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Spatial & temporal variations of PM10 and particle number concentrations in urban air

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Abstract The size of particles in urban air varies over four orders of magnitude (from 0.001 μ m to 10 μ m in diameter). In many cities only particle mass concentrations (PM10, i.e. particles $< 10 \,\mu m$ diameter) is measured. In this paper we analyze how differences in emissions, background concentrations and meteorology affect the temporal and spatial distribution of PM10 and total particle number concentrations (PNC) based on measurements and dispersion modeling in Stockholm, Sweden. PNC at densely trafficked kerbside locations are dominated by ultrafine particles ($<0.1 \ \mu m$ diameter) due to vehicle exhaust emissions as verified by high correlation with NOx. But PNC contribute only marginally to PM10, due to the small size of exhaust particles. Instead wear of the road surface is an important factor for the highest PM10 concentrations observed. In Stockholm, road wear increases drastically due to the use of studded tires and traction sand on streets during winter; up to 90% of the locally emitted PM10 may be due to road abrasion.

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Swedish Meteorological and Hydrological Institute, S-601 76 Norrköping, Sweden PM10 emissions and concentrations, but not PNC, at kerbside are controlled by road moisture. Annual mean urban background PM10 levels are relatively uniformly distributed over the city, due to the importance of long range transport. For PNC local sources often dominate the concentrations resulting in large temporal and spatial gradients in the concentrations. Despite these differences in the origin of PM10 and PNC, the spatial gradients of annual mean concentrations due to local sources are of equal magnitude due to the common source, namely traffic. Thus, people in different areas experiencing a factor of 2 different annual PM10 exposure due to local sources will also experience a factor of 2 different exposure in terms of PNC. This implies that health impact studies based solely on spatial differences in annual exposure to PM10 may not separate differences in health effects due to ultrafine and coarse particles. On the other hand, health effect assessments based on time series exposure analysis of PM10 and PNC, should be able to observe differences in health effects of ultrafine particles versus coarse particles.

Keywords Coarse particles · Health effect assessment · Vehicle emissions · Resuspension · Traffic exhaust · Ultrafine particles · Urban aerosol

1 Introduction

The size of particles in the atmosphere varies over four orders of magnitude, from a few nanometers to tens of micrometers. Size, shape and chemical properties (such as hygroscopicity) govern the lifetime of particles in the atmosphere and the site of deposition within the respiratory tract. Health effects of ultrafine particles (UFP, diameter <100 nm) are likely very different from those caused by coarse particles (>2.5 μ m). While freshly emitted UFP, consisting of soot and organic compounds, may be less hygroscopic and penetrate deep into lung alveols, coarse particles, consisting of soil mineral elements and salts, tend to deposit in the upper part of the respiratory system. UFP is part of the fine particle fraction ($<2.5 \ \mu m$), which is suspected to be more important for mortality. Nevertheless, the EU directive regulates the total mass of all particles less than 10 μ m irrespective of size, morphology and chemistry and also irrespective of their health effects. Measures aimed at reducing the negative health impact of particles must necessary build on an understanding of the controlling factors not only for PM10 concentrations but also for other particle size fractions.

