban, traffic sites) and Leppävaara (suburban site), together with $PM_{2.5}$ concentrations at Vallila and regional background PM_{10} concentrations at Luukki (Pohjola et al. 2000)

In Stockholm, the highest PM_{10} levels are also observed during spring from March to April, caused mainly by vehicle induced airborne material from street surfaces. Most exceedances of the daily mean EU limit value for PM_{10} occur during the spring (Fig. 5.4). The spring maximum is particularly pronounced at the kerb sites close to traffic, but can also be seen at urban background sites. For fine particulate matter fraction, $PM_{2.5}$, there is almost no annual variation and the urban background concentration is almost the same as the rural background.

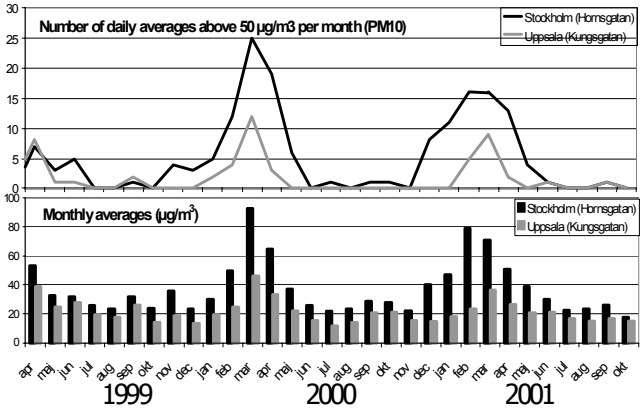

Fig. 5.4. Upper panel: Number of days with average PM_{10} concentration higher than 50 $\mu g/m^3$ (EU limit) in Stockholm (upper line) & Uppsala (lower line). Lower panel: Monthly average PM_{10} concentration at the same sites (Stockholm: black bars, Uppsala: grey bars)

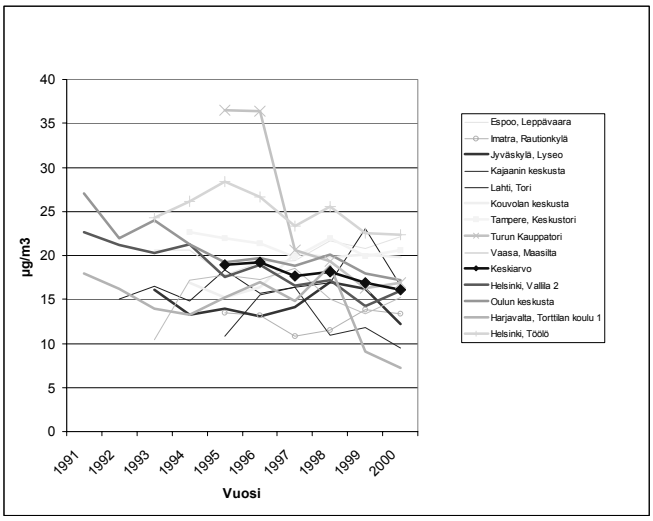

Fig. 5.5. The trends of annual average PM_{10} concentrations at various urban measurement stations located in 12 cities in Finland during the 1990's. The average of all stations is denoted as bold black line with squares (Pietarila et al. 2001)

5.3.2 Temporal trends of particulate matter in urban areas

The trends of annual average PM_{10} concentrations are presented in Fig. 5.5 for various urban measurement stations in Finland since 1991. The data includes urban measurement stations located in 12 cities. These trends show a substantial variability both from one station to another, and from year to year. However, the annual average value of all stations shows a slight decreasing trend. The measurement data of fine particulate matter, $PM_{2.5}$ is not yet sufficient for detailed analysis to reveal any reliable year-to-year trends.

Since the measurements started in 1994, the PM_{10} concentrations in Stockholm (Sweden) have shown no clear temporal trend. Urban background concentrations of PM_{10} in Stockholm range from 13.5 to 16 $\mu g/m^3$. While local vehicle exhaust emissions have probably significantly decreased during this period, due to the renewal of vehicle fleet and the introduction of cleaner fuels, they do not substantially contribute to the total PM_{10} concentrations. The main sources of PM_{10} in Stockholm are background air transported to the urban area and vehicle induced airborne material from street surfaces.

5.4 Field campaigns involving particulate matter

In SATURN, field measurement campaigns were conducted in Denmark, Finland, France, Greece, Hungary, Sweden and the United Kingdom. In the following, five of the above mentioned campaigns are described in a structured format that includes (i) major scientific issues to be investigated with the campaign, (ii) explicit relevance to SATURN aims, (iii) experimental set-up and (iv) main results.

5.4.1 Field measurement campaigns in Copenhagen and other cities in Denmark

Major scientific issues investigated (Copenhagen)

The objective was to measure and determine the particle emissions under normal driving conditions in ambient air, in order to establish the relationship between the sources and the exposure of the population (Palmgren et al. 2001).

The population spends most of the time indoors. The ambient (outdoor) air quality has a significant effect on indoor pollution levels (Lai and Nazaroff 2000). There is thus a strong need for quantifying the processes governing the particle composition and size distributions also in the indoor environment.

Relevance to SATURN aims (Copenhagen)

A four-year project on particle studies was initiated in 2000/2001. The main objectives are (i) to characterise the geographic and temporal variability in particle composition and size distributions in Danish ambient air, (ii) to determine particle emission factors for various vehicle categories, (iii) to determine indoor - outdoor relationships for buildings in busy streets and (iv) to develop the Danish air quality models for local (OSPM), urban (BUM) and regional scales to include particles and to validate the models for particles.

