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Variability of particulate matter concentrations along roads and motorways determined by a moving measurement unit

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Abstract

The spatial variability of aerosol number and mass along roads was determined in different regions (urban, rural and coastal-marine) of the Netherlands. A condensation particle counter (CPC) and an optical aerosol spectrometer (LAS-X) were installed in a van along with a global positioning system (GPS). Concentrations were measured with high-time resolutions while driving allowing investigations not possible with stationary equipment. In particular, this approach proves to be useful to identify those locations where numbers and mass attain high levels ('hot spots'). In general, concentrations of number and mass of particulate matter increase along with the degree of urbanisation, with number concentration being the more sensitive indicator. The lowest particle numbers and PM_1-concentrations are encountered in a coastal and rural area: $<5000\,cm^{-3}$ and $6\,\mu g\,m^{-3}$, respectively. The presence of sea-salt material along the North-Sea coast enhances $PM_{>1}$-concentrations compared to inland levels. High-particle numbers are encountered on motorways correlating with traffic intensity; the largest average number concentration is measured on the ring motorway around Amsterdam: about $160\,000\,cm^{-3}$ (traffic intensity $100\,000\,veh\,day^{-1}$). Peak values occur in tunnels where numbers exceed $10^6\,cm^{-3}$. Enhanced PM_1 levels (i.e. larger than $9\,\mu g\,m^{-3}$) exist on motorways, major traffic roads and in tunnels. The concentrations of $PM_{>1}$ appear rather uniformly distributed (below $6\,\mu g\,m^{-3}$ for most observations). On the urban scale, (large) spatial variations in concentration can be explained by varying intensities of traffic and driving patterns. The highest particle numbers are measured while being in traffic congestions or when behind a heavy diesel-driven vehicle (up to $600\times10^3\,cm^{-3}$). Relatively high numbers are observed during the passages of crossings and, at a decreasing rate, on main roads with much traffic, quiet streets and residential areas with limited traffic. The number concentration exhibits a larger variability than mass: the mass concentration on city roads with much traffic is 12% higher than in a residential area at the edge of the same city while the number of particles changes by a factor of two (due to the presence of the ultrafine particles (aerodynamic diameter $<100\,nm$). It is further indicated that people residing at some 100 m downwind a major traffic source are exposed to (still) 40% more particles than those living in the urban background areas.

Keywords: Mobile measurements; Spatial variability; Traffic; Aerosol; PM_1; $PM_{>1}$; CPC; LAS-X

1. Introduction

Traffic is a major emission source of particles especially in urban agglomerations. Because of variations in vehicular intensity, driving behaviour, type and age of vehicles, and building infrastructure (open or connected, high or low), their spatial distribution will be highly inhomogeneous. Monitoring networks with

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stationary equipment give a quantitative impression of prevailing average concentrations near the instrumental site. Troublesome situations (''hot-spots'') however will usually not be discovered in the network's data.

The direct daily personal exposure to airborne particulate matter is therefore difficult to assess. Many epidemiological studies compare results of (statistical) health research with a (city) average. It is likely to result in (significant) errors of exposure (Ashmore, 2001). The influence of air pollution on people's health depends on the doses that organs receive, which is related to the individual's actual exposure, the individual's contact with an air-polluting component. Hence, the actual exposure requires detailed knowledge of the concentration variability at the various locations where people live or pass by. The goal of this study is to give more quantitative insight for typical Dutch circumstances.

To this purpose, differences in concentrations of particulate matter were measured for a number of cases. On the regional scale, the difference in mass and number concentration between a densely populated urbanised agglomeration and a clean marine region is of interest. On the local scale, concentration gradients for typical urban areas (suburban outskirt vs. centre area) or locations (e.g. when approaching a heavily trafficked road) are estimated here.

The experimental data presented here were collected in 1999 and 2000 with a moving (small) van. The vehicle contained high-time resolution equipment to measure on-line the ambient levels of mass and particle number. The use of mobile laboratories have become a common feature (e.g. Seakins et al., 2002) but performing measurements ''while driving'' is a rather new approach. During an extensive campaign Bukowiecki et al. (2002, 2003) measured on-road concentrations of trace gases and various aerosol parameters in a mountainous Swiss area. In their studies the suitability of the mobile-measurement approach for short- and long-term air-pollution investigations was shown. Another variant is the ''real-world'' measurement of the pollutants' concentrations in the freshly emitted gaseous exhaust produced by ''on-road'' vehicles pursued by the mobile laboratory on a short distance (e.g. Kittelson et al., 2000; Canagaratna et al., 2004).

In our campaign the mobile measurement technique was used along roads and motorways in different areas in the Netherlands, a comparatively flat country strongly influenced by the nearby presence of the North-Sea. Measured aerosol properties were restricted to mass, still the most important parameter from a regulating point of view, and particle number, subject to investigation in medical research with respect to possible adverse health effects (though not exclusively). The shortest length scale of interest here is that of an urban street (~ 30 m). Even with the low propagation speeds typical in urban agglomerations ($\sim 30\,\mathrm{km\,h^{-1}}$), parameters need to be measured with high-time resolutions (<10 s).