High concentrations of UFP are found in vehicle exhaust, contributing to high number concentrations in street environments (Shi et al., 1999; Molnar et al., 2002; Longley et al., 2003; Ketzel et al., 2003a, 2003b; Gidhagen et al., 2004). At kerbside locations UFP, contribute only marginally to the total mass concentration measured as PM10. This is because the UFP generated in traffic exhaust are concentrated in the 10-30 nm size intervals, too small to build up aerosol mass even in high number concentrations. However, the comparatively low importance of the UFP particles for the PM10 levels is also due to the large contribution of road wear particles (Rogge et al., 1993; Chow et al., 1996; Pakkanen et al., 2001; Querol et al., 2004). In Berlin about 45% of the local traffic contributions to PM10 concentrations at kerb side are due to resuspended soil material (Lenschow et al., 2001). The remaining traffic contribution is due to vehicle exhaust and tyre abrasion. This is similar to the conclusion by Harrison et al. (1997) that about 50% of PM10 during summer months in Birmingham was due to the coarse particle fraction (PM10-PM2.5), indicating the importance of road dust. In addition they found a positive dependence of coarse particles of wind speed due to resuspension (i.e. higher concentrations as wind speed increases), in contrast to what is expected for the exhaust particles, which will be more diluted as wind speed increases. Kukkonen et al. (2001) concluded that short term PM10 concentrations may not be modeled using NOx as indicator of traffic emissions, indicating that other processes than vehicle exhaust such as road dust and background, determine the PM10 levels in Helsinki. There are still relatively few papers on the long term variability of number concentrations in urban areas (Tuch et al., 1997; Harrison et al., 1999; Pitz et al., 2001) and data on UFP exposure and health effects are even more limited (Englert, 2004). Tuch et al. (1997) found very poor correlation between particle number concentrations (PNC) and mass concentrations in the size range 0.01–2.5 μ m. In their study, performed during 8 winter months in Erfurt in eastern Germany, they found that PNC was dominated by particles smaller than 0.1 μ m, whereas particle mass concentrations were dominated by particles in the range 0.1–0.5 μ m. Poor correlation between fine particle mass concentration and PNC were also found at urban locations in Austria (Gomiscek et al., 2004). In three eastern German cities during 1993 to 1997, Pitz et al. (2001) analysed the particle size distribution and total number concentration and found that while the mass concentration had decreased during this period, there was no significant change in the total number concentrations. In addition they could observe a shift in the particle size distribution to smaller particle sizes, possibly caused by changes in local emissions. Based on data from an urban site in Birmingham, Harrison et al. (1999) concluded that measurements of PNC are far better indicator of traffic exhaust emissions than PM10 (mass), the latter being strongly influenced by the background aerosol. Similarly, Van Dingenen et al. (2004) compiled aerosol concentration data from 31 European sites and concluded that particle number concentrations increase more than proportionally to PM mass going from rural sites to kerb side locations. Even though there is a fairly good correlation between PM2.5 (or PM10) and PNC at clean sites, poor correlation is generally found at polluted sites (Keywood et al., 1999; van Dingenen et al., 2004). PNC in urban air are affected not only by primary emissions but also by particle processes. Ketzel et al. (2003b) have observed, during periods with low primary particle emissions, particle formation events that are due to nucleation in background air. They found that total particle number concentrations in Copenhagen increased by up to 5-10 times within a few hours in connection with clean air and high solar radiation. Also the generation rate of vehicle exhaust particles, formed in the combustion process or just after leaving the exhaust tube, may vary due to environmental conditions and not solely as a function of traffic intensity. Charron and Harrison (2003) observed for a street canyon data set that the concentration of particles in the size range 11–30 nm increased relatively to larger particles during periods with lower temperatures as well as with higher wind speeds, indicating the importance of ambient conditions for particle dynamic processes such as nucleation, coagulation, condensation in the immediate vicinity of the vehicle exhaust tube.

In this paper we have used both model calculations and measurements to assess the differences in temporal and spatial variability in Stockholm of PNC and PM10 respectively. The different behavior of the two particle measures is explained in terms of the sources and meteorological factors controlling the emissions. Understanding the causes of the variability of particle concentrations is crucial for assessing the relative importance of PNC and PM10 for health effects based on epidemiological studies. Finally, the paper includes a discussion on the implications of the temporal and spatial variability of PM10 and PNC for the possibility to clarify which component of PM plays the most important role in eliciting adverse effects on health in time-series versus long-term epidemiological studies is being discussed.

2 Methodology

Particle mass concentration measurements were performed by the Environment and Health Protection Administration of Stockholm using automatic TEOM instruments (Tapered Element Oscillating Microbalance, model 1400, Rupprecht and Pataschnik) equipped with PM10 and PM2.5 inlets. Measurement stations were located in street canyons with busy streets (10000–35000 vehicles/day) in central Stockholm (Hornsgatan¹, Sveavägen² and Norrlandsgatan³) and one urban background station at roof level; ~20 meter above street (Rosenlundsgatan⁴) in the city. Meteorological parameters including wind speed, wind direction, temperature and relative humidity are also measured at roof level. The wetness of the road surface was monitored at Norrlandsgatan using a simple electrical resistance wire and at Horngatan using a commercial Road sensor (IRS-21, Signalbau Huber, Germany). Total number of particles (>7 nm aerodynamic diameter) was measured using a CPC3022 (TSI Inc.), both at one of the kerb side stations (Hornsgatan) and at the urban background station (Rosenlundsgatan). The annual mean PM10 and particle number concentrations were calculated using a wind model and a Gaussian air quality dispersion model, both part of the Airviro Air Quality Management System (SMHI, Norrköping, Sweden; http://airviro.smhi.se). Meteorological conditions were based on a climatology that was created from 10 years of meteorological measurements (15 minute averages) in a 50 meters high mast located in the southern part of Stockholm. The climatology consists of a list of hourly events, each of them with a certain frequency of occurrence, which together will yield a distribution of different weather conditions that is similar to the distribution of the full scenario period. We have used a scenario that consist of 60 wind direction classes with 6 stability classes within each wind sector, making a total of 360 hourly events. The wind field for the whole model domain was calculated based on the concept first 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. Any non-linear interaction between the scales is neglected. It is also assumed that the adaptation process is very fast and that horizontal processes can be described by non-linear equations while the vertical processes can be parameterised as linear functions. The large scale winds as well as vertical fluxes of momentum and temperature are estimated from profile measurements in one or several meteorological masts (called principal masts). For the model domain analysed in this study (35 km²) only one principal mast is used. This is located in the southern part of the city. Topography and land use data for the Danard model are given by 500 meter resolution. Since the topography of Stockholm is relatively smooth, without dominating ridges or