Experimental set-up (Copenhagen)

Most of the measurements in these studies were performed in central Copenhagen in a street canyon, Jagtvej, which is a 10 m wide main road and during rush hours in practise a 4 lane road. Both sides of the roadway have bicycle lanes and pavement. In addition, Jagtvej is lined on both sides by 5-6 storey buildings. The traffic density is approx. 26,000 vehicles per 24 hours, including 6-8 % heavy vehicles, i.e. buses, lorries and larger vans.

A fixed monitoring station of the Danish Air Quality Monitoring Programme has been in operation at this location for many years (Kemp and Palmgren 2000). Data from this station include half-hour measurements of NO_x (sum of the nitrogen oxides NO and NO_2), CO (carbon monoxide), and other traditional pollutants. In addition, 24 hours particle filter samples were collected of TSP (Total Suspended Particulates) and PM_{10}. Other measurement campaigns were performed at H.C. Andersen's Boulevard (approx. 60,000 vehicles per day), a street in Copenhagen. Some measurement campaigns were performed at Albanigade (22,000 vehicles per day, 12 % diesel), a street in Odense, a city of 180,000 inhabitants about 150 km west of Copenhagen. All three stations are included in the Danish Air Quality Monitoring programme with the above mentioned measurement programme. The urban background air pollution was measured at rooftop stations in the Danish Air Quality Monitoring programme.

A Scanning Mobility Particle Sizer (SMPS) was used to measure the fine and ultrafine particles. Campaigns of PM_{10} measurements with a time resolution of half an hour were carried out at Jagtvej in Copenhagen by Tapered Element Oscillating Microbalance. The measurements of the PM_{10} elemental and organic carbon (EC/OC) were performed using the automatic speciation analyser 5400 Ambient Carbon Particulate Monitor (R&P).

Studies of exchange of particles in outdoor/indoor air and the transformation took place in the street canyon, Jagtvej, in Copenhagen and in an apartment along the street. Four measurement campaigns were performed in 2001/2002.

Main results (Copenhagen)

The Constrained Physical Receptor Model (COPREM) has been used for the source apportionment. For the apportionment it is important to find the emission profiles for the sources. In Wåhlin et al. (2001a), it has been shown that the emission profiles, in principle, can be determined, if the average CO/NO_x emission ratios for the two traffic categories are known. As the CO/NO_x emission ratio for diesel is practically zero compared with the ratio for petrol, the critical unknown parameter is the average CO/NO_x emission ratio for petrol.

This ratio was determined on the basis of traffic counts in Albanigade, Odense, and a specific solution was found for the particle concentrations. The solution for the total particle concentrations in an average week at Albanigade is shown in Fig. 5.6. The small diurnal variation of the non-traffic contribution is different from the traffic pattern and is probably due to the general activity in the city (Wåhlin et al. 2001a).

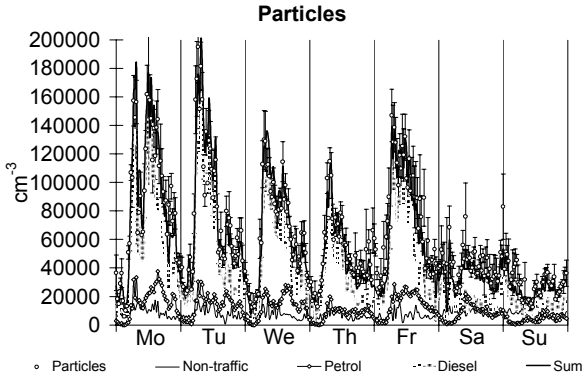

Fig. 5.6. The average weekly cycle of particle number concentrations measured at Albanigade, 3/5/99-20/5/99, and the fitted contributions from the different sources (Wåhlin et al. 2001a)

5.4.2 A field measurement campaign in Helsinki, Finland

Major scientific issues investigated (Helsinki)

The principal aim was to reveal the influence of long-range transport and major local sources on the concentrations of various chemical species in ultrafine ($PM_{0.1}$), fine ($PM_{2.3}$) and coarse ($PM_{2.3\text{-}15}$) aerosol particles. In order to obtain reliable results, the performance of different size-segregating aerosol collectors was compared for 14 different ions. Gas-particle interactions and particle deposition characteristics were also studied.

Relevance to SATURN aims (Helsinki)

The results are useful in estimating the health effects of atmospheric particles and in planning emission abatement strategies. Further, the results can be used as input data for modelling studies, and contribute to an improved understanding of urban particulates and source-receptor relationships.

Experimental set-up (Helsinki)

In April 1996 - June 1997, size-segregated atmospheric aerosols were measured simultaneously at Vallila, an urban site in Helsinki, and at Luukki, a rural site in Espoo. At both sites, separated by about 20 km, virtual impactors (VI, 53 samples) and Berner low-pressure impactors (BLPI, 10 samples) were operated in parallel. In addition, black carbon (BC, particle diameter < 2.5 μm) was monitored at the urban site, and gaseous SO_2, NO_x and O_3 were monitored at both sites.

The different size fractions were analysed utilising inductively coupled plasma - mass spectrometry (ICP-MS) and ion chromatography (IC). About 100 samples were analysed using instrumental neutron activation analysis (INAA). Quality control was introduced by comparing the parallel sampling and parallel analysis (Pakkanen et al. 2001a; Pakkanen and Hillamo 2002).