2. Experimental set-up

The high-time resolution equipment in this study consists of a Condensation Particle Counter (CPC3022, TSI Inc.) and a Laser Aerosol Spectrometer (LAS-X, PMS Inc.). The CPC counts the (total) number of particles in air with a diameter larger than 7 nm; its maximum detectable concentration is $10^7\,\mathrm{cm^{-3}}$. The LAS-X measures particles from 0.1 up to 7.5 μm in 15 size bins and registers particles greater than 7.5 μm in an oversize channel. The LAS-X data allows the determination of the volume concentration of particles for each size bin and the integrated volume concentration after summation over all bins. The aerosol mass is then estimated from the volume by assuming a density of $1.65\,\mathrm{g\,cm^{-3}}$. This is based on a comparative experiment between calculated mass concentrations derived from LAS-X with filter measurements and in agreement with Tuch et al. (2000, and references therein). Even though the exact value of aerosol density may differ from $1.65\,\mathrm{g\,cm^{-3}}$ this is not of vital importance as we are predominantly interested in relative concentration levels.

Both measuring devices were installed in a van equipped with an online data acquisition system. The inlet is placed on top of the roof at the front-side of the car (2.5 m above the ground) and directed forwards. This position is chosen to avoid influence from the van's own exhaust emissions at its lower rear end (at a height of 30 cm), and to reduce the effect of dust blown up by wind and car movement as much as possible. The possible effect of the van exhaust on the measurements was assessed by driving and standing at a low-concentration location, and found to be negligible.

The time resolution of the CPC and LAS-X measurements was 2 and 10 s, respectively; in case of non-urban measurements the CPC-data were averaged over 10 s. The exact location while driving was registered by GPS. In total, data of three 1-day campaigns have been used in this study. These campigns took place in the coastal and rural areas of the province of Noord-Holland (near the North-Sea shore in the northwest of the Netherlands, see Fig. 1) and in the cities Amsterdam and Nijmegen.

The accuracy of particle concentration measurements for $PM_{2.5}$ is within 20% as was estimated by comparing these instruments with other aerosol counters and spectrometers (Khlystov et al., 2001; Mirme et al., 2002). Hinds and Kraske (1986) showed that the LAS-X sampling efficiency decreases for this fraction: about 30% loss of 10 μm particles at the sampling rate of $5\,\mathrm{cm^3\,s^{-1}}$. Hence, the coarse fraction (here defined as all particles larger than 1 μm) will be underestimated.

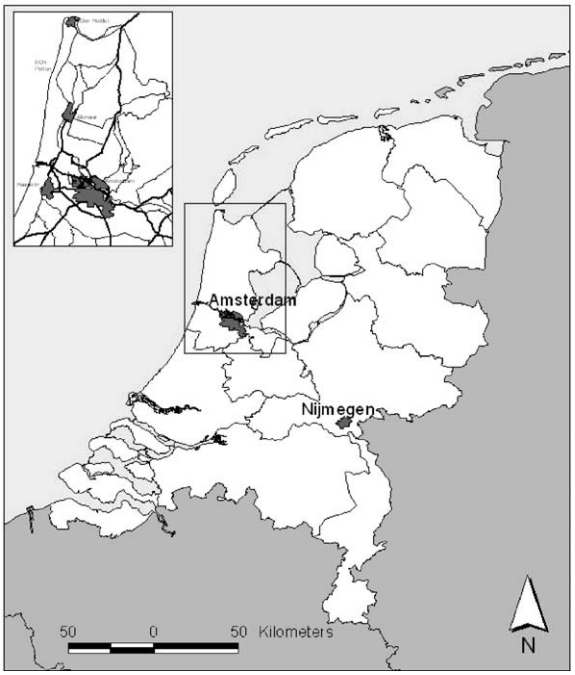

Fig. 1. Locations of the province of Noord Holland, Amsterdam and Nijmegen in the Netherlands.

Wind-driven particles belong mostly to this fraction. Due to the height of the inlet and the measuring range of the LAS-X, their contribution is expected to be limited in our measurements.

3. Results and discussion

3.1. Regional variability

3.1.1. Aerosol number concentration

Fig. 2 shows the particle number variability measured during a 1-day campaign (27 October, 2000) in the province of Noord, Holland. The trip went from the Amsterdam urban area to the ECN site at Petten near the North-Sea coast. The dominant wind direction was SSW. The instantaneous measurements were averaged over 500 m. In general, the number concentration appears to relate to the intensity of traffic in the province: a gradual increase is found when going from the north to the more densely populated regions in the southwest. Along the way several characteristic areas can be identified. First, near the coast and in the adjacent rural area (up to the city of Alkmaar) with predominantly marine air is characterised by the lowest numbers: $<3\times10^3\,\mathrm{cm}^{-3}$ in the immediate proximity of the sea. In this area the traffic is relatively low, but the (few) passages of oncoming cars incidentally raise numbers to above $20\times10^3\,\mathrm{cm}^{-3}$ for short time periods.

