Differences among Black Smoke, PM_{10}, and $PM_{1.0}$ Levels at Urban Measurement Sites

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In Amsterdam, the Netherlands, we measured airborne particulate matter (PM) during winter 1998–1999, taking daily average measurements at an urban background site, at a busy street, and at a motorway. Comparison of black smoke, PM_{10}, and $PM_{1.0}$ levels showed that daily averages were highly correlated over time. Median daily concentrations were elevated at sites affected by traffic. The highest increase relative to the background in median daily concentration was noted for black smoke at the motorway (300%), whereas for PM_{10} and $PM_{1.0}$ the increase was only 37% and 30%. These results indicate that mass measurements of ambient particulate matter underestimate the exposure to particles generated by traffic. *Key words*: black smoke, exposure assessment, PM_{10}, $PM_{1.0}$, particulate matter, traffic. *Environ Health Perspect* 109:151–154 (2001). [Online 24 January 2001]
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Studies in the last two decades have shown associations between particle mass and adverse short- and long-term health effects (*1–4*). The focus shifted from coarse particles such as total suspended particulates (TSP) and particles with a 50% cut-off aerodynamic diameter of 10 µm (PM_{10}) to finer particles such as those with a 50% cut-off aerodynamic diameter of 2.5 or 1.0 µm ($PM_{2.5}$, $PM_{1.0}$).

Amsterdam, the Netherlands, has about 700,000 inhabitants and relatively little heavy industry, so the main sources of air pollution are emissions from long-range transport and traffic. In 1998 the municipal air pollution monitoring network of Amsterdam was re-equipped to estimate the impact of air pollution on public health. About 10% of the population lives along roads traveled by more than 10,000 motorized vehicles per day (population registration, Amsterdam). We measured pollution at two sites along busy roads and urban background concentrations at another site. Measurements at this stage included PM_{10}, $PM_{2.5}$, and $PM_{1.0}$. We measured black smoke to compare measurements of earlier years, because it indicates emissions of diesel engine vehicles (*5*), and because black smoke rather than PM_{10} was associated with daily mortality in Amsterdam (*6*).

Here we describe the differences among sites measuring daily concentrations of particulate matter (PM) in background and traffic sites in autumn and winter 1998–1999.

Materials and Methods

Sites. In the Netherlands, an urban measurement site is considered a background site if in a circle of 35 m around that site fewer than 2,750 motor vehicles pass during 24 hr. We measured background concentrations at a site positioned at the edge of a large park (Vondelpark), which is the center of the city. The nearest road is about 50 m away and is separated from the measurement site by a four-story hospital.

One traffic-influenced site is at the Stadhouderskade, a street running along the edge of the oldest part of town with a legal speed limit of 50 km/hr. The site is situated next to an intersection with traffic lights. The other traffic-influenced site is at the Einsteinweg, a 4–6 lane motorway that encloses the center of Amsterdam and has a legal speed limit of 100 km/hr. The inlets are placed at the roadside, above the crash barrier (Table 1).

We measured hourly PM_{10} and $PM_{1.0}$ concentrations with tapered element oscillating microbalance (TEOM), model 1400A, operating at 50°C and equipped with size-specific inlets (Rupprecht and Patashnick, Albany, NY, USA). The total flow through the sampling heads was 16.67 L/min; this was split into a flow of 3 L/min through the filter and into a 13.67 L/min bypass flow. We calculated mass concentrations using the standard algorithm provided by the manufacturer.

We measured 24-hr concentrations of black smoke with SX-200 black smoke continuous monitors (ETL, Hereford, England) operating with a sampling flow of 1.38 L/min through Whatman No. 1 paper filters with inverted funnels as sampling inlet. Sampling of particles, reflectance measurements, and calculation to mass concentrations were done according to specifications of the European community (*7,8*).

Data analysis. We calculated daily means over autumn and winter 1998–1999 (10 October 1998 to 31 March 1999) by averaging hourly values of the TEOM measurements. Days with more than 25% of missing hour values were noted as missing. In addition, if a component was missing, the same component was noted as missing on the other stations. Differences in distribution were tested with Wilcoxon rank-sum test; correlation coefficients were calculated with Spearman rank–order correlation.