Main results (Helsinki)

Average fine particle ($PM_{2.3}$) mass concentrations were 11.8 μg/m^3 and 8.4 μg/m3 and those of coarse particles ($PM_{2.3-15}$) were 12.8 μg/m^3 and about 5 μg/m^3 at the urban and rural sites, respectively, indicating that relatively strong local sources existed around the urban site. On average, fine particle mass at the urban site consisted of not-analysed material (43 %, mainly carbonaceous material and water), sulphate (21 %), nitrate (12 %), crustal material (12 %), ammonium (9 %) and sea-salt (3 %), while coarse particle mass consisted of crustal material (59 %), not-analysed material (28 %, mainly carbonaceous compounds and water), sea-salt (7 %), nitrate (4 %) and sulphate (2 %) (Pakkanen et al. 2001b). According to an earlier study (Pakkanen et al. 2000) made at the urban site, the average black carbon (BC) concentration in fine particles was 1.4 μg/m^3, of which 0.4 μg/m^3 was long-range transported. On average, local traffic contributed to the BC concentrations 63 % on working days and 44 % on Sundays.

Average mass concentrations of ultrafine particles ($PM_{0.1}$) were about 0.5 μg/m3 at both the urban and rural sites (Pakkanen et al. 2001a). The ultrafine mass seemed to consist mainly of carbonaceous material (70 %) and water (10 %), since the average concentrations of analysed components were low (at the urban site sulphate 0.032 μg/m^3, ammonium 0.022 μg/m^3, Ca_2^+ 0.005 μg/m^3 and nitrate 0.004

0.004 μg/m^3). The analyses of ultrafine particles consisted of more than 40 chemical components, whose average concentration was above the limit of detection.

Detailed mass size distributions (aerodynamic particle diameter 0.03 - 15 μm) of particulate mass and 27 elements were utilised in estimating fine particle contributions arising from local sources versus long-range transport (Pakkanen et al. 2001c). It was estimated that at the urban site 46 % of fine particle mass and most of, for instance, fine particle Ni (nearly 100 %), Cu (77 %), V (72 %), Bi (67 %), and Sb (55 %) was of local origin. The local contributions of Tl (23 %), As (26 %), Pb (31 %) and Cd (33 %) were estimated to be low. Local road dust was the dominant source for fine particle crustal elements, such as, Al, Ba, Ca, Fe, Mg and Ti, leading to local contributions close to 100 % for these elements. The average particle mass size distributions measured at the urban and rural sites are presented in Fig. 5.7.

Concentrations and size distributions of particle bound low-molecular-weight dicarboxylates were studied at the urban and rural sites (Kerminen et al. 2000). Compared to winter, concentrations were much higher during summer, the likely principal sources being (i) secondary production in the long-range transported air masses and (ii) local traffic.

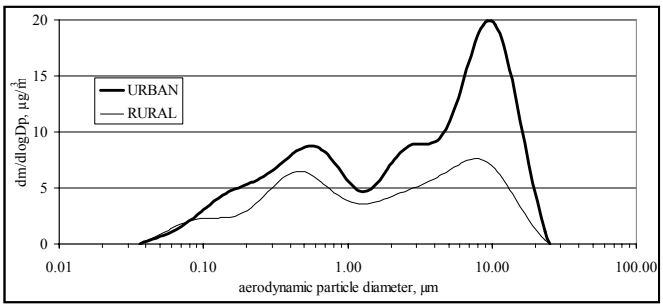

Fig. 5.7. Average particle mass size distributions at the urban and rural site in the Helsinki Metropolitan Area

5.4.3 A field measurement campaign in Budapest, Hungary

Major scientific issues investigated (Budapest)

The origin of aerosol particles is of crucial importance for environmental management in an urban scale. Source characterisation and receptor modelling applied in connection with the measurement campaign provides quantitative estimates of the source contributions on ambient air PM. In contrast to dispersion modelling, no highly detailed meteorological and emission data is needed.

As stated previously, aerosol particles are of great concern due to their adverse health effects. In order to better formulate mitigation strategies to reduce such impacts in urban areas, an important first step is to identify the major sources and composition of ambient particles. Previously, regional scale investigations have been performed in Hungary regarding the transport and chemical composition of particles (Bozó 2000; Havasi et al. 2001). Fine aerosol sampling and related source-receptor modelling for Budapest was started in 1997.

Relevance to SATURN aims (Budapest)

Local sampling and measurements, as well as the Chemical Mass Balance method has been applied to estimate the source contributions to ambient air concentration levels of trace elements in Budapest. The general aim is to optimise the emission control strategies that will be effective in reducing the ambient concentrations of pollutants considered. Effects of regional pollution could also be pinpointed during the sampling and measurements campaign.

The experimental results are utilised in combination with the ADMS model computations (these are described in more detail in the modelling part of this section) in order to locate the most polluted areas in Budapest. The temporal variation of particle concentrations can also be evaluated based on measurements conducted during several years.

Experimental set-up (Budapest)

Source profiles of Cd, Cu, Ni, Pb, V and Zn for waste incineration, traffic, oil and coal burning were applied for model calculations. Aerosol sampling for fine size range aerosol particles was carried out by Harvard-type impactors at the sources and at two receptor points, in the downtown of Budapest during November and December in 1999, and summer months in 2000 and 2001.

One of the sampling sites (OKI) is located at the SE edge of the downtown area with relatively heavy traffic in its vicinity, while another site (ELTE) is located in the vicinity of the Danube River that crosses the city in the N-S direction. This site is fairly intensively ventilated; it can therefore be expected that a mixture of various source categories be detected at that site.

Main results (Budapest)

It was found that substantial amounts of Zn, Pb and Cu are originated from a waste incinerator that is located in Budapest. Regarding the traffic profile, the most important element is still Pb; however, its relative contribution has decreased rapidly during the past five years.