¹ Hornsgatan is 24 meter wide street canyon with 24 meter buildings and 35 000 vehicles/day.

 $^{^2}$ Sveavägen is a 33 meter wide street canyon with 24 meter buildings and around 35 000 vehicles per day.

³ Norrlandsgatan is 15 meter wide with 24 meter high buildings and around 10 000 vehicles per day.

⁴ Rosenlundsgatan is a roof top site in central Stockholm that can be regarded as representative for the urban background since it

is not directly affected by nearby local vehicle emission. This is also verified by urban scale modelling of particle number concentrations by Gidhagen *et al.* (2005).



Fig. 1 Monthly average total particle number (PNC) and PM10 concentrations at Hornsgatan in central Stockholm (street canyon). The concentrations represent the contribution from the local traffic on this street since the measured urban background concentrations of PM10 and PNC have been subtracted. The monthly mean values were obtained from hourly measurements

taken from January 2001 to May 2005. Vertical bars indicate standard deviation of diurnal averages. Only months with data covering more than 50% of the time have been included (for PM10 the averages are based on between 111 and 149 days; for PNC the averages are based on between 24 and 94 days)

valleys, the free wind can be assumed to be horizontally uniform in the whole domain.

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

Emission factors for PM10 were obtained from measurements in a street canyon using NOx as tracer (Omstedt et al., 2005). Information on traffic flows, vehicle types etcetera was obtained from the emission inventory of the Regional Air Quality Management Association of Stockholm and Uppsala (Johansson *et al.*, 1999). The inventory includes some 20000 road links and an annual traffic volume of 12500 million vehicle km's (data from 2001). The emission factors for particle number concentrations are those suggested by Gidhagen et al. (2005). Chemical and physical transformation processes of particles as well as dry and wet deposition were neglected in the model calculations of annual mean PM10 and number concentrations. See also Gidhagen et al. (2005) regarding the influence of dry deposition and coagulation on particle number concentrations over the urban scale of Stockholm.

3 Results and Discussion

3.1 Differences in temporal variations

Figure 1 shows monthly average PNC and PM10 concentrations at Hornsgatan (street canyon). The concentrations represent the contribution from the local traffic on this street since the measured urban background concentrations of PM10 and PNC have been subtracted. For both PNC and PM10 there is a strong seasonal variation. However, the PM10 levels peak during spring, March to April, whereas the PNC is highest during the winter season, November to February. The spring-time peak in PM10 is caused by suspension of road wear particles (Omstedt et al., 2005). The combined use of studded tires and sanding of street during winter make it likely that road wear is much more important for the PM10 levels in Stockholm as compared to many other cities in Europe. PNC is lower in summer for several reasons; i) lower vehicle exhaust emissions occur due to decreased traffic intensity, ii) higher temperatures during summer decrease the potential formation of new particles, and iii) there is generally more efficient dilution due to more intense turbulent mixing. During some occasions in spring and summer one might expect new particle formation (nucleation) that does not take place directly behind the tailpipes of the vehicles, but in the urban background. Ketzel et al. (2003b)



Fig. 2 Total particle number (PNC) and PM10 concentrations in a street canyon and urban background in central Stockholm (data from February 2002)

reported sudden increases in number concentrations at an urban background station in Copenhagen, which, as they were associated with clean air and preceded by low surface area together with high solar radiation, were interpreted as nucleation events. However, as indicated by the model calculations of Gidhagen *et al.* (2005) particle number emissions from vehicles seems much more important during most part of the year in Stockholm.