Results

A description of the 24-hr mean values is given in Table 2. PM_{10}, $PM_{1.0}$, and black smoke are lowest at the background site, higher at the street site, and highest at the motorway. The relative increase for each component differs among the traffic-influenced sites and the background site. The increase in median PM_{10} concentration is 16% at the street site and 37% at the motorway. For $PM_{1.0}$ this increase is 20% and 30%, respectively. For black smoke the increase is 138% (street) and 300% (motorway). The 24-hr mean values are plotted against date in Figures 1–3. The patterns of $PM_{1.0}$ and PM_{10} on the background and street sites are very similar (Figures 1 and 2). The pattern of PM_{10} at the motorway differs somewhat from that of PM_{10} on background and street, whereas the pattern of $PM_{1.0}$ at the three sites is very similar. This is reflected in the correlation coefficients in Table 3, which are very high among $PM_{1.0}$ on background, motorway, and street, and between PM_{10} background and PM_{10} street. In Figure 3 black smoke concentrations are plotted against date. At the motorway and street sites the variability is larger compared to background but the correlation coefficients between background and the other two sites are still high.

Table 4 shows the correlation coefficients between different components at the same site. At the motorway site the correlation between the components is higher than at the other two sites.

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Discussion

The measurements at the three sites indicate clear differences in particle concentration not only among the background site and the traffic sites but also between the two traffic sites. Median levels differed among the sites whereas the correlation over time among the same components on different sites was high. The high correlation over time indicates that meteorological conditions such as wind speed, wind direction, and precipitation significantly influence the daily levels. The differences among the distributions are most strongly pronounced in the black smoke concentrations and less so in the $PM_{1.0}$ and PM_{10} concentrations.

A part of the increase in PM_{10} mass can be attributed to the increase in $PM_{1.0}$ mass. $PM_{1.0}$ particles result from combustion sources, condensation of volatile species, and gas-to-particle conversion (*9*). The rest of the increase comprises particles larger than 1 µm. The mass of this fraction is dominated by particles that have a crustal origin (*9*) and might be resuspended by passing traffic. This is supported by Janssen et al. (*10*), who found elevated levels of elements with a crustal origin such as iron and silicon in PM_{10} street samples compared to PM_{10} sampled at background locations. That the two traffic-influenced sites differ in their PM_{10} and $PM_{1.0}$ levels can be attributed to differences in traffic density and in average speed of the passing traffic. The street site is near a traffic light, which lowers the average speed. In addition, the speed limit is 50 km/hr at the street site and 100 km/hr at the motorway site. Thus the average speed of the motorway traffic is higher and this can cause more resuspension of PM_{10} particles. In addition, the high correlation between the different components on the motorway site indicates a common source.

The most striking result is the large increase of black smoke at the traffic-influenced sites. Black smoke is measured by the reflectance of a sampled filter (*7*) and does not account for the portion of aerosol mass that does not absorb light (*9*). The main fraction of light-absorbing particles in ambient air is formed by elemental carbon (*11*), and the size of the particles sampled with the black smoke sampler is below 5 µm (*9*). The formulas used to transform reflectance to mass concentrations are based on older studies in which the air pollution mixture was different from the current composition (*7*). Therefore, the black smoke figures should be interpreted as an indication of elemental carbon, not as mass concentrations. The increase in black smoke is highest at the motorway, the site with the highest traffic density, highest average speed, and the highest number of trucks. These levels are probably caused by diesel engines because diesel contributes to elemental carbon levels (*5*). Besides practically all of the trucks, 11% of the passengers cars have diesel engines in the Netherlands (*12*).

One reason for the larger increase of black smoke compared to PM_{10} and $PM_{1.0}$ may be the different number of observations for each component during the measurement period. Instrument malfunctioning meant that many observations of black smoke were missing. However, after we reduced the data set to days where all components on all sites were present (n = 65) the results remained the same (Table 2).

Another reason for the difference between the black smoke and the PM_{10} and $PM_{1.0}$ measurements might be the method. PM_{10} and $PM_{1.0}$ were measured with TEOM equipment which heats the sampled particles to 50°C, causing evaporation of semivolatile components and particle-bound water. Compared to manual gravimetric methods, which measure at ambient temperatures, this method may underestimate particle mass. This underestimation might be higher at the traffic sites due to the larger contribution of semivolatile components from vehicle emissions. However, comparisons between manual gravimetric methods and TEOM in various cities with differing traffic intensities and seasons showed that the underestimation of the mean concentration was at most 38% (*13*). This underestimation is not large enough to explain the relatively smaller difference of PM concentrations

Table 1. Description of measurement sites.

Sampling site	Sampling height	Distance to road axis (m)	No. motorized vehicles per day	No. trucks per day
Background	4 m	–	–	–
Street	4 m	7	30,000	1,000
Motorway	3 m	12–14	94,000	9,000

Table 2. Description of 24-hr mean concentrations in µg/m^3.