It was concluded that waste incineration provides the most significant contribution to the toxic metal load in Budapest (from 65 to 70 %). The relative contribution of traffic sources ranges from 11 to 17 % (Table 5.1). Coal burning has no significant importance in Budapest, regarding the receptor profile. The coal consumption has significantly decreased in Budapest during the past decade, since it has been replaced by natural gas at most industrial, power-production and residential sources.

In co-operation with NOAA, three-dimensional backward trajectories were also computed in order to estimate the origin of air masses. Episodes with elevated concentrations of Ni and V measured in Budapest are accompanied by Southerly winds, indicating the effects of an oil refinery located at a distance of 25 km South of Budapest.

Measurement campaigns are continued at three receptor points in Budapest as well as in the vicinity of relevant stationary and mobile sources. The objective is to extend the investigations towards the relationships between meteorological conditions and receptor profiles.

Table 5.1. Relative contribution (%) of source categories to ambient trace element concentrations (Cd, Cu, Ni, Pb, V and Zn) in Budapest

Pollutant source category	Measurement site	
	OKI	ELTE
Waste incineration	65	70
Traffic	17	11
Coal burning	5	6
Oil burning	6	6
Other sources	7	7

5.4.4 A field measurement campaign in Stockholm, Sweden

Major scientific issues investigated (Stockholm)

During three years (1998-2000), data has been obtained in order to evaluate source-receptor relationships of both gaseous and particulate air pollution in the urban area of Stockholm. The project has included both emission and air quality measurement campaigns, and modelling using dispersion models and source receptor models (Johansson et al. 2001).

Detailed emission databases have been created in co-operation with the Environment and Health Protection Administration of Stockholm (URL 5.1). These databases include NO_x, CO, PM, benzene and polycyclic aromatic hydrocarbons.

Relevance to SATURN aims (Stockholm)

The overall scientific objectives were:

- to establish source-receptor relationships for hydrocarbons and PM,
- to evaluate the relative contribution of individual sources for the distribution of these compounds and
- to evaluate emission inventories using a combination of measurements, source-receptor models and dispersion models.

Experimental set-up (Stockholm)

Three different experimental activities have been carried out:

- a road tunnel study; on-road emissions were characterised,
- a wood combustion experiment; characterisation of the aerosol and some gaseous compounds and
- an urban field campaign in Stockholm.

In the road tunnel study, two sampling points were used: at a distance of 100 m from the entrance and 900 m into the tunnel. Both sites were equipped with Sierra Anderson Hi-Vol PM_{10} inlets, through which the tunnel air was brought through the tunnel wall and into an adjacent space housing the instruments. The following aerosol and gas measurements were performed: aerosol size distributions (from 3 to 800 nm; DMPS), PM_{10} mass (TEOM), coarse and fine fraction elemental composition (PIXE), organic and elemental carbon (ACPM), CO, NO, NO_2, light hydrocarbons (C2-C7), volatile hydrocarbons (> C5), alkanes (> C10), PAH's, benzene, toluene, xylene and ethyl-benzene. These measurements (except for PAH) were carried out with a time resolution of between 15 minutes and 1 hour. Furthermore, categorised traffic flows and speeds were recorded as well as the wind velocity, temperature and relative humidity in the tunnel.

For the urban field campaign, the same equipment as in the tunnel was installed at three sites in Stockholm. The sites included a street canyon with around 40 000 vehicles per day, a residential area just outside the centre and a rooftop site in central Stockholm.

Main results (Stockholm)

In the tunnel, with around 40,000 vehicles per day, hourly average total particle number concentration reach 1 million during morning rush hours. Most of the particles are in the size range from 10 to 60 nm diameter (Kristensson et al. 2000; Johansson *et al.* 2001). Total numbers of particles in the size range from 3 to 7 nm tend to decrease significantly during morning rush hours, due to coagulation and deposition on tunnel walls as shown by Gidhagen et al. (2002).

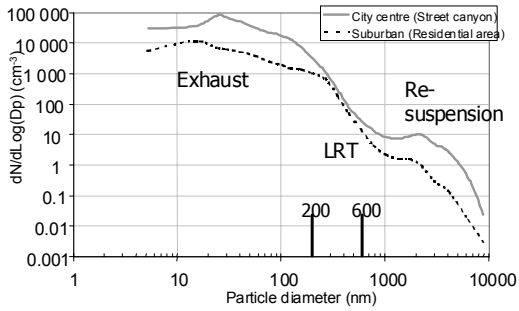

Fig. 5.8. Measured particle size distribution in the city centre (upper thick line) and in a residential area (dotted line, lower) in Stockholm. ”Exhaust” (particles < 200 nm), ”Resuspension” (> 600 nm) and ”LRT” (200 – 600 nm) indicate the particle size ranges that are heavily affected by local vehicle exhaust emissions, particles generated by local wear processes and particles which are mainly due to long-range transport, respectively.

The particle size distribution varies considerably from a road tunnel and densely trafficked streets to a background site far from the city. The results suggest that local traffic is the main source of ultra-fine particles and most of the particles close to traffic are in the range from 3 to 30 nm. The size distribution changes markedly as a function of distance from the traffic. At the rooftop location, the size distribution shifts towards larger sizes, compared with the corresponding results at the street level location.

The particle size distributions measured at two different sites in Stockholm were compared with each other. Fig. 5.8 shows three distinct modes in the size distribution: < 200 nm, from 200 to 600 nm, and > 600 nm (as aerodynamic diameters). Local vehicle exhaust particles have a dominant contribution to the number concentration of particles less than 200 nm, whereas long-range transport dominates the sizes between 200 and 600 nm. For the large particles (> 600 nm), local vehicles are again the main source, but these particles are mainly originated from the wear of roads, tyres, brakes etc.

