PERGAMON

Atmospheric Environment 35 (2001) 3875–3884

ATMOSPHERIC ENVIRONMENT

www.elsevier.com/locate/atmosenv

Assessment of exposure to traffic related air pollution of children attending schools near motorways

Nicole A.H. Janssen, Patricia H.N. van Vliet, Francée Aarts, Hendrik Harssema, Bert Brunekreef*

Environmental and Occuptational Health Group, Utrecht University, P.O. Box 80 176, 3508 TD Utrecht, Netherlands

Received 19 April 2000; received in revised form 2 February 2001; accepted 10 February 2001

Abstract

To assess exposure to air pollution from traffic of children attending schools near motorways, traffic related air pollution ($PM_{2.5}$, NO_2 and benzene) was measured in and outside 24 schools located within 400 m of motorways in the Netherlands. Reflectance of $PM_{2.5}$ filters was measured as a proxy for elemental carbon (EC). The relationship between this proxy and measurements of EC was studied in a sub-sample and a high correlation was established. In both indoor and outdoor air, concentrations of $PM_{2.5}$ and "soot" significantly increased with increasing truck traffic density and significantly decreased with increasing distance. Indoor NO_2 concentrations significantly increased with increasing car traffic. The percentage of time that the school was downwind of the motorway during the measurements was significantly associated with "soot" and NO_2, but not with $PM_{2.5}$ and benzene. Estimated yearly averaged concentrations, calculated after standardising for differences in the background concentrations during the measurements, showed an about 2.5 fold range in "soot", benzene (indoors and outdoors) and NO_2 (indoors) concentrations. For $PM_{2.5}$ (indoors and outdoors) and NO_2 outdoors the range was smaller (1.4–1.7). Standardised concentrations were highly correlated with the results of two other approaches that were used to order the exposures at the schools. This study has shown that concentrations of air pollutants in and outside schools near motorways are significantly associated with distance, traffic density and composition, and percentage of time downwind. These variables can therefore be used to assess exposure to traffic related air pollution of subjects living near motorways. Furthermore, the yearly averaged concentrations of $PM_{2.5}$, "soot", NO_2 and benzene can be used as a more direct measure of long-term exposure in epidemiological studies of the children attending the 24 schools.

Keywords: Traffic pollution; Particles; Soot; Nitrogen dioxide; Benzene; Roads

1. Introduction

Motorised traffic is a major source of air pollutants such as nitrogen dioxide (NO_2) and suspended particulate matter (SPM). Several recent studies suggest an association between living near busy roads and respiratory health (Weiland et al., 1994; Nitta et al., 1993; Ciccone et al., 1998). Most of these studies have used (self-reported) traffic density on the street of residence and/or distance of the home address to busy roads as measures of exposure. Few studies have incorporated actual measurements of traffic related air pollutants. Krämer et al. (2000) measured personal and outdoor NO_2 concentrations of children living near major roads in two urban and one suburban area. Outdoor concentrations were correlated with a traffic index based on the traffic density at the home address ($R = 0.70$). Outdoor NO_2 concentrations at the front of the children's homes were associated with atopy and allergic

*Corresponding author. Fax: +31-30-253-5077.
E-mail address: b.brunekreef@vet.uu.nl (B. Brunekreef).