PM10 concentrations show very different short term variations (hour to hour and day to day variation) as compared to PNC and NOx. Figure 2 shows hourly mean concentrations of PNC and PM10 during 2 weeks in a street canyon in Stockholm. For exhaust components like PNC and NOx, the concentrations at kerb-side locations depend on traffic emissions and turbulent dilution generated by traffic and wind (Ketzel et al., 2002; Ketzel et al., 2003a; Gidhagen et al., 2004). For PM10, concentrations are not affected by vehicle exhaust but on factors that control road dust generation, such as wear of the road surface and suspension of particles from the road (Norman and Johansson, 2006; Omstedt et al., 2006). In Stockholm road wear is more important for PM10 concentrations than in other countries where the use of studded tires is not so common. Approximately 70% of all light duty vehicles use studded tires in Stockholm during wintertime, November to March (Norman and Johansson, 2006).

Figure 3 shows the effect of road surface wetness on the PM10 and PNC concentrations. The wetness of the road surface was monitored at Norrlandsgatan. PNC and PM10 data are from Hornsgatan (a street canyon site, but the urban background concentrations have been subtracted). Only hours with simultaneous data on PM10, PNC and road wetness were included in the monthly average. Unfortunately road surface wetness was not monitored at the same street as PNC and PM10 concentrations. However, in general the wetness of the different streets in Stockholm is expected to closely follow each other, even though the rate at which the streets dry up may depend on the direction of the street in relation to wind direction and incoming solar radiation. As shown in Fig. 3 road surface wetness is very important for PM10 with systematically higher levels during dry conditions as compared to wet, while no significant difference can be observed for PNC. Particle number concentrations are mainly due to vehicle exhaust emissions that are not affected by road surface wetness. As already discussed above, PM10 concentrations are dominated by the coarse particles coming from the road surface; during wet conditions much less particles are suspended to the air. Measurements have shown that for roads in Sweden where studded tires are used, the total road wear is higher on wet roads (Jacobson, 1994). Of this only a small fraction is particles with diameters less than 10 μ m, but it is likely that also more PM10 is generated during wet conditions as



Fig. 3 Monthly median levels of PM10 and PNC (particle number concentrations) for wet and dry road surface conditions respectively, at Hornsgatan in central Stockholm. Note that the PM10, PNC and road wetness were measured in the street canyon and the levels at roof level (urban background) have been sub-

tracted. Only hours between 07–19 and with data on both PM10, number concentrations and road wetness were included in the monthly average (this is the reason for some months having missing data). Vertical lines indicate 25 and 75 percentiles

compared to dry. But it is not until the road is dry that the particles are being suspended to the air.

Figure 4 shows the variation of the hourly mean PNC and PM10 levels at Hornsgatan during wet and dry road surface conditions. During this period road surface wetness and PM10/PNC were measured at the same street. During the shaded periods the street surface was dry and during the unshaded periodsit was wet. Obviously there is a substantial difference in the temporal variations of PNC and PM10 during this period. The temporal variations in PNC levels follow closely the traffic intensity (not shown) and is essentially unaffected by the road surface humidity, whereas PM10 levels are very low during wet periods and high during dry periods. During wet periods PM10 levels at street level is very close to that measured at the urban background site.



Fig. 4 Hourly mean concentrations of particle number and PM10 at Hornsgatan (street) in central Stockholm. During the shaded periods the street surface was dry and during the unshaded periods it was wet according to measurements using a road sensor



Fig. 5 Scatter plots of daily mean PNC (total number concentration, left) and PM10 (right) versus NOx. Kerb side data from Hornsgatan (Stockholm, October 2001–July 2005)

3.2 Importance of exhaust and non-exhaust emissions for PM10 and PNC

Figure 5 shows scatter plots of PNC and PM10 versus NOx. Obviously, the correlation between NOx and PNC is much higher than for NOx versus PM10. This confirms that PM10 to a large degree is due to nonexhaust particles, whereas PNC is maily due to vehicle exhaust particles. The relative contribution of exhaust versus non-exhaust particles at Hornsgatan in central Stockholm may be estimated using NOx as a tracer (Omstedt et al., 2005). Omstedt et al. (2005) arrived at an annual average emission factor for PM10 of 244 mg $(\text{vehicle km})^{-1}$. This may be divided into the contributions from particle emissions due to vehicle exhaust and brake, tire and road surface wear. Based on the Swedish traffic model (EVA) the vehicle exhaust emission factor for PM at Hornsgatan was about 25 mg/vehicle km (for the vehicle fleet of 2002).