Principal component analysis was performed to aerosol and gas phase data from measurements in a road tunnel. NO_x, CO, CO_2, VOC’s and copper were mainly associated with gasoline exhaust, whereas particulate organic carbon and NO_2 shows high loadings on both the gasoline and diesel factor. Particle number concentration is dominated by particles with a diameter around 20 nm and associated with diesel exhaust. Elemental carbon, particle surface area (<900 nm) and volume (<900 nm) show highest loadings on the diesel factor. The road dust factor has high loadings of $PM_{2.5}$ and a number of elements (Si, Fe, Mn etc.).

Birch wood is widely used as fuel in stoves and boilers of Swedish households. An experiment has been undertaken to characterise the combustion aerosol and gaseous emissions of wood combustion in a small stove (Hedberg et al. 2002). In

traffic exhaust, the number concentration of particles was largest at 20 nm, while the number distribution from wood burning ranged from 20 to 300 nm.

5.4.5 A field measurement campaign in Hatfield, United Kingdom

Major scientific issues investigated (Hatfield)

PM in the atmosphere is present in different size ranges, as low as few nanometer to tens of micrometer, with various shapes (McMurry 2000). The environmental impact of these particles, such as, health hazards, acid rain, global albedo and visibility degradation as well as their fate and transport is basically determined by factors including the size of the particles and their chemical composition, in addition to other environmental variables (McMurry 2000). Therefore, to understand the environmental and health impact of ambient particles it is important to study their chemical composition as a function of their size. Concentrations of several metals and ions have been determined for size-differentiated particles. As different sources emit particles in different size ranges, such a study can also assist in the understanding of source-receptor relationships.

Relevance to SATURN aims (Hatfield)

Chemical analysis results of size-fractionated samples of ambient particles will help in arriving at source-receptor relationships for air pollutants. Such data will also assist in apportioning the sources of particles and in determining the key particle size ranges contributing to particulate mass metrics such as PM_{10}, $PM_{2.5}$ and PM_1.

Experimental set-up (Hatfield)

The ambient particle samples in 10 size ranges were collected using a MOUDI impactor from a suburban rooftop site, on the campus of University of Hertfordshire, Hatfield. The samples were collected in August and September 2000, and March 2001. The samples from the both campaigns were analysed gravimetrically and for metals (Pb, Zn, Fe, Ni and Cu) using FAAS and ICPES. The samples from first campaign were also analysed for water-soluble ions (SO_4^{-2}, NO^{-3}, Cl^-) using ion chromatography.

Main results (Hatfield)

The results of the chemical characterisation from two sampling campaigns are presented in Fig. 5.9. Figure shows the distribution of ambient particle mass in the

measured size fractions. The percentage concentrations of the analysed species in PM_{10}, $PM_{2.5}$ and PM_1 fractions for August - September 2000 campaign are shown in Fig 5.9a, and for March 2001 campaign are shown in Fig. 5.9b. The general size distribution pattern exhibited by the particulate mass is quite similar during both campaigns. The mass concentration during the campaign of March 2001 is slightly lower compared to the August - September campaign, mainly because during the March campaign, the average wind speed was higher and there was a higher level of precipitation.

The PM_{10} mass and all but iron of the analysed species are dominated by the fine fraction. More than two thirds of PM_{10} mass and over 80 % of zinc and lead are in fine fraction (particles of 2.5 micron and less). The contribution of PM_1 is dominant except for Fe and Cu. Around 50-60 % of the PM_{10} mass is accounted by particles in the range of PM_1. The overall size distribution of particles is similar for the spring and summer campaigns, except for Fe, where the coarse fraction was more significant during the March measurements.

The dominance of the fine fraction in the particle mass and the analysed species point towards the influence of anthropogenic activities on the suspended PM at this site. As expected, the concentration of lead in the campaigns was quite low and comparable (6.6 in summer and 8.0 ng/m^3 during spring).

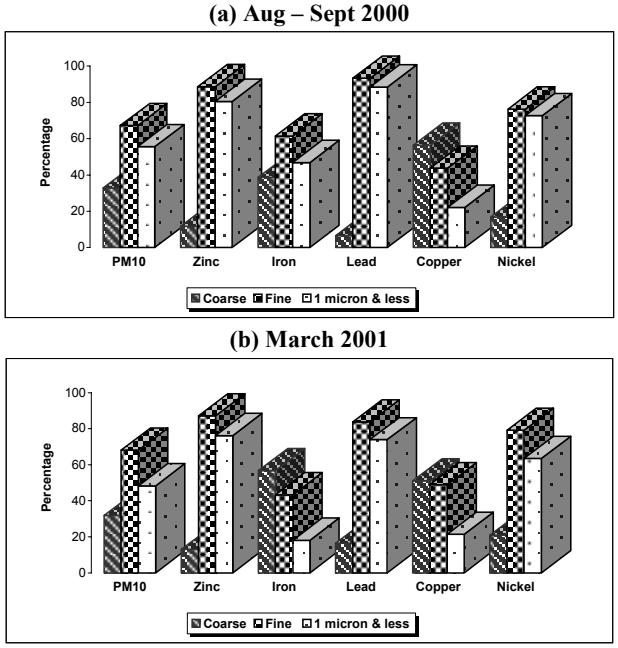

Fig. 5.9. Percentage fraction of coarse, fine and PM_1 fractions in the metal species and particulate mass during two campaigns (Sokhi *et al.* 2001)

5.5 Modelling of particulate matter in urban areas

Mathematical models have been developed, evaluated and applied in order to predict aerosol processes, and the number and mass concentrations, such as $PM_{2.5}$ and PM_{10}, in urban areas. Source apportionment modelling has been discussed in relation to the field measurement campaigns.