1352-2310/01/$ - see front matter
PII: S1352-2310(01)00144-3

symptoms. No such associations were found for personal NO_2. Ciccone et al. (1998) included some validation of the exposure indicator used by simultaneously measuring outdoor NO_2 concentrations in 85 locations in one of the 10 study areas. NO_2 concentrations were on average about 10 μg m^{-3} (23%) higher in streets where lorries were reported often to pass than in streets where lorries were reported not or seldom to pass. Nitta et al. (1993) included measurements of NO_x at 3 or 4 different distances from the road in 7 locations. The average concentration at the roadside (0 m) was 14–46% higher than the concentration at 150 m. Mean SPM concentrations, measured at one of the locations, declined from 77 μg m^{-3} at 0 m to 64 μg m^{-3} at 150 m. Nakai et al. (1995) measured personal, indoor and outdoor concentrations of NO_2 of subjects selected from participants of a health effects study which was conducted some years earlier. Measurements were conducted 10 times during several seasons. Mean outdoor concentrations were always higher in the zone closest to the road and lowest in the reference zone. The overall average NO_2 concentration was 81 μg m^{-3} in zone A (<20 m), 67 μg m^{-3} in zone B (20–150 m) and 39 μg m^{-3} in the reference zone. Mean personal and indoor NO_2 concentrations were generally higher in zone A during non-heating seasons. During the heating season this was only the case for residents who did not have an unvented heater.

In 1995, respiratory health of children living and attending schools within 1000 m from 6 major motor ways in the Netherlands was studied (Brunekreef et al., 1997; van Vliet et al., 1997). In addition to traffic counts for cars and trucks and distance of the homes to the motorway as exposure variables, air pollution in the children's schools was measured. Black smoke concentrations were significantly correlated with truck traffic density and percentage of time downwind. Classroom NO_2 concentrations were significantly correlated with car and total traffic density, percentage of time downwind and distance of the school to the motorway (Roorda-Knape et al., 1998). Truck traffic density and classroom black smoke concentrations were significantly associated with chronic respiratory symptoms and lung function. In 1997/1998 we conducted a similar, more extensive, study, involving children from 24 schools situated within 400 m of 22 different motorway stretches. $PM_{2.5}$, "soot", NO_2 and benzene were measured in and outside all 24 schools. This paper describes the relation between air pollution measurements on the one hand and traffic density of the motorway, distance to the motorway and percentage of time downwind during the measurements on the other hand. Furthermore, yearly averaged air pollution concentrations in and outside the schools were estimated, to be used as measures of exposure in the epidemiological study.

2. Methods

2.1. Sampling strategy

Traffic related air pollution was measured at 24 schools, located within 400 m of motorways in the Netherlands. The schools were situated along 22 different motorway stretches. Three schools, which were also involved in a study on respiratory health of children living near Schiphol airport, were situated along the same motorway stretch (2 on the Northeast side and 1 on the Southwest side of the road). The other 21 schools were all situated along different motorways stretches. In the selection of the sites, a maximum variation in traffic densities and a minimal correlation between car and truck traffic was aimed at. In and outside each school, $PM_{2.5}$, NO_2 and benzene was measured. Reflectance of $PM_{2.5}$ filters was measured as a marker for elemental carbon (EC), a major part of diesel soot (Wolff, 1985). Weekly averaged measurements were conducted. For $PM_{2.5}$ and benzene, indoor sampling was done during school-hours only (Wednesday 9.00–13.00; 9.00–15.00 on all other weekdays). Outdoor sampling for these components was done on all days of the week during 15 min out of every hour, to avoid overloading of the samples. NO_2 sampling was continuous in both indoor and outdoor air. Each school was measured 5 to 10 times, spaced over 16 weeks in the period April 1997–May1998. Every measuring week, on average 12 schools (range 4–18) were measured simultaneously. Indoor samples were taken in one of the classrooms, at 1.5 m height and away from the door, window or blackboard. Outdoor sampling was mostly done on the roof of the school or in a patio. In 2 cases, outdoor measurements were conducted in the garden of a neighbouring house because no suitable monitoring sites were available on the grounds of the schools.