Sampling site	No. observations	Percentile 5th	Percentile 50th	Percentile 95th	Max	50th complete
Background						
PM_{10}	167	12	19	36	74	19
$PM_{1.0}$	128	5	10	20	49	10
Black smoke	109	2	8	20	47	7
Street						
PM_{10}	167	15	22*	40	78	21
$PM_{1.0}$	128	6	12*	23	53	12
Black smoke	109	8	19*	40	59	18
Motorway						
PM_{10}	167	15	26*	52	88	27
$PM_{1.0}$	128	6	13*	26	52	14
Black smoke	109	6	36*	93	159	36

Abbreviations: max, maximum; P_{50} complete, median value using days with complete measurement on all sites (n = 65).
*p < 0.01 Wilcoxon rank-sum test comparing street or motorway to background.

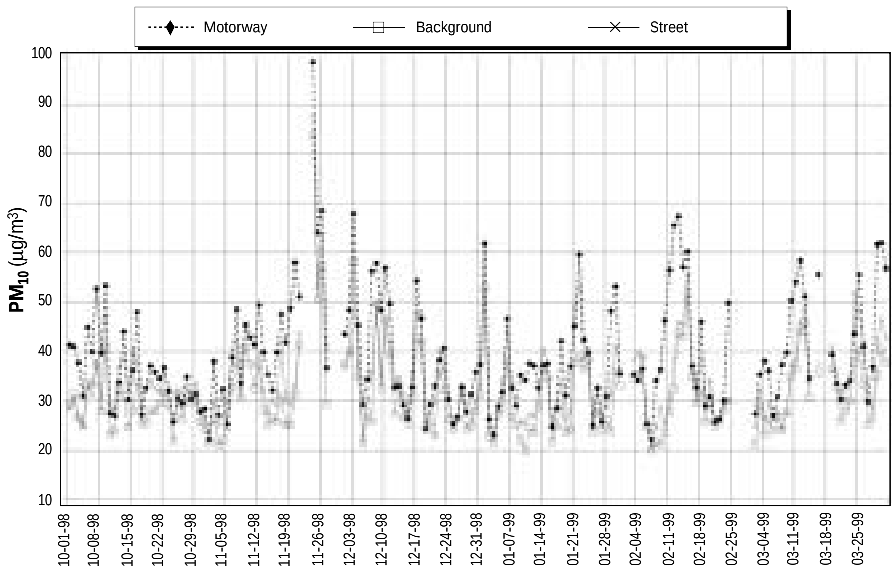

Figure 1. Daily PM_{10} concentrations (µg/m^3) at motorway, street, and background sites, against date.

between the sites compared to the difference in black smoke concentration. Other studies not using TEOM equipment have also reported a larger increase in elemental carbon or black smoke than in particle mass measurements when comparing measurements of different exposure to traffic emissions. A study conducted in Harlem, New York (USA), comparing sidewalk sites on roads with low or high bus and truck counts found that elemental carbon was 4 times higher and $PM_{2.5}$ only 1.3 times higher (*14*). In a study in Arnhem, The Netherlands, PM_{10} and $PM_{2.5}$ concentrations were on average 1.3 higher at a street site than at a background site, whereas the black smoke concentrations were 2.6 times higher (*10*). In another study in the Netherlands, ambient measurements of PM_{10}, $PM_{2.5}$, and black smoke were made at different distances from motorways. Black smoke but not PM_{10} and $PM_{2.5}$ decreased with increasing distance from the freeway, up to about 250 meters (*15*). This indicates that elevated exposure can occur also at distances further away from the road than the measurement sites. Finally, a study in Amsterdam showed that homes along busy roads had 15–20% higher outdoor levels of PM_{10} and $PM_{2.5}$ compared to homes along quiet roads. Total polycyclic aromatic compounds, benzo[*a*]pyrene, absorption coefficient of particle filters, and benzene differed by a factor of two between the two types of homes, outdoor as well as indoors (*16*).