5.5.1 Aerosol process models

In Finland, an aerosol dynamical model MONO32 was applied that was originally developed at the University of Helsinki (Pirjola and Kulmala 2000). The model takes into account gas-phase chemistry and aerosol dynamics (nucleation, coagulation, condensation/evaporation and deposition). The particles are classified into four different size modes, which are monodisperse. The first objective was to evaluate quantitatively the influence on aerosol evolution of various chemistry and aerosol processes.

Pohjola et al. (2003) have compiled vehicular exhaust scenarios in selected urban environments. They studied the effects of coagulation, condensation, the concentration of condensable organic vapour, and the dilution of the exhaust plume on the number concentration, composition and particle size.

A particle dynamic module has been developed by Gidhagen et al. (2002). It is intended for studying the dynamics of different particle size distributions in a heavily trafficked urban environment. The model was tested by comparison with measurements in a road traffic tunnel (Kristensson et al. 2001b). A CFD model is coupled to the above mentioned MONO32 aerosol dynamical model. For the tunnel application, a single compound is used to represent the particle composition, although it is possible to handle separately inorganic salts, organic and elemental carbon, sea salt and mineral dust.

The CFD solver is used to calculate the transport equations for the particle number concentration and the particle mass concentration of particle size mode. The effect on particle number concentrations of coagulation and dry deposition was evaluated under morning rush hour conditions. The model simulations showed:

- For the two smallest size modes used in the model (diameter below 29 nm), coagulation had the largest effect on the resulting number concentrations, but dry deposition was also found to be important.
- With the high particle number concentrations that characterise the morning rush hour, coagulation will generate a maximum of the nucleation mode in middle of the tunnel, with decreasing concentration towards the ends of the tunnel.
- For the coagulation process, the hygroscopicity of the particles is not important. A change of sulphate content from 3 % to 50 % in the particles, with resulting

particle growth due to increased water uptake, did not produce any significant changes in the coagulation rate.
- The determination of emission factors of ultrafine particles must, therefore, include the effects of coagulation and wall losses due to dry deposition.
- The transient behaviour of particle number concentrations during 24 hours was also investigated.

From the case study we conclude:
- Constant emission factors cannot explain the observed time series of number concentrations for particles smaller than 29 nm. The model study suggests that the emission factors for light duty vehicles are strong functions of speed; the emissions are reduced when the speed is decreased.
- The total number emission factors of the light duty vehicles (in average 95 % gasoline fuelled cars, the remaining being diesel fuelled) determined from the present tunnel model study ($1.6\ 10^{14}$ to $8.4\ 10^{14}$ $veh^{-1}\ km^{-1}$) are in general higher than what has been reported from laboratory measurements.
- While translating the estimated particle number emission factors to emission factors for particle mass, the present study yields values comparable to those presented from other tunnel measurements.

If particle number concentration turns out to be important from a health point of view, it will be essential to quantify the emission rates and dynamic behaviour of the most abundant particles, i.e. those with a diameter around 20 nm.

5.5.2 Roadside dispersion models

CALINE4 is a US EPA approved roadside dispersion model that incorporates the effects of thermal and mechanical turbulence for roadside situations (Benson 1984; Sokhi et al. 1998). The team at the University of Hertfordshire (UK) has applied the CALINE4 model as well as the ADMS-Urban model to a site at a major motorway (M25). In addition to air quality monitoring, the site also included traffic counts and meteorological measurements.

With regard to PM_{10}, CALINE4 and ADMS predictions show good agreement with measured data. In this case the measurements along with the model predictions indicate that the 24 hour running mean standard of 50 μgm^{-3} will be exceeded. However, this needs further investigation, as there can be considerable contribution from urban background in terms of mass of particles.

Similar work was conducted regarding the model CAR-FMI (e.g., Kukkonen et al. 2001b). This roadside emission and dispersion model was extended to allow for the emissions (discussed in section 5.2.3) and dry deposition of PM. The model predictions were compared with a $PM_{2.5}$ measurement campaign in the vicinity of a major road (Tiitta et al. 2002).

5.5.3 Semi-empirical and statistical models

Besides the detailed modelling of aerosol processes, a simple model was developed for predicting the concentrations of $PM_{2.5}$ in urban areas (Tiitta et al. 2002; Karppinen et al. 2002). The influence of primary vehicular emissions is evaluated using the roadside emission and dispersion model CAR-FMI (e.g., Kukkonen et al. 2001b), used combined with the meteorological pre-processing model MPP-FMI.

The regionally and long-range transported (LRT) contribution to the concentrations of fine particulate matter ($PM_{2.5}$) are required for dispersion model computations on an urban scale. However, direct measurements of $PM_{2.5}$ on proper regional background locations are commonly not available. A statistical model for evaluating the regionally and long-range transported concentration of $PM_{2.5}$ has therefore been developed, that can be used for evaluating the fine particulate matter concentrations; based on available measurements at the nearest EMEP (Co-operative programme for monitoring and evaluating of the long-range transmission of air pollutants in Europe) stations (Karppinen et al. 2002).

The modelling system was tested against the results from a measurement campaign in a suburban environment near a major road in Kuopio, Central Finland. The mass concentrations of fine particles ($PM_{2.5}$) were measured simultaneously at four distances from a major road, together with traffic flows and relevant meteorological parameters (Tiitta et al. 2002). This modelling system could also be applied in other European cities for analysing the source contributions to measured fine particulate matter concentrations.