The second part of the route is the passage westerly of Alkmaar (a small town with 90 000 inhabitants), a busy 4-lane through-traffic road with a number of traffic lights. From the moment the municipal borderline has been crossed, particle numbers exhibit a base level (i.e. concentrations are visually higher for most measurements) of $20\times10^3\,\mathrm{cm}^{-3}$. In this rural area not far from the North Sea the background air is still relatively clean. Occasionally, instantaneous concentrations rise above $80\times10^3\,\mathrm{cm}^{-3}$ due to the stop-and-go car movements at the traffic lights.

The third part is the A9-motorway between Alkmaar and the ring motorway in Amsterdam (750 000 inhabitants) that shows that baseline levels further increase to some $40\times10^3\,\mathrm{cm}^{-3}$. On the Amsterdam ring motorway the base level is $80\times10^3\,\mathrm{cm}^{-3}$; here maximum concentrations ($>160\times10^3\,\mathrm{cm}^{-3}$) are found over extended tracks, in particular along the western and southern leg of the ring. Even by averaging over a distance of 500 m, variability in number appears still large in the centre of Amsterdam: levels are between $5\times10^3\,\mathrm{cm}^{-3}$ to well above $160\times10^3\,\mathrm{cm}^{-3}$. The highest instantaneous concentrations ($\sim10^6\,\mathrm{cm}^{-3}$) are encountered when driving inside tunnels.

3.1.2. Aerosol mass concentration

The particle-mass estimations are given in Figs. 3a,b for PM_1 (particles with an aerodynamic diameter smaller than 1 μm) and $PM_{>1}$ (particles larger than 1 μm), respectively. Distance of averaging is again 500 m. In case of PM_1 the measured aerosol mass is in the range 0.1–1 μm (accumulation mode), because the size segregated information is only available in this size range.

PM_1-concentrations were mostly below $9\,\mu\mathrm{g\,m}^{-3}$. In the marine and rural area (between the coast and Alkmaar town) concentrations are $<6\,\mu\mathrm{g\,m}^{-3}$. More elevated levels (i.e. above $9\,\mu\mathrm{g\,m}^{-3}$) occur at locations characterised by an increase in traffic intensity (like the busy provincial road between Haarlem and Amsterdam), by a lower air dilution (inside tunnels), and at locations near Alkmaar and in the Amsterdam city centre where stop-and-go traffic prevails (note the enhancement at the road junction in the very centre). Fig. 3b shows the contribution of the larger particles ($PM_{>1}$, upper detection limit 7 μm). In general, this contribution is relatively small. One difference is found in the coastal area northwest of Alkmaar where the contribution of the larger seasalt aerosols is measured ($6–8\,\mu\mathrm{g\,m}^{-3}$); in this respect the decrease of $PM_{>1}$ up to the city border of Alkmaar is notable. Significant $PM_{>1}$-contributions are further noticed near Alkmaar and in the Amsterdam agglomeration, probably related to the re-suspension of road dust or direct emission of