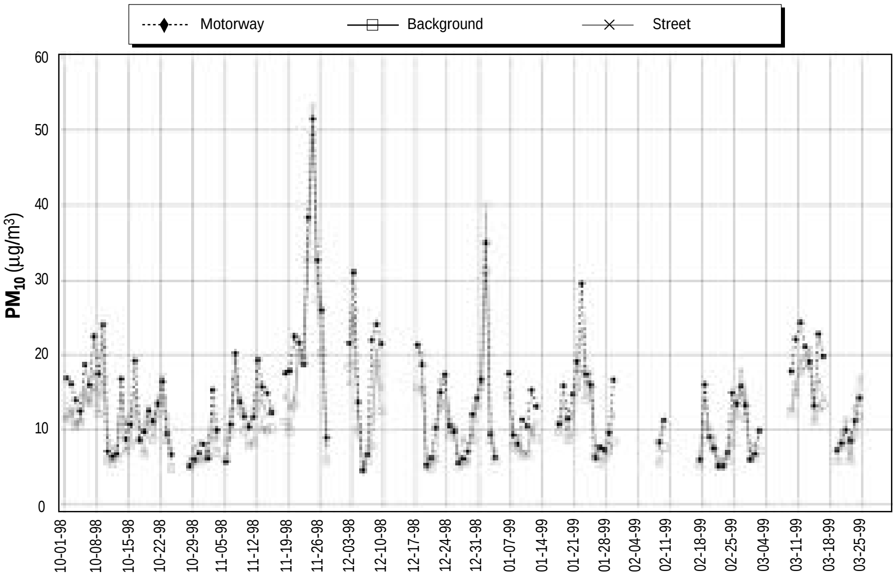

Figure 2. Daily $PM_{1.0}$ concentrations (µg/m^3) at motorway, street, and background sites, against date.

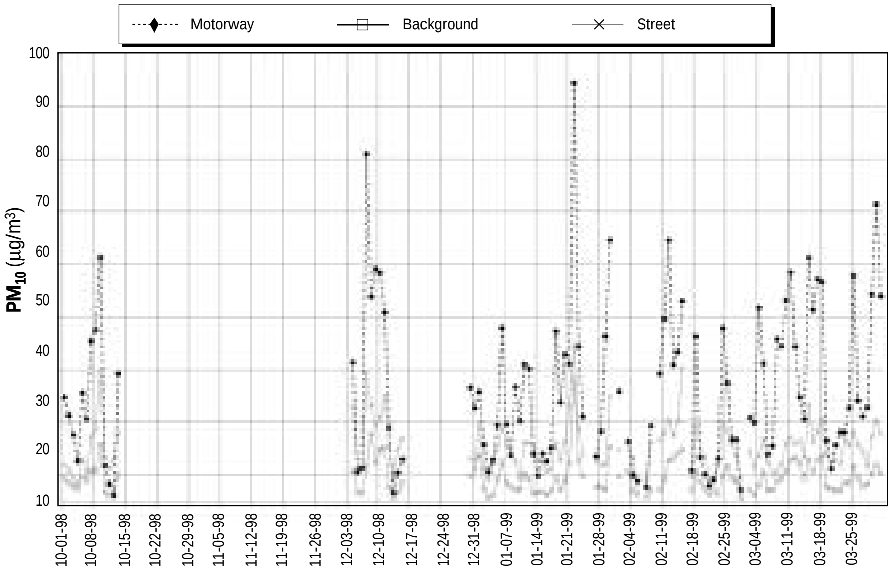

Figure 3. Daily black smoke concentrations (µg/m^3) at motorway, street, and background sites,against date.

Table 3. Spearman's correlation coefficient among 24-hr average concentrations of same component on different sites.

Type	Street	Motorway
PM_{10}		
Background	0.93*	0.75*
Street		0.75*
$PM_{1.0}$		
Background	0.96*	0.92*
Street		0.92*
Black smoke		
Background	0.83*	0.85*
Street		0.76*

*$p < 0.01$

Table 4. Spearman's correlation coefficients among 24-hr average concentrations of different components on same sites.

Site	$PM_{1.0}$	Black smoke
Background		
PM_{10}	0.75*	0.64*
$PM_{1.0}$		0.86*
Street		
PM_{10}	0.74*	0.65*
$PM_{1.0}$		0.68*
Motorway		
PM_{10}	0.90*	0.84*
$PM_{1.0}$		0.89*

*$p < 0.01$.

Adverse health effects such as cancer and lung function decrements have been shown in animal studies and occupational settings for diesel exhaust (*17*). Studies of health effects in general populations related to traffic emissions are less frequent and are often limited by lack of exposure measurements (*18*). Health effects associated with black smoke range from increased respiratory symptoms (*15*) and reduced lung function (*19*) to mortality (*6*). The European Union and the United States have formulated threshold values for particulate matter in ambient air in terms of mass concentrations of PM_{10} and/or $PM_{2.5}$ to protect public health. The results in this paper indicate that given the high correlation over time, mass concentrations of particulate matter can be used to estimate day-to-day variations in exposure to particles generated by traffic, such as black smoke. However, due to the large difference in median levels, mass concentrations may underestimate the differences in long-term exposure to particles emitted by traffic.

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