2.2. Sampling methods

$PM_{2.5}$ was measured using Harvard impactors (Marple et al., 1987) at 10 l min^{-1}. Andersen 2 μm pore size Teflon filters were used. Flows were measured at the beginning and end of each sampling period with calibrated rotameters and elapsed time indicators were used to calculate the sampled volumes. Filters were weighed using a Mettler AT261 analytical balance with 10 μg reading, after equilibrating at about 20°C and 45% relative humidity for 24 h in a desiccator. Reflectance of $PM_{2.5}$ filters were measured using an EEL43 reflectometer and transformed into an absorption coefficient according to ISO 9835 (ISO, 1993). Absorption coefficients were converted into black smoke ("soot") concentrations using a regression equation of the relation between absorption of PM_{10} filters and black smoke concentrations measured simultaneously at

the same site, reported by Roorda-Knape et al. (1998) (Black smoke ($\mu g\ m^{-3}$) = $-3.663+9.897 \times$ Absorption; $R^2 = 0.94$; n = 40). Co-located measurements of both $PM_{2.5}$ and PM_{10} in outdoor air conducted by us showed that absorption coefficients of $PM_{2.5}$ filters are highly correlated with and only slightly lower than absorption coefficients of PM_{10} filters ($ABS_{PM2.5} = 0.03 + 0.93 \times ABS_{PM10}$; $R^2 = 0.99$; n = 26). Fischer et al. (2000) also documented little difference between absorption of PM_{10} and $PM_{2.5}$ filters. The mean coefficient of variation (CV) of 14 indoor duplicates was 7% (S.D. 5; range 0–15%) for $PM_{2.5}$ and 3% (S.D. 1; range 0–6%) for absorption. For outdoor duplicates ($n = 26$) mean CV values were 5% (S.D. 6; range 0–28%) for $PM_{2.5}$ and 1% (S.D. 1; range 0–5%) for absorption.

Benzene was sampled at a flow rate of about 700 ml min^{-1} through SKC charcoal tubes. Flows were measured at the beginning and end of each sampling period with calibrated rotameters and elapsed time indicators were used to calculate the sampled volumes. Valve boxes were used to avoid passive loading of the tubes during non-sampling times. Samples were analysed on a GLC Hewlett Packard type 5890 gas chromatograph, after desorption with 1 ml carbon disulphide (CS_2). The mean CV was 13% (S.D. = 9; range 1–29%) for 13 indoor duplicates and 7% (S.D. = 7; range 0–24%) for 27 outdoor duplicates.

NO_2 was measured using diffusion tubes described by Palmes et al. (1976). These tubes consist of a cylindrical tube, which at one end is closed with a cap containing a metal grid coated with tri-ethanol amine. In the first 8 months of the fieldwork, somewhat transparent caps were used instead of the red caps we had used before, as these were no longer available from the distributor. Outdoor NO_2 measurements conducted with the transparent caps appeared to give too low NO_2 concentrations. Laboratory experiments showed that after exposing some highly exposed tubes with ultraviolet light, hardly any NO_2 was measured anymore. No such effect was observed when red caps instead of transparent caps were used. From December 1997 onward, therefore, red caps were obtained again and used. Field comparison measurements conducted throughout the remainder of the fieldwork showed that in outdoor air significantly lower concentrations (up to > 100% lower) were measured with the transparent caps compared to the red caps, especially in the spring. No such difference was found for indoor air. We therefore decided to exclude all outdoor measurements conducted with transparent caps from the data analysis. Indoor measurements were not excluded. Chemical analysis was done by colorimetry using Saltzman reagent. All measurements were done in duplicate. Two observations were excluded because of bad repeatability (CV > 30%). The mean CV of the remaining duplicates was 6% (S.D. 5; range 0–27%) for both indoor and outdoor measurements.

To establish the correlation between reflectance of $PM_{2.5}$ filters and direct EC measurements, EC was measured in a subset of 47 outdoor and 32 indoor samples. For the EC measurements duplicate sample collection was conducted using Harvard $PM_{2.5}$ impactors and Schleicher and Schuell QF 20 quartz filters. Samples were analysed the AMU TUV GMBH laboratory in Munchen, Germany, according to VDI 2465 standard (VDI, 1995). The mean CV of four outdoor EC duplicates was 10% (S.D. 5; range 3–13%).