According to CEPMEIP (2002) tire wear contributes with 3.5 mg PM10/vehicle km and 18.6 mg/vehicle km, for light and heavy duty vehicles. Wear of brake linings in Stockholm is around 17 and 84 mg/vehicle km for light and heavy duty vehicles (Westerlund and Johansson, 2002). Of this around 35% may become airborne according to Garg *et al.* (2000), which then for the Stockholm case would translate to emission factors of 5.1 and 25 mg/vehicle km due to wear of brake linings of light and heavy duty vehicles, respectively. Considering that the fraction heavy-duty vehicles on Hornsgatan is around 5% we obtain 4.3 and 6.2 mg/ vehicle km for tire and brake wear respectively. Then the emission factor for road wear can be calculated as the difference between the total and the sum of exhaust, tires and brakes; 244-25-4.3-6.2 = 209 mg/vehicle km. Thus, almost 90% of the total PM10 emission is due to road wear. This consist mainly of coarse (>1 μ m) stone mineral particles. Vehicle exhaust particles are mainly smaller than 0.1 μ m and consist mainly of unburned fuel and motor oil, soot and some metals due to wear motor and exhaust system. Thus, both the chemical composition and the size distribution of PM10 and PNC are totally different.

3.3 Spatial variations in PM10 and PNC

In this section the annual mean spatial variation of PM10 and PNC is compared. The ratio of the total concentration measured at kerb side locations to that measured at roof indicates the importance of local road traffic emissions versus influence of other emissions and background concentrations (Table 1). For PM10 kerb side concentrations are 2 to 3 times higher than urban roof level. For NOx and PNC kerb side concentrations are 4.5 to 8.2 times the roof concentrations, indicating a much higher influence of local traffic exhaust emissions compared to road wear (PM10). For NOx the ratio is substantially higher at Hornsgatan compared to the other streets. This is mainly due to the road sloping upward (2.3%) and that westward going vehicles accelerate after stopping at traffic lights, some 70 meters east of the measurements station. Eastward traffic on

Table 1Annual mean ratioof kerb-side and urbanbackground concentrations(roof at a height of ca20 meters) for PM10, NOxand PNC	Mean ratio Kerb-side/Roof \pm Standard deviation			
	Street average number of vehicles	PM10	NOx	PNC
	Hornsgatan 35 000 veh/day Norrlandsgatan 10 000 veh/day Sveavägen 30 000 veh/day	2.5 ± 1.5 2.0 ± 1.3 2.1 ± 1.5	$\begin{array}{c} 8.2 \pm 3.3 \\ 4.5 \pm 2.3 \\ 4.6 \pm 2.5 \end{array}$	5.6±2.1

the other hand decelerate going down-slope towards the traffic lights, but the measurements presented here are from the sampling point located on the same side as the westward traffic. NOx and particle emissions depend on local driving conditions (start/stop, speed) whereas PM10 emissions depend on factors that show less spatial variability such as road wetness and use of studded tires.

The difference in the dilution of PNC and PM10 concentrations due to the high background concentrations of PM10 as compared to PNC and NOx is similar to what has been observed in Copenhagen by Ketzel *et al.* (2003b). Based on their data from two different streets we calculate average ratios of kerb-side to urban background of between 3.1 and 5.7 for PNC, 5.7 and 8.7 for NOx and 1.7 and 2.9 for PM10. This is similar to the values found in Stockholm (Table 1).

Long-range transport is much more important for the urban and kerb-side PM10 annual mean levels as compared to PNC. Figure 6 shows the calculated relative variation of the annual mean urban background concentrations of PNC and PM10 in the Greater Stockholm area (35 km², 100 meter spatial resolution). The values have been normalized to the rural background level, which is 3200 cm⁻³ for PNC (Gidhagen *et al.*, 2005) and 10 μ g/m³ for PM10 (Forsberg *et al.*, 2005). Only road traffic emissions are included in the calculated values since this is the dominating source of both PM10 and particle number (Gidhagen et al., 2005; Mårtensson et al., 2006). Maximum PNC levels in central Stockholm are more than 5 times higher than background levels. For PM10 much smaller gradients are observed, maximum levels in central Stockholm are only 2 times higher than rural background levels.



Fig. 6 Model calculated relative spatial variation of annual mean concentrations of A) PM10 and B) PNC (total particle number concentration) in Greater Stockholm area ($35 \text{ km} \times 35 \text{ km}$). The

colors indicate the increase above rural background (10 μ g m⁻³ for PM10 and 3500 cm⁻³ for PNC) due to local traffic emissions in the area



Fig. 7 Annual mean population exposure of PNC and PM10 for the Greater Stockholm area as modelled in Figure 6. Each value in the figure is the concentration in 100×100 square meter times the number of people (based on their home address, not working place) in the same area (in total 122 500 points in the 35 × 35 km² model domain) divided by the maximum value of all data

3.4 Implications for exposure and epidemiological studies

So far there is very limited epidemiological data to distinguish health effects of particulate matter with respect to their origin, chemistry and size (Forsberg *et al.*, 2005). Specific exposure-response functions for different particle fractions are urgently needed for optimal control of urban air pollution.