A semi-empirical model has been developed for evaluating the PM_{10} concentrations in urban areas (Kukkonen et al. 2001a). The basic model assumption is that local vehicular traffic is responsible for a substantial fraction of the street-level concentrations of both PM_{10} and NO_x, either due to primary emissions or vehicle-driven PM from various surfaces. The modelling system utilises the data from an air quality monitoring network in the Helsinki Metropolitan Area. The model also includes a treatment of the regional background concentrations, and resuspended PM. The model performance was evaluated against the measured PM_{10} data from five air quality stations in the Helsinki area in 1999.

5.5.4 Sample results on modelled spatial concentration distributions

The spatial concentration distribution of PM_{10} in Budapest

The spatial distribution of the PM_{10} concentrations was estimated for Budapest using the ADMS dispersion modelling system (Singles and Carruthers 1999). Example results for one specific day are presented in Fig. 5.10. High-resolution emission inventory included contribution from both point and line sources in the city. Meteorological input of the model was based on data gained from surface meas-

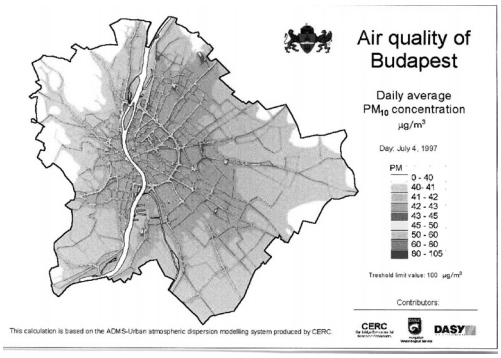

Fig. 5.10. Daily average concentrations of PM_{10} in Budapest on 4/7/1997 (μg m^{-3}).

urements as well as on vertical sounding data from Budapest. The model results showed that the highest PM_{10} concentrations occur in the vicinity of major roads in Budapest.

The spatial concentration distribution of PM_{10} in Helsinki

As an example application for the semi-empirical model developed by Kukkonen et al. (2001a), we have computed the annual average spatial concentration distribution of PM_{10} over central Helsinki in 1998. The predicted distribution is presented in Fig. 5.11.

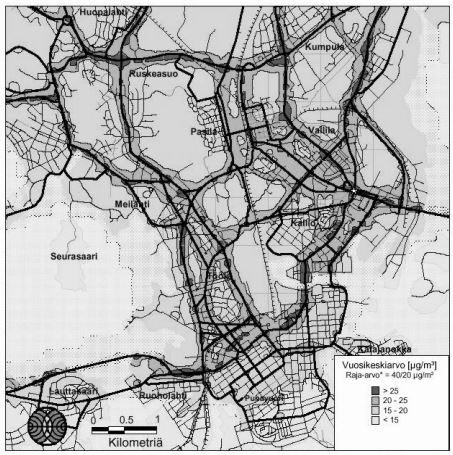

Fig. 5.11. Predicted spatial distribution of yearly means of PM_{10} (μg m^{-3}) in central Helsinki in 1998. The size of the depicted area is 7 km x 7 km. The location of the main streets is shown. The white star indicates the predicted max concentration in the area (41 μg/m^3)

The computations considered here correspond to urban background concentrations, averaged over the length scale of the grid size applied. The grid interval of the receptor point network in the area considered varies from 50 to 200 m.

Clearly, the concentrations of PM_{10} are highest in the vicinity of the main streets. It is of interest to compare the predicted concentration values with the EU limit values. The limit values in the first and second stages are intended to be in use in 2005 and 2010, respectively; the corresponding numerical values are 40 and 20 $\mu g/m^3$. The first stage limit value would be violated only at the centre of one specific junction of busy streets. However, the second stage limit value is exceeded over a relatively wide area in the vicinity of the main streets of central Helsinki.

The spatial concentration distribution of PM_{10} in Stockholm

The spatial distribution and population exposure to PM_{10} have been estimated in an earlier project in Stockholm (SHAPE; Stockholm study on Health Effects of Air Pollution and its Economic consequences; Johansson et al. 1999). The calculations are based on an Information and Air Quality Assessment System that is administered by the Regional Association for Air Quality Management in the counties of Stockholm and Uppsala.

Road traffic emissions are calculated from detailed information on traffic volumes, vehicle compositions and driving conditions. All this information is given for short links with a length ranging from less than 100 m up to 2 km. The database also includes emissions from burning of different fuels in district and residential heating, emissions from ships and a special model for airborne re-suspension of PM from road dust. The re-suspension model considers the influence of meteorological conditions, the wetness of the road surface and effect of the traffic on the re-suspension rate.

5.6 Exposure to urban particulate matter

5.6.1 Variability in exposure

Measurements of road-user exposure to $PM_{2.5}$ in London (Adams et al. 2001a) showed that people moving along city streets are exposed to levels of pollution that are not only substantially higher than urban background fixed-point measurements, but also higher than roadside and kerbside measurements. Volunteers carried high volume portable $PM_{2.5}$ sampling equipment (Adams et al. 2001b) on bicycles, buses, and cars along three routes in Central London, each route approximately 5 km in length. One month of measurements four times a day were made in July, followed by a smaller set of measurement in winter. An additional

set of measurements were made by cyclists following their usual commuting routes in the morning and evening.

Day-to-day changes in meteorological conditions, especially wind speed, was the most important determinant of exposure that could be identified using regression modelling (Adams et al. 2001c). Mode of transport was a much weaker determinant, with no detectable effect in the total $PM_{2.5}$ measurements. Isolation of the diesel exhaust fraction of $PM_{2.5}$, using reflectance as a surrogate for elemental carbon aerosol, indicated that car drivers were exposed to higher concentrations than cyclists (Adams et al. 2002).