2.3. Relation between air pollution concentrations, traffic density, distance and percentage of time downwind

Traffic counts were obtained from the Ministry of Public Works, which routinely collects counts for all motorway stretches in the Netherlands using induction loops. This method distinguishes into vehicles smaller than and larger than 5.1 m, which we classified as cars and trucks respectively. Weekday counts for 1997 were used. Distances of the measuring points to the motorways were measured using 1 : 1000 or 1 : 1500 scale maps. Data on wind direction per hour were obtained from measuring site Zestienhoven (Rotterdam Airport) of the Royal Dutch Meteorological Institute. For each measurement, the percentage of time that the school was downwind during the measurement was calculated. A school was considered downwind if the wind was within 60° from perpendicular to the road in the direction of the school. The relationship between air pollution concentrations, traffic density, distance and percentage of time downwind was assessed using multiple regression analysis. The logarithm of distance was used because from general dispersion models an exponential decay in the contribution from the road with distance can be expected. All measurements of a particular pollutant were included in the same model. Regression results were adjusted for measurement period by including 15 dummy variables (for 16 measurement periods) in the model. The SAS procedure "Proc Mixed" was used to adjust regression results for correlations between repeated measurements of the same school. A random intercept model was used.

2.4. Estimation of yearly averaged air pollution concentrations

For logistical reasons it was not possible to measure all schools simultaneously. Instead, on average 12 schools at a time were measured during 16 different measurement weeks, resulting in 5 to 10 weekly averaged concentrations per school (see sampling strategy). Because of the temporal variation in air pollution concentrations, average background concentrations

during the measurements differed from school to school. For example, the average black smoke background concentration during the measurements varied between 11 and 16 μg m^{-3} for the different schools. Air pollution concentrations were therefore standardised for the background concentrations during the measurements according to

$$C_{\text{standardised},t} = (C_{i,t}/C_{\text{background},t}) \times C_{\text{background,year}}.$$

Where $C_{\text{standardised},t}$ is the standardised concentration during week t; $C_{i,t}$ is the measured concentration during week t; $C_{\text{background},t}$ is the average background concentration during week t; $C_{\text{background,year}}$ is the yearly averaged background concentration. Background concentrations were obtained from the Dutch National Air Quality Monitoring Network, which is operated by the Dutch National Institute of Public Health and the Environment (RIVM). For $PM_{2.5}$ and "soot", PM_{10} and Black smoke concentrations were used from 4 regional background network stations spread over the study area. For benzene, data from only one background station was available. For NO_2, information from 9 background stations was available. Since no background $PM_{2.5}$ concentrations were available, PM_{10} data were used to standardise $PM_{2.5}$ concentrations. Before standardising, $PM_{2.5}$ concentrations were converted into PM_{10} concentrations using a regression equation of the relation between $PM_{2.5}$ and PM_{10} obtained from 26 co-located measurements in outdoor air ($PM_{2.5} = -6.2 + 0.77 \times PM_{10}$; $R^2 = 0.93$). After standardising, PM_{10} concentrations were converted into $PM_{2.5}$ again. This procedure was necessary because of the significant intercept in the relation between $PM_{2.5}$ and PM_{10}.

In addition to the calculation of standardised concentrations, 2 other approaches were used. First, instead of the ratio $C_{i,t}/C_{\text{background},t}$, the difference between measured and background ($C_{i,t} - C_{\text{background},t}$) concentrations was calculated and average differences were calculated for each school. Second, differences between the schools were estimated using a regression model: concentration $= \alpha + \beta * \text{school} + \delta * \text{period}$ in which 'school' and 'period' were included as 23 and 15 dummy variables respectively. Results of the last two approaches were correlated with the average standardised concentrations to evaluate if the different approaches reflect the same order of exposures.

3. Results

Characteristics of the various motorways (Table 1) showed a 4–5-fold range in traffic densities. The correlation between car and truck traffic was low (Fig. 1, $R = 0.31$). Distance of the school to the motorway was moderately correlated with truck traffic ($R = 0.52$), but not with total or car traffic.