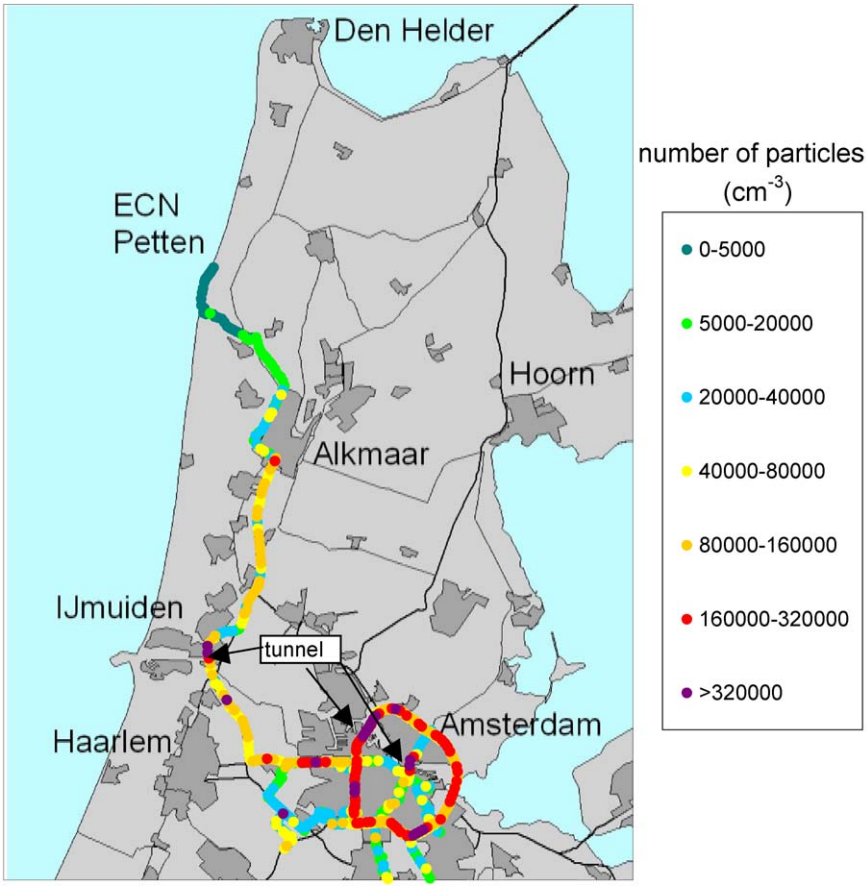

Fig. 2. Particle number concentrations along the way from the urban agglomeration of Amsterdam to the marine area near Petten (averages over 500 m; CPC-measurements).

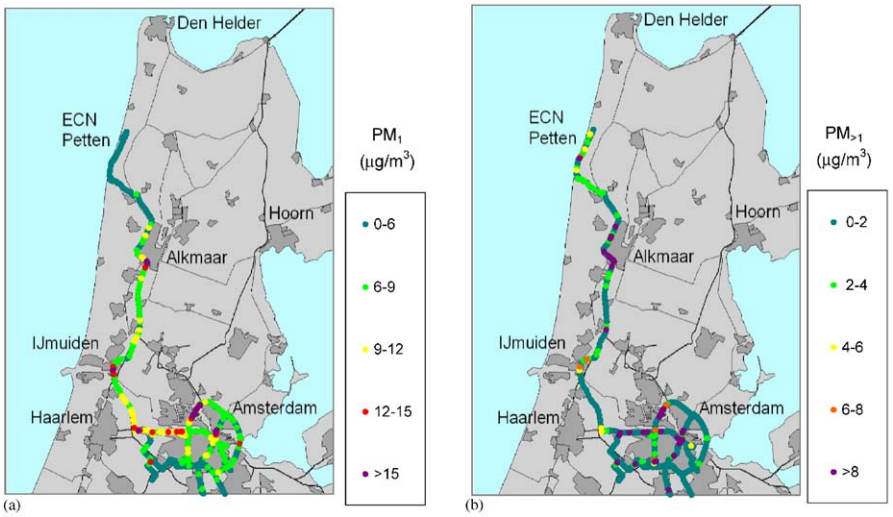

Fig. 3. As Fig. 2 but now for the mass fractions (a) PM_1 and (b) $PM_{>1}$ (LAS-X measurement range: 0.1–7 µm).

larger particles by the traffic (e.g. coarse soot particles re-entrained from the exhaust pipe).

On average, the largest contribution here originates from PM_1. Measurements on the motorway indicate increased PM_1-levels with respect to the rural or marine areas. The mass concentration immediately near the seacoast (distance within 1 km) is higher than in the rural region further inland and is due to particles with sizes larger than 1 μm (sea-salt aerosol). A remarkable increase of particles with large masses is observed in the urbanised area of Amsterdam. Most likely this can be attributed to the single events in which measurements take place in tunnels and the short time periods in which the measurement unit is near or inside emission plumes of heavy trucks or buses. In these cases concentrations might rise to above $400\,\mu g\,m^{-3}$; the emission of such vehicles also contains a number of particles with diameters above 1 μm (also see the next section).

3.2. Variability of number and mass concentrations within a city

3.2.1. Examples of time series

The variability of mass and number of particles in urban air is demonstrated in Figs. 4a–d that show four time series of aerosol concentrations measured in the cities of Nijmegen (19 March, 1999) and Amsterdam (26 October, 2000). Fig. 4a contains measurements during a traffic congestion (at the Berg en Dalseweg) in a quiet side street (Tooropstraat) and again at the Berg en Dalseweg (now with a great deal of traffic passing through). In the congestion line (13:42–13:50), the measuring inside almost undiluted exhaust plumes of vehicles standing still, accelerating or decelerating leads to massive fluctuations: standard deviations for this period are $7.7\,\mu g\,m^{-3}$ and $1.3\times10^5\,cm^{-3}$, respectively.

The concentration levels in the quiet side street are much more stable: mass and number decrease rapidly and standard deviations are now $2.3\,\mu g\,m^{-3}$ and $2.9\times10^4\ cm^{-3}$. An increase in variability marks the moment when the vehicle entered the Berg en Dalseweg again (around 14:56). Obviously, standard deviations ($4.2\,\mu g\,m^{-3}$; $1.1\times10^5\,cm^{-3}$) are higher than in the Tooropstraat but lower than what is measured in the congestion line. A larger distance between vehicles and the turbulence aroused by the driving enable dilution to a larger extent than in the traffic jam.