The most surprising finding was that individual level variability in exposure was as high as the variability due to changes in meteorological conditions. For example, three cyclists following the same route at the same time of the same day measured exposures with geometric standard deviation similar to the geometric mean. This variability is much larger than the possible error in an individual measurement.

5.6.2 Modelling road-user exposure and its source apportionment

Dispersion modelling using an operational street canyon plume model in ADMS-Urban was used to investigate likely causes and implications of the observed variability in road-user exposure to $PM_{2.5}$. The street canyon model was run with a high spatial resolution of output receptors along the routes where measurements were made. The modelled exposure was found to be dominated by very high concentrations of $PM_{2.5}$ at the most congested, poorly ventilated canyons along the route, with differences as large as a factor of ten being found between some pairs of road links. This is capable of explaining, in part, the individual level variability in exposure, as small differences in cyclist behaviour including speed lead to differences in the amount of time spent at the most polluted locations. It is also consistent with the observation (Adams et al. 2002) that the individual level exposure is found in the carbonaceous fraction of the $PM_{2.5}$.

Furthermore, the modelling shows how the source apportionment of the $PM_{2.5}$ is very different at the most polluted locations to other parts of the route. Emissions on the nearest road dominate almost to the exclusion of all others at the most polluted points. Along most of the rest of the route, source apportionment is similar to that at roadside locations, with similar contributions from transboundary imported secondary $PM_{2.5}$ to nearest road contribution, and a smaller but significant contribution of primary $PM_{2.5}$ from the rest of London.

Some sources of variability are, however, omitted from this modelling. While the main features of two-dimensional street canyon plume circulation are captured by the semi-empirical canyon model, complex features near junctions are not resolved (e.g. Scaperdas and Colvile 1999). Spatial and temporal variability associ-

ated with individual vehicles is also smoothed out, and temporal variability associated with signal control of traffic flows is ignored. It is not known to what extent each of these sources of variability are important in determining variability in exposure. A major programme of work to investigate this in detail in Central London is therefore taking place during 2002-2006 (URL 5.2).

The model results outlined above are all for the mass of PM to which people are exposed. A preliminary study (Bowsher 2000) of the microphysical evolution of aerosol after emission from diesel exhaust indicated that coagulation causes the particle size to increase during the time taken for dispersion to carry the emissions from the vehicle slipstream to the edge of the road. Exposure to ultrafine particles will therefore be even more strongly dominated by short-term peak concentrations than exposure to $PM_{2.5}$.

5.7 Conclusions

Several field scale measurement campaigns that focused on aerosols were conducted within the SATURN project. Such campaigns include those performed in Denmark, Finland, France, Greece, Hungary, Sweden and the United Kingdom. A key aspect of these campaigns was the source apportionment of PM. The analysis of these results has contributed to a substantially improved insight on the source contributions, and the chemical and physical characteristics of polluted urban air. For instance, it was demonstrated that local traffic is the dominating source of ultrafine particles, while regional and long-range transport can have a dominating effect on the fine particulate mass ($PM_{2.5}$).

Contributions from various mobile sources may be assessed, e.g., using averaged Scanning Mobility Particle Sizer (SMPS) data in combination with routine monitoring data and traffic rates. Based on the combination of urban sampling, measurement and 3D trajectories, the rural influence on fine particle concentrations and chemical composition can also be evaluated. Chemical mass balance or principal component analysis provide a fairly simple, but useful tool for the assessment of source contributions.

Improved knowledge is needed regarding the chemical and physical properties of urban air particles (especially EC, OC, PAH and metals), surface properties, volatility, hygroscopicity and morphology. It is therefore necessary to apply other advanced analytical techniques, such as SEM and micro probe analysis.

A systematic urban monitoring network is needed in Europe for the evaluation and analysis of fine particles. Such measurements should include not only mass fraction measurements, but also size distribution and chemical composition, in order to achieve reliable information on the origin and potential health effects of PM.

Somewhat smaller resources have been devoted to the mathematical modelling of particulate matter within the SATURN project, compared with those of the experimental work. However, in many cases the measurements were supplemented with one or several modelling methods, such as source apportionment analyses, deterministic and semi-empirical modelling.

Two research teams have performed detailed computations of aerosol transformation, using the aerosol process model MONO32. Interesting results have also been obtained concerning the extension of roadside emission and dispersion models to include a treatment for particle mass fractions. Semi-empirical (partly statistical) models have also been developed for evaluating the concentrations of both $PM_{2.5}$ and PM_{10}.

For modelling purposes, a better knowledge is needed regarding the emissions of PM. Information is needed on the number and particle size distribution, and chemical content in vehicular exhaust. For regulatory dispersion modelling, the emission modelling should also be comprehensive in terms of major vehicle categories and driving speeds. Information would also be welcome on the dependence of vehicular emissions, e.g., on cold starts, driving conditions including idling, and fuel composition. Similar information is also needed for biomass combustion, as this is relevant in residential areas in some parts of Europe.

The combined use of aerosol process models and atmospheric dispersion models is another major challenge for the future. Work towards this aim is in progress within the EMEP programme. Evaluating and modelling of the exposure to urban PM is another important topic for future research.

Clearly, atmospheric aerosols have been studied also in many other EUROTRAC-2 projects, such as GENEMIS, AEROSOL, CMD, GLOREAM and PROCLOUD. Closely related work is in progress also within the WMO GURME programme and the COST 715 action.

Urban aerosols will be investigated in several projects of the CLEAR cluster; this is a network of projects that have been granted funding from the "City of Tomorrow" programme of the EU 5th Framework Programme. Several contributors of this review are participants in one or more CLEAR projects; this will therefore be a natural forum in order to utilise and forward the expertise gained in SATURN.