Indoor concentrations of "soot" and benzene exceeded outdoor concentrations (Table 2). This can be caused by the difference in the sampling times: indoor measurements were conducted during school hours only, whereas outdoor sampling was done on all days of the week, thus including night-times and weekends during which traffic flows and therefore emissions are lower than during school hours. NO_2 concentrations, which were measured continuously in both indoor and outdoor

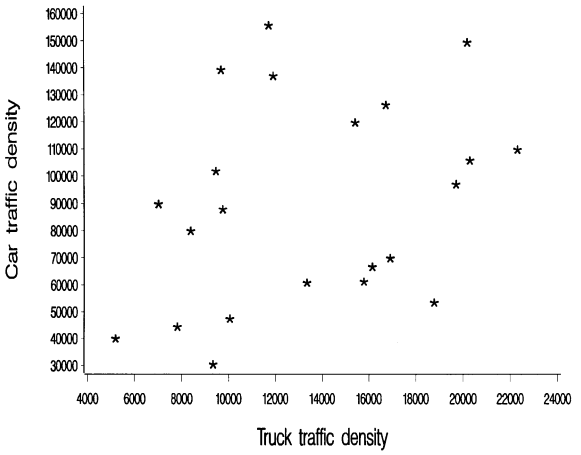

Fig. 1. Relation between car and truck traffic density (vehicles per weekday) on the 22 motorway stretches ($R = 0.31$).

Table 1
Characteristics of the study locations ($n = 24$)

	Mean	S.D.[a]	Median	Range
Truck traffic density (vehicles/weekday)	13 146	4880	11 814	5190–22 326
Car traffic density (vehicles/weekday)	89 544	35 843	87 843	30 399–155 656
Total traffic density (vehicles/weekday)	102 690	37 620	97 603	39 737–169 637
Distance outdoor measurement site (m)	206	108	210	32–377
Distance indoor measurement site (m)	208	115	227	32–390
Yearly average % of time downwind	38	10	38	27–49

[a] Standard deviation.

Table 2
Distribution of air pollution concentrations (μg m^{-3})

	Indoor					Outdoor				
	N	Mean	Median	S.D.[a]	Range	*N*	Mean	Median	S.D.	Range
PM2.5	178	23.0	23.2	6.3	7.7–52.8	181	24.8	23.4	11.6	5.2–60.8
"Soot"	177	14.7	15.0	5.6	1.9–35.4	179	12.2	12.6	5.0	0.7–24.5
NO_2	192	19.1	17.5	8.1	2.8–44.7	84	39.2	38.5	15.9	10.7–76.6
Benzene	160	3.2	2.9	1.7	0.6–8.1	177	2.2	2.1	1.1	0.3–5.0

[a] Standard deviation.

Table 3
Relation between air pollution concentrations, traffic density, distance and percentage of time downwind (adjusted for measurement week; trucks per 10.000; cars per 50.000)

	n	Cars		Trucks		Log(distance (m))		% downwind	
		Estimate[a]	S.E.[b]	Estimate	S.E.	Estimate	S.E.	Estimate	S.E.
Indoor									
PM25	178	0.09	0.95	3.31*	1.52	−2.19*	0.95	1.48	1.15
"Soot"	177	1.20*	0.54	1.91*	0.87	−2.82**	0.55	3.61**	0.64
NO_2	192	4.01*	1.51	−2.95	2.42	−0.84	1.53	3.31**	1.22
Benzene	160	0.36	0.24	0.66	0.38	−0.14	0.24	0.68*	0.28
Outdoor									
PM25	181	−1.06†	0.51	4.01**	0.82	−1.89**	0.53	−0.45	0.84
"Soot"	179	−0.06	0.40	2.23**	0.63	−2.24**	0.42	2.49**	0.44
NO_2	84	2.46	1.49	2.41	2.33	−2.40	1.47	5.84*	2.26
Benzene	178	0.16	0.10	0.35*	0.16	−0.34**	0.11	−0.10	0.13