A different situation is observed in Fig. 4b where measurements in a residential area of Nijmegen are shown. The nearly total absence of traffic around this time of day (approximately 15:30) is reflected in low masses (base line value around $12\,\mu g\,m^{-3}$) and numbers ($2.5\times10^4\,cm^{-3}$). Standard deviations are $1.8\,\mu g\,m^{-3}$ and $2.3\times10^4\,cm^{-3}$, respectively. The limited variability in the series is likely to reflect fluctuations in background concentrations. The same holds for the series measured in the quiet municipality of Beuningen (10 km northwest of Nijmegen) demonstrated in Fig. 4c. Here a period of at least 50 s is present in which the measuring took place while driving directly behind a diesel-powered bus. Mass and number concentration levels go up to some $55\,\mu g\,m^{-3}$ and $600\times10^3\,cm^{-3}$, clearly demonstrating the negative impact of emissions of a heavy diesel vehicle on local air quality.

Fig. 4d shows a particle number series registered when driving in one of the Amsterdam tunnels; wind direction was almost parallel to the tunnel line. Clearly, the number distribution inside the tunnel is nonsymmetrical. Turbulence aroused by wind and moving cars possibly cause the diminishing dilution in the first halve. In the midpart of the tunnel the increase vanishes which may be due to lower car emissions going down hill and perhaps coagulation at these very high concentrations ($>10^6\,cm^{-3}$). To overcome the slope to the tunnel ending, drivers step harder on the gas pedal inducing a further rise decreasing abruptly near the end of the tunnel.

3.2.2. Average concentrations in urban air

The figures above demonstrate that variability of mass and number concentrations on a local scale can be interpreted by studying the presence of traffic and driving behaviour at those locations. This feature leads to the grouping of the collected dataset on basis of the characteristics of the routes traversed. Selected here are residential areas, streets, through traffic routes (traffic lights excluded) and crossings. Traffic routes are characterised by an intensity of more than $10\,000\,veh\,day^{-1}$; for a street this is $<6600\,veh\,day^{-1}$ (these data are provided by the municipality of Nijmegen). For residential areas, traffic intensity has been estimated to be $<2\,veh\,min^{-1}$ (i.e. a maximum of $2800\,veh\,day^{-1}$). For each subset of corresponding routes, weighed averages and standard deviations were calculated for mass and number concentrations, with the weighing factor being the number of measurement points in each individual trajectory. Results of these computations are shown in Fig. 5 for the various locations. The indications 'congestion' and 'bus' refer to the measurements during periods described in Section 3.2.1 and Figs. 4a and c, and have been added for comparison.

Clearly, the increasing presence of traffic is reflected in increasing levels and standard deviations for mass and number concentrations. The average number concentrations are lowest ($5.9\times10^4\,cm^{-3}$) in residential areas in the outskirts of the city. Higher number concentrations are found on streets and roads with more traffic, in the "quiet" streets concentrations are up by some 40%, on the "busy" traffic routes this percentage is 83% while at crossings particle number concentrations are more than doubled compared to the levels in residential areas. It is