In long-term cohort studies the contrast in yearly mean exposure of a population across a region is being used to find relevant exposure-response relationships (e.g. the American Cancer Society study of Pope et al., 1995). Figure 7 shows a scatter plot of the annual mean exposure concentrations of PNC versus PM10, i. e. the same data as in Fig. 6 but multiplied by the number of people in each grid cell and then normalized by dividing by the maximum value. As shown in Fig. 7 the relative change in exposure to primary emitted PNC and PM10 going from one spot to another in the area is almost exactly the same. This is expected, as local contributions to both PNC and PM10 originate from the same source (which in Stockholm is mainly road traffic). But the interesting consequence is that in different parts of a metropolitan area (dominated by transport-related air pollution) people experiencing, e.g. a factor of 2 different PM10 exposure concentration from local traffic (as an annual mean value) will also experience a factor of 2 different exposure in terms of number of particles (PNC). Thus, epidemiological studies based solely on spatial differences in annual exposure will not be able to separate the health impacts due to PNC and PM10.

From an air pollution point of view, epidemiological studies based on time-series analysis have much better chance to identify which constituents of PM are responsible for adverse health effects. As shown above (Fig. 4) the temporal variations of PM10 and PNC concentrations are much less correlated than the spatial variations. Consequently short term time series analysis of e.g. daily PM10 and PNC concentrations, should be more suitable for identifying potential differences in health effects of PNC and PM10. Since long-range transported (LRT) particles are evenly distributed over a metropolitan area they would not give rise to any spatial contrast in exposure of the population. However, in time series analysis the health impact of LRT particles may also be distinguished from effects due locally produced primary particles.

4 Conclusions

There are substantial differences in terms of the temporal and spatial distribution of total particle number (PNC) and mass (as PM10) concentrations in Stockholm. This is due to differences in source contributions and influence of meteorological conditions. The annual mean urban background PM10 levels are relatively uniformly distributed over the city. This is due to the importance of long range transport, which contributes with 60% to 70% to the annual mean value. For particle number, concentrations are more than 5 times higher in the city center compared to outside the city. Only 20% to 30% of the PNC is caused by non-local sources.

Even close to roads, local vehicle exhaust emissions contribute to less than 10% of PM10 which is manifested in a poor correlation between PM10 and NOx. For PNC there is a very high correlation with NOx indicating the importance of vehicle exhaust emissions. Road wear due to studded tyres and sanding of streets is very important for the highest PM10 concentrations observed during spring. PNC show strong spatial gradients and is highly correlated to NOx. The "effective" exhaust emissions of PNC depend on ambient conditions such as temperature and particle surface area of the existing aerosol, whereas the PM10 road dust emission is controlled by road wetness. Both PNC and PM10 emissions depend on the speed of the vehicles and on vehicle fleet composition.

These results may be important from an epidemiological point of view. In health impact studies of short term health effects based on time series analysis particle mass and number concentrations may not be well correlated and different health effects of number versus mass of particles may be observed. In long term studies analysing spatial variations in annual mean levels, PM10 and particle number concentration are well correlated since they have a common source in traffic and possible differences in health effects of the different particle metrics may not be readily seen. Short term studies of temporal variations will yield better possibilities to identify potential differences in the health response to PM10 and ultrafine particles.

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References

- CEPMEIP. (2002). Co-ordinated European Programme on Particulate Matter Emission Inventories, (http://www.air.sk/ tno/cepmeip/).
- Charron, A., & Harrison, R.M. (2003). Primary particle formation from vehicle emissions during exhaust dilution in the roadside atmosphere. *Atmospheric Environment*, 37, 4109– 4119.
- Chow, J.C., Watson, J.G., Lowenthal, D.H., & Countess, R.J. (1996). Sources and chemistry of PM₁₀ aerosol in Santa Barbara County, CA. *Atmospheric Environonment*, 30(9), 1489–1499.
- Danard, M. (1977). A simple model for mesoscale effects of topography on surface winds. *Monthly Weather Review*, 99, 831–839.
- Englert, N. (2004). Fine particles and human health a review of epidemiological studies. *Toxicology Letters*, 149, 235–242.
- Forsberg, B., Hansson, H.-C., Johansson, C., Areskoug, H., Persson, K., & Järvholm, B. (2005). Comparative health impact assessment of local and regional particulate air pollutants in Scandinavia. *Ambio*, 34, 11–19.
- Garg, B.D., Cadle, S.H., Mulawa, P.A., & Groblicki, P.J. (2000). Brake wear particulate matter emissions. *Environmental Science & Technology*, 34, 4463–4469.
- Gidhagen, L., Johansson, C., Langner, J., & Olivares, G. (2004). Simulation of NOx and ultrafine particles in a street Canyon in Stockholm, Sweden. *Atmospheric Environment*, 38, 2029–2044.