[a] Parameter estimate.
[b] Standard error.
$^{*}p<0.05$.
$^{**}p<0.01$.
$^{\dagger}p<0.10$.

air, were about 50% lower in indoor compared to outdoor air. For benzene, however, the indoor concentration was almost 8 times higher than the outdoor concentration on 2 occasions. One was caused by an extremely high indoor concentration of 14 μg m^{-3}; the other was due to a low outdoor concentration of 0.6 μg m^{-3} in a period that outdoor concentrations that were measured simultaneously at 14 other schools ranged from 2.1 to 3.3 μg m^{-3}. These 2 concentrations also showed an extremely high (indoor) and low (outdoor) residual in the results of the regression analyses presented in Table 3 and were therefore excluded from the analysis (already excluded in Table 2).

Absorption coefficients were highly correlated with EC concentrations (Fig. 2). The estimated regression equations are: Absorption ($m^{-1} \times 10^{-5}$) = 0.46 + 0.34 ×EC (μg m^{-3}) ($R = 0.92; n = 47$) in outdoor air, and absorption ($m^{-1} \times 10^{-5}$) = 0.45 + 0.42 × EC (μg m^{-3}) ($R = 0.85; n = 32$) in indoor air. After excluding one outlier in indoor air (circled value) the correlation coefficient in indoor air increased to 0.93. Fig. 3 shows that the relation between reflectances (%) of Teflon

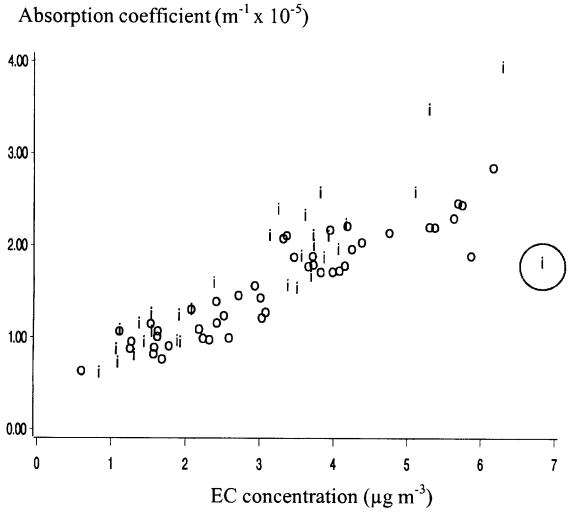

Fig. 2. Relation between absorption coefficients and EC concentrations (O = outdoor measurement; i = indoor measurement).

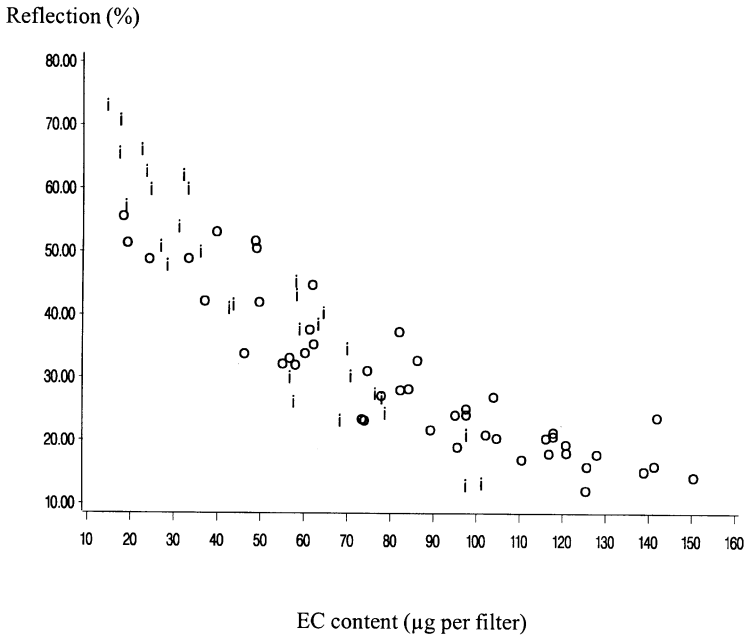

Fig. 3. Relation between reflectance (%) and EC content (μg per filter) (O = outdoor measurement; i = indoor measurement).