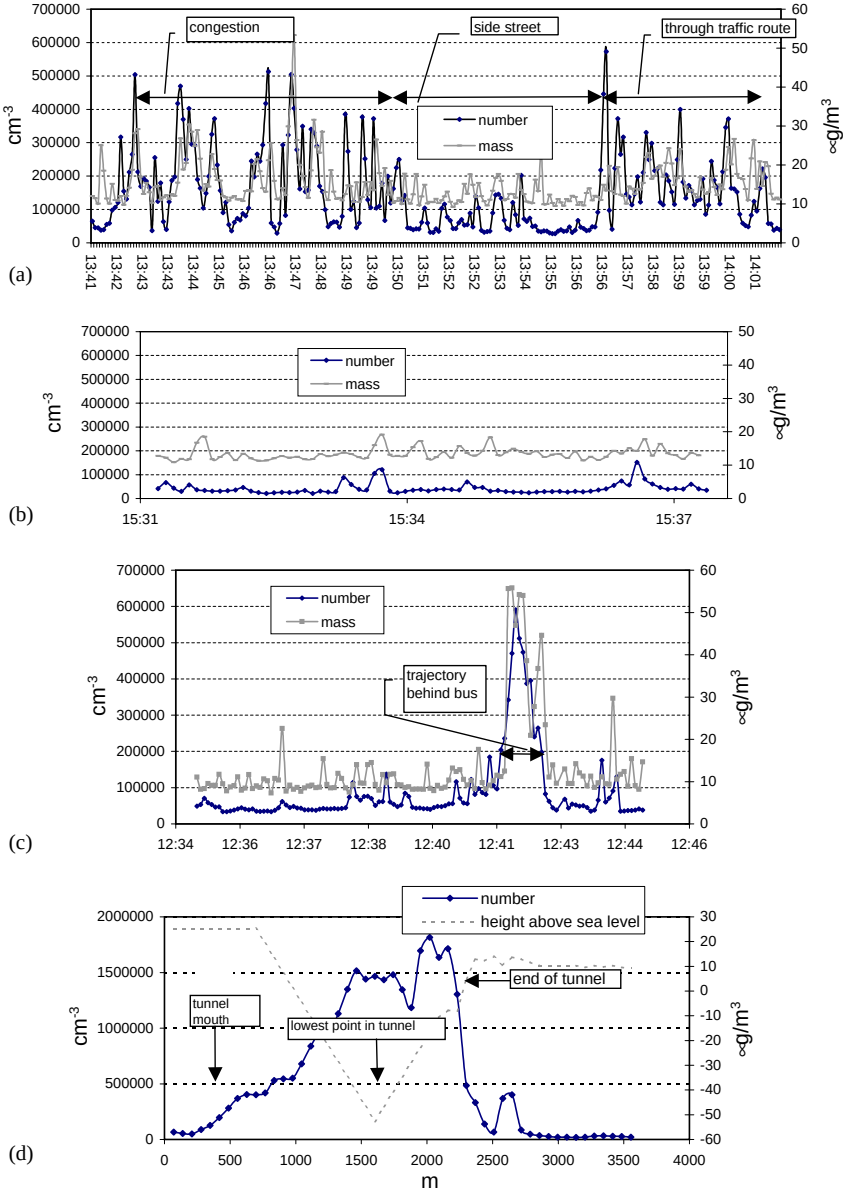

Fig. 4. Mass and number of particles on three trajectories in Nijmegen: (a) congestion until 13:50), Tooropstraat (side street, until 13:56) and Berg en Daalseweg (traffic route), (b) residential area, (c) Beuningen, and (d) in an Amsterdam tunnel (also given height a.s.l.).

further noted that the increase in mass concentrations does not keep up with this pace: between residential areas and crossings the mass increase amounts to merely 12%. Being more sensitive, particle number seems to be a better indicator for the presence of traffic in urban areas than mass. Very high concentrations of mass (+29% compared to residential area) and number (almost 4 times as high) are found in the congestion. Not surprisingly, the concentrations measured directly behind the diesel bus (approx. $38\,\mu g\,m^{-3}$ and $3.6\times10^5\,cm^{-3}$, respectively) are the highest that were measured this day.

3.2.3. Particle size distribution

Fig. 5 suggests that an increase in traffic intensity results in higher numbers of particles in the air. The mass and number distributions as a function of particle size confirm this. Figs. 6a,b illustrate how number and mass are distributed over 11 diameter classes recalculated from the LAS-X data. The LAS-X registers

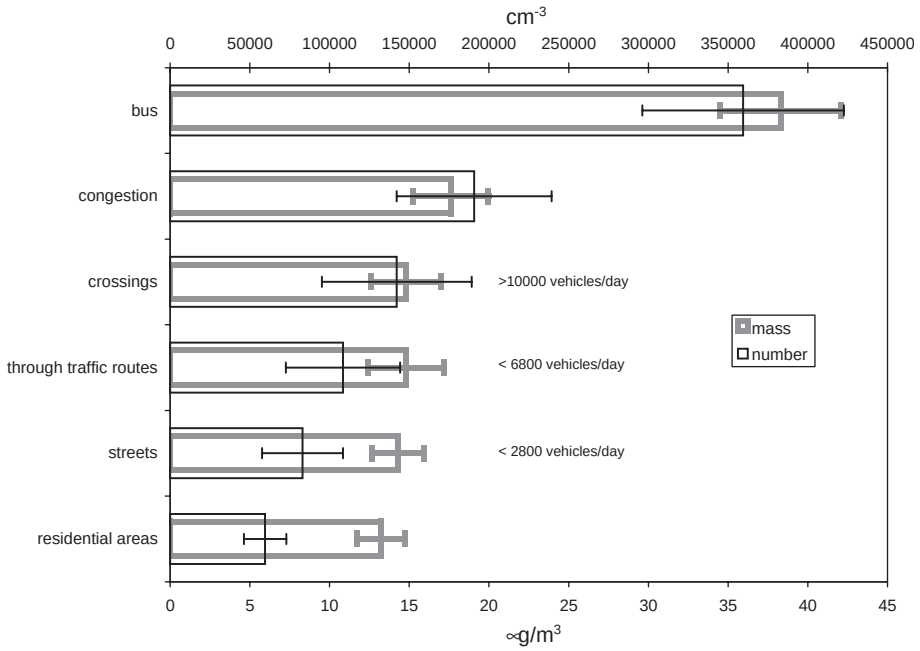

Fig. 5. Average concentrations and standard deviations for total mass and number of particles at typical locations in the city of Nijmegen.

particles between 0.1 and 7.5 μm; in the figures the upper limit of each size class is given along the horizontal axis. The number of particles in the ultrafine fraction (<0.1 μm) is added to Fig. 6a. Because the LAS-X measures particles with diameters above 100 nm and the CPC has a lower detection limit of 7 nm, the difference is a measure of number concentration of ultrafine particles.