- Gidhagen, L., Johansson, C., Langner, J., & Foltescu, V. (2005). Urban scale modeling of particle number concentration in Stockholm. *Atmospheric Environment*, 39, 1711– 1725.
- Gomiscek, B., Hauck, H., Stopper, S., & Preining, O. (2004). Spatial and temporal variations of PM1, PM2.5, PM10 and particle number concentration during the AUPHEP–project, *Atmospheric Environment*, 38, 3917–3934.
- Harrison, R.M., Shi, Ji Ping, & Jones, M.R. (1999). Continuous measurements of aerosol physical properties in the urban atmosphere. *Atmospheric Environment*, 33, 1037–1047.
- Harrison, R.M., Deacon, A.R., & Jones, M.R. (1997). Sources and processes affecting concentrations of PM10 and PM2.5 particulate matter in Birmingham (UK). *Atmospheric Envi*ronment, 31, 4103–4117.
- Hoek, G., Brunekreef, B., Goldbohm, S., Fischer, P., & van den Brandt, P.A. (2002). Association between mortality and indicators of traffic-related air pollution in the Netherlands: a cohort study. *Lancet*, 360, 1203–1209.
- Jacobson, T. (1994). Investigation of the road wear and light reflection characteristics of different road surfaces. (Only in Swedish: Undersökning av slitstyrkan och ljusreflektionsegenskaper hos vägbeläggning: Försök i VTIs provvägsmaskin). The Swedish National Road and Transport Research Institute (VTI), Report Notat 47–94, Linköping, Sweden.
- Johansson, C., Hadenius, A., Johansson, P.-Å., & Jonson, T. (1999). The Stockholm study on Health effects of Air Pollution and its Economic Consequences. Part I. NO₂ and Particulate Matter in Stockholm. AQMA Report 6:98, Stockholm Environment and Health Protection Administration, Box 38 024, 100 64 Stockholm, Sweden.
- Ketzel, M., Louka, P., Sahm, P., Guilloteau, E., Sini, J.-F., & Moussiopoulos, N. (2002). Intercomparison of numerical urban dispersion models—part II: street canyon in Hannover, Germany. *Water, Air, and Soil Pollution: Focus, 2*, 603–613.
- Ketzel, M., Wåhlin, P., Berkowicz, R., & Palmgren, F. (2003a). Particle and trace gas emission factors under urban driving conditions in Copenhagen based on street and roof-level observations. *Atmospheric Environment*, *37*, 2735–2749.
- Ketzel, M., Wåhlin, P., Kristensson, A., Swietlicki, E., Berkowicz, R., Nielsen, O.J., & Palmgren, F. (2003b). Particle size distribution and particle mass measurements at urban, near-city and rural level in the Copenhagen area and Southern Sweden. *Atmos. Chem. Phys. Discuss.*, *3*, 5513– 5546. (www.atmos-chem-phys.org/acpd/3/5513/).
- Keywood, M.D., Ayers, G.P., Gras, J.L., Gillett, R.W., & Cohen, D.D. (1999). Relationships between size segregated mass concentration data and ultrafine particle number concentrations in urban areas, *Atmospheric Environment*, 33, 2907– 2913.
- Krzyzanowski, M., Cohen, A., Anderson, R., & WHO Working Group. (2002). Quantification of health effects of exposure to air pollution. *Occupational and Environmental Medicine*, 59(12), 791–793.
- Kukklonen, J., Härkönen, J., Karpinen, A., Pohjola, M., Pietarila, H., & Koskentalo, T. (2001). A semi-empirical model for urban PM10 concentrations, and its evaluation against data from an urban measurement network. *Atmospheric Environment*, 35, 4433–4442.