filters and the amount of EC (μg) collected on the co-located quartz filter, corrected for the difference in the sampled volumes of the Teflon and quartz filter, is not completely linear. From about 110 μg EC onward the reflectance hardly decreases anymore (a filter can not get blacker than black). This suggests that absorption coefficients calculated from filters with very low reflectances (<15%) are unreliable. This mainly occurred when the outdoor pump accidentally had run continuously instead of 15 min out of every hour. Since this could have resulted in a serious underestimation of the absorption coefficient, these samples were excluded from the analysis.

In both indoor and outdoor air, $PM_{2.5}$ and "soot" concentrations significantly increased with increasing truck traffic density and significantly decreased with increasing distance (Table 3). NO_2 concentrations significantly increased with increasing car traffic density in indoor air. The lack of significant associations in outdoor air can be caused by the smaller sample size ($n = 84$). Including total traffic density instead of separate variables for cars and trucks in the NO_2 model resulted in a significant regression coefficient for total traffic in both indoor and outdoor air (2.9 and 3.0 μg m^{-3} per 50,000 vehicles respectively). The coefficient for the logarithm of distance in this model is −1.9 (s.e. 1.3) for both indoor and outdoor air. Outdoor benzene concentrations are significantly associated with truck traffic density and distance, whereas indoor benzene concentrations are only significantly associated with percentage of time downwind. The significant association with truck traffic, however, is partly caused by one school, along a highway with a truck traffic density of 19,724 vehicles per day, that was also situated close to a busy local roundabout and gasoline station. When this school is excluded from the analysis a significant association is found for both car and truck traffic. The coefficient for trucks decreases from 0.35 to 0.26 μg m^{-3} per 10,000 trucks (s.e. 0.12), whereas for cars only the standard error decreases to 0.08. Exclusion of this school had little influence on the results for indoor benzene concentrations. Percentage of time downwind is significantly associated with "soot", NO_2 and benzene indoors, but not with $PM_{2.5}$ and benzene outdoors. The average percentage of time downwind during the measurements ranged from 25 to 65% for the different schools. The difference between the percentage of time downwind during the measurements and the yearly average percentage ranged from −21 to 21% (average 0%). The associations of $PM_{2.5}$ and "soot" with distance imply that, at an average traffic density and % of time downwind, $PM_{2.5}$ and "soot" concentrations at 50 m are about 20 and 50%, respectively, higher than at 400 m.

Table 4 shows that for "soot" and benzene in both indoor and outdoor air and for NO_2 in indoor air there is an about 2.5 fold range in estimated yearly averaged concentrations at the schools. For $PM_{2.5}$ (indoor + outdoor) and NO_2 outdoors the range is smaller (1.4–1.7). Average standardised concentrations were highly correlated with the 2 other approaches that were used to order the exposures at the schools: Pearson's R ranged

Table 4
Distribution of standardized average air pollution in and outside the schools ($n = 24$; $\mu g\ m^{-3}$)

	Indoor					Outdoor				
	Mean	Median	S.D.[a]	Min	Max	Mean	Median	S.D.	Min	Max
PM2.5	20.3	20.1	2.7	16.6	27.1	20.5	20.4	2.2	17.3	24.4
"Soot"	12.7	11.8	3.0	8.7	21.5	10.3	10.5	2.1	6.2	15.5
NO_2	18.8	17.8	5.3	11.9	30.4	34.8	34.0	5.2	26.8	44.4
Benzene	2.9	2.5	0.7	2.0	4.8	1.9	1.8	0.5	1.2	3.1

[a] Standard deviation.

from 0.93 to 0.97 for the correlation between standardised concentrations and school specific averages of the difference between the concentrations at the schools and corresponding background concentrations. The correlation coefficients with the school specific β's (obtained from the regression model concentration= $\alpha + \beta *$ school $+ \delta *$ measurement period) were slightly lower ($R = 0.93$–0.96 for indoor air and 0.81–0.91 for outdoor air). Average understandardised concentrations were generally less well correlated with the results of any of the 3 approaches.