Fig. 6a shows that most of the registered particles have diameters smaller than 0.5 μm; this is irrespective of location. The ultrafine fraction shows high numbers in case of measurements at crossings, in congestions and behind a diesel bus. The numbers of larger particles (>0.5 μm) turn out to be too small to indicate. The mass distribution (Fig. 6b) appears bimodal. The first mode corresponds to the coarse particles (>1 μm). Though few in number, these make a substantial contribution to the aerosol mass. The second mode is identified as the accumulation mode (between 0.2 and 0.4 μm) and the result of coagulation and accumulation of aerosol mass from gas-to-particle conversion processes. The mass distribution decreases below 0.2 μm. Despite their large numbers, the ultrafine fraction does not contribute substantially to the total mass. With a number-median diameter between 10 and 20 nm for the ultrafine mode in urban air (Whitby and Sverdrup, 1980), and the numbers seen in Fig. 6a, its corresponding mass at residential areas, in streets and at through-traffic routes would be negligible. Only when measuring directly behind the diesel bus the ultrafine mass contribution could be as high as $3\,\mu g\,m^{-3}$. It is concluded that both accumulation and coarse mode are present in the mass distributions at the different urban locations; the ultrafine (nuclei) mode dominates the number distribution. These results are comparable with the findings for urban air as published by Whitby and Sverdrup (1980) and Keywood et al. (1999).

The size distributions at the various locations appear rather similar. Yet, there are some interesting differences. The fraction of particles with sizes between 0.1 and 0.2 μm increases on through traffic routes and on crossings with the enhanced presence of traffic (Fig. 6b). The mass concentration in this size range measured on the through traffic routes is some 30–40% larger than in the residential areas. The increase in the 0.2–0.3 μm range is less pronounced and even negligible in larger-size classes. This observation is consistent with the contribution of traffic emission. Internal combustion engines are known to emit predominantly ultrafine particles (e.g. Kittelson et al., 2000). The effect of traffic emissions on ultrafine particle formation in urban areas is described by Woo et al. (2001).

The LAS-X measurements behind the diesel bus also indicate the presence of a coarse mode in the mass distribution. Obviously, the "in-plume" size distributions are a combination of both exhaust and ambient aerosol. Part of it will be due to resuspension. However, diesel exhaust particle size distributions are found to be

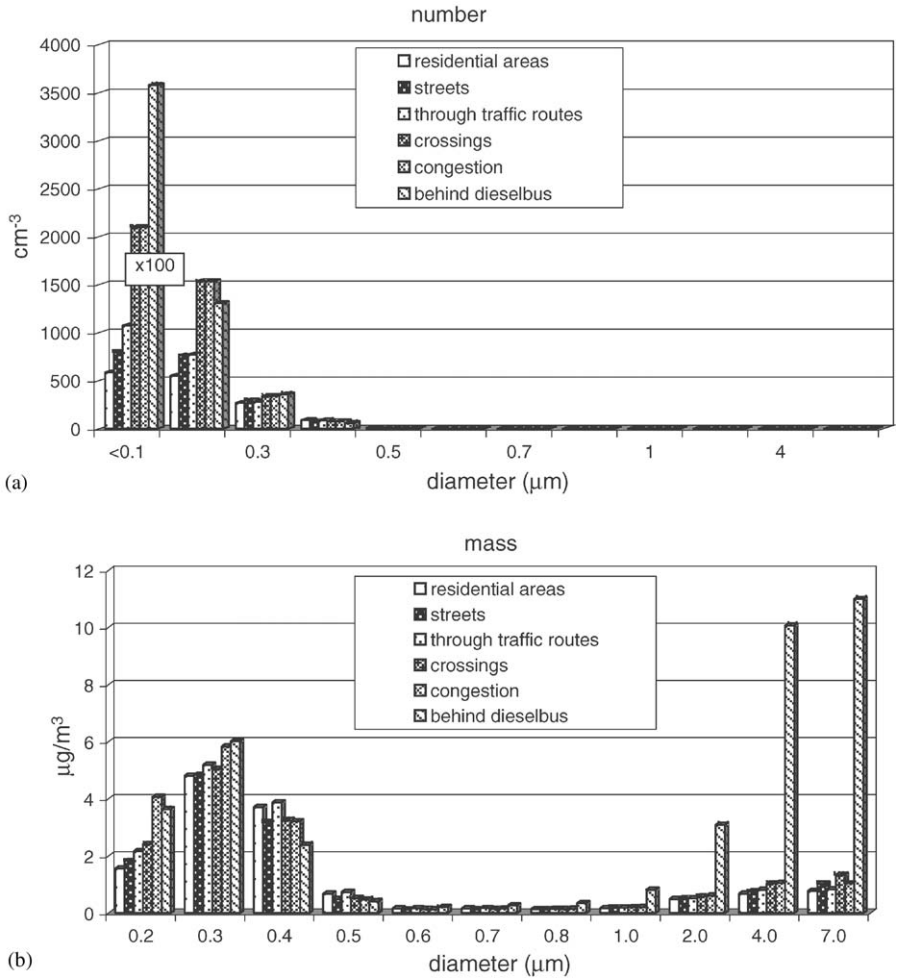

Fig. 6. Number (a) and mass (b) concentrations for 11 particle-size classes measured at various locations in Nijmegen.

typically trimodal (Kittelson, 1998; Worsnop et al., 2002).