- Lenschow, P., Abraham, H.-J., Kutzner, K., Lutz, M., Preuss, J.-D., & Reichenbächer, W. (2001). Some ideas about the sorces of PM10. *Atmospheric Environment*, 35(Supplement no. 1), 823–833.
- Longley, I.D., Gallagher, M.W., Dorsey, J.R., Flynn, M., Allan, J.D., Alfarra, M.R., & Inglis, D. (2003). A case study of aerosol (4.6 nm <Dp < 10 μm) number and mass size distribution measurements in a busy street canyon in Manchester, UK, 2003. Atmospheric Environment, 37, 1563–1571.
- Mårtensson, E.M., Nilsson, E.D., Buzorius, G., & Johansson, C. (2006). Eddy correlation measurements and parameterisation of particle emissions in an urban environment. *Atmospheric Chemistry & Physics*, 769–785, DOI: 1680-7324/acp/2006-6-769.å.
- Molnár, P., Janhäll, S., & Hallquist, M. (2002). Roadside measurements of fine and ultrafine particles at a major road north of Gothenburg. *Atmospheric Environment*, 36, 4115–4123.
- Nafstad, P., Lund Håheim, L., Wisløff, T., et al. (2004). Urban air pollution and mortality in a cohort of norwegian men. Environ Health Perspect., 112, 610–615.
- Norman, M., & Johansson, C. (2006). Studies of some measures to reduce road dust emissions from paved roads in Scandinavia. *Atmospheric Environment*, in press.
- Omstedt, G., Bringfelt, B., & Johansson, C. (2005). A model for vehicle induced non-tailpipe emissions of particles along Swedish roads. *Atmospheric Environment*, 39, 6088–6097.
- Pakkanen, T., Veli-Matti Kerminen, A., Korhonen, Christina H., Hillamo, Risto E., Aarnio, Päivi, Koskentalo, Tarja, & Maenhaut, Willy. (2001). Use of atmospheric elemental size distributions in estimating aerosol sources in the Helsinki area. Atmospheric Environment, 35, 5537–5551.
- QUARG. (1996). Airborne particulate matter in the United Kingdom. Quality of urban air review group, University of Birmingham, School of Chemistry, Edgbaston, Birmingham, UK. ISBN 0 9520771-3-2.
- Pitz, M., Kreyling, W.G., Hölscher, B., Cyrys, J., Wichmann, H.E., & Heinrich, J. (2001). Change of the ambient particle size distribution in East Germany between 1993 and 1999. *Atmospheric Environment*, 35, 4357–4366.

- Pope, C.A., 3rd, Thun, M.J., Namboodiri, M.M., Dockery, D.W., Evans, J.S., Speizer, F.E., *et al.* (1995). Particulate air pollution as a predictor of mortality in a prospective study of US adults. *American Journal of Respiratory Critical Care Medicine*, 151, 669–674.
- Querol, X., Alastuey, A., Ruiz, C.R., Artiñano, B., Hansson, H.C., Harrison, R.M., Buringh, E., ten Brink, H.M., Lutz, M., Bruckmann, P., Straehl, P., & Schneider, J. (2004). Speciation and origin of PM10 and PM2.5 in selected European cities. *Atmospheric Environment*, 38, 6547–6555.
- Rogge, W.F., Hildemann, L.M., Mazurek, M.A., & Cass, G.R. (1993). Sources of fine organic aerosol. 3. Road dust, tire debris, and organometallic brake lining dust: roads as sources and sinks. *Environmental Science and Technology*, 27, 1892–1904.
- Shi, J.P., Khan, A.A., & Harrison, R.M. (1999). Measurements of ultrafine particle concentration and size distribution in the urban atmosphere. *Science of Total Environment*, 235, 51–64.
- Tuch, Th., Brand, P., Wichmann, H.E., & Heyder, J. (1997). Variation of particle number and mass concentration in various size ranges of ambient aerosols in eastern Germany. *Atmospheric Environment*, 31, 4193–4197.
- Tunved, P., Hansson, H.-C., Kulmala, M., Aalto, P., Viisanen, Y., Karlsson, H., Kristensson, E., & Swietlicki, M., *et al.* (2003). One year boundary layer aerosol size distribution data from five Nordic background stations. *Atmos. Chem. Phys.*, *3*, 2183–2205.
- Van Dingenen, R., Raes, F., Putaud, J.-P., Baltensperger, U., Charron, A., Facchini, M.-C., Decesari, S., Fuzzi, S., Gehrig, R., Hansson, H.-C., *et al.* (2004). A European aerosol phenomenology–1: physical characteristics of particulate matter at kerbside, urban, rural and background sites in Europe. *Atmospheric Environment*, 38, 2561–2577.
- Westerlund, K.G., & Johansson, C. (2002). Emissions of metals and particulate matter due to wear of brake linings in Stockholm. In: Brebbia, C.A., & Martin-Duque, J.F. (eds.), *Air Pollution X*. Southampton, UK: WIT Press, ISBN 1-85312-916-X.