3.2.4. *Decrease of concentrations as a function of distance from the road*

Assessments of human exposure to vehicular pollution require knowledge of concentration changes near roads. To demonstrate how on-road measurements can be used to assess such a gradient, nine specific periods were selected in the Nijmegen data set (19 March, 1999). During each series the van drove off a heavily trafficked route (more than 10 000 veh day^{-1}) into a quiet side-street. Each series lasted at least 30 s. Based on these data, average horizontal gradients for PM_1, $PM_{>1}$ and total number were estimated by an 'ensemble' averaging procedure.

In the computation of ensemble averages the positional data of each of the nine time series was divided into 10 equally spaced 'bins'; each bin comprises position data (with respect to road axis) with corresponding concentration with bin 1 containing the concentrations measured on the road (position value: 0 m) while bin 10 is 'filled' with data belonging to the position when difference between actual and background concentration has become near zero. Naturally, the total number of data points depends on the duration of a time series and is the same for each bin. After this, data were averaged for both concentration and position. For more details see Wilczak (1984). Resulting gradients are shown in Fig. 7. The urban background concentration has already been subtracted.

The figure indicates that both mass and number aside a road behave like exponentially decaying functions. The decay parameter (denoted by a in the legend of Fig. 7) is largest in the case of $PM_{>1}$, i.e., the return to background level is fastest for $PM_{>1}$. This is not

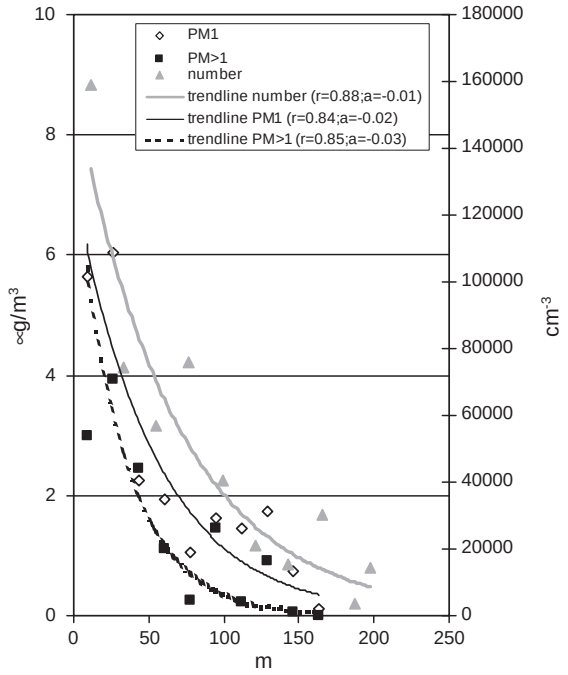

Fig. 7. Estimated ('ensemble') average gradients and best-fit exponential functions for mass and number aside an urban through-traffic route; the city background level has been subtracted.

surprising, as large particles are removed faster due to sedimentation. The particles in the PM_1-fraction remain longer in the air and may travel over a greater distance. The distance over which the $PM_{>1}$-contribution has decreased (e-fold) with 90%, is about 75 m; in case of PM_1 and number this is around 120 and 150 m, respectively. Also, at a distance of 100 m the number of traffic-related particles is still some $40 \times 10^5\,cm^{-3}$ above the background level.

4. Conclusions

A moving measurement system has proven to be a useful tool to study spatial variability of particle concentrations in various areas, and urban areas in particular. It allows investigation of location-specific characteristics that cannot be performed with a single or multiple stationary monitoring sites and 'ordinary' filter measuring. Repetition of these measurements in different periods as well as on other routes will further increase the reliability of these spatial and temporal studies.

The number and mass concentrations have been observed to increase with increasing traffic intensity. Highest concentrations are found on motorways, road crossings, and inside tunnels. Aerosol mass concentrations along roads with high traffic density are approximately 12% higher than in residential areas in the same city, while particle number concentrations are 2 times higher. The number of particles is a more sensitive indicator of the contribution of traffic than the aerosol mass, because traffic emits mostly ultrafine particles that dominate the number, but not the mass.

It is known that number concentration is not a unique indicator for traffic and that nucleation may produce very high number numbers even in remote areas (Baltensperger et al., 2002). However, these events take place on a much larger scale (hundreds of kilometers) as recent studies suggest (e.g. Stanier et al., 2004). In contrast, our measurements clearly show that the number concentration inside a city changes on a scale of a hundred meters, and that these fluctuations correlate with the local traffic intensity and driving conditions.

Aerosol concentrations decrease exponentially with increasing distance from the road. The distances along which the traffic contribution is reduced by 90% are 75 m for $PM_{>1}$, 120 m for PM_1 and 210 m for the number concentration. It is further shown that people residing at some 100 m downwind a major traffic source are exposed to (still) 40% more particles than those living in the urban background areas.

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