4. Discussion

4.1. Relation between air pollution concentrations, traffic density, distance and percentage of time downwind

$PM_{2.5}$ and "soot" concentrations in both indoor and outdoor air significantly increased with increasing truck traffic density. A significant association with car traffic was only found for "soot" in indoor air. This is in line with the higher particle emissions from diesel vehicles compared to gasoline-powered automobiles observed in emission studies (Hildemann et al., 1991). Kinney et al. (2000) found a 4.1 times higher EC concentration on the sidewalk of a street with 18,375 cars and a total of 2467 trucks and buses per 8 h compared to a background location. For a street with an about 20% lower car traffic count (14,229 per 8 h) but with much less trucks and buses (927 per 8 h) the street/background ratio was only 1.5 Horvath et al. (1988) observed a strong correlation ($R = 0.93$) between estimated concentrations of diesel particles and the number of diesel vehicles measured at 7 sites in Vienna. Estimated concentrations of diesel particles ranged from $10.3\ \mu g\ m^{-3}$ in a residential low traffic area to $22.4\ \mu g\ m^{-3}$ at 37 m distance from a motorway with heavy traffic (> 850 diesel vehicles h^{-1}). In our previous study on air pollution along major motorways (Roorda-Knape et al., 1998), concentrations of "soot" in classrooms were also significantly associated with truck traffic density. In contrast to the present study, no association was found between indoor "soot" and distance to the motorway. In outdoor air, however, where black smoke concentrations were measured at varying distances from the same motorway, black smoke concentrations significantly decreased with increasing distance. No such gradient was found for $PM_{2.5}$ or PM_{10}. Two other studies documented 10–40% higher $PM_{2.5}$ and/or PM_{10} concentrations 15–40 m downwind of motorways compared to upwind concentrations or concentrations at 3 75 m distance (Lamoree and Turner, 1999; Hitchins et al., 2000), which is in line with the estimated ratio between concentrations at 50 and 400 m of 1.2 that was found in our study.

The association with distance was stronger for "soot" than for $PM_{2.5}$. Furthermore, "soot" was significantly associated with percentage of time downwind, whereas no such association was found for $PM_{2.5}$. This is in line with several other studies that have documented a stronger influence of traffic on "soot" or EC compared to $PM_{2.5}$ or PM_{10} concentrations, with street/backging from 1.8 to 4.1 for "soot" or EC and from 0.9–1.3 for $PM_{2.5}$ or PM_{10} (Janssen et al., 1997; Fischer et al., 2000; Kinney et al., 2000).

"Soot" and NO_2 concentrations were significantly associated with the percentage of time that the school was downwind of the motorway during the measurements. In our previous study (Roorda-Knape et al., 1998), black smoke and NO_2 concentrations decreased more strongly with distance from the road in periods with a high percentage of time downwind ($> 33\%$) compared to periods with a low percentage of time downwind ($< 33\%$). Hitchins et al. (2000) found a steeper decline in particle numbers with distance to a motorway for conditions that the wind blew directly from the road compared to when the wind blew parallel to or towards the road. Morawska et al. (1999) found an increase in particle numbers along a motorway that coincided with the morning peak hour, during which the wind was directed from the road to the sampling points. No such increase was observed during the afternoon peak hour, when the wind no longer came from the road.

NO_2 concentrations were significantly associated with car traffic (indoors only), total traffic and percentage of time downwind. This is in line with the significant

correlations between indoor NO_2 concentrations in classrooms and car traffic, total traffic and percentage of time downwind that were found in our previous study (Roorda-Knape et al., 1998). Kuhler et al. (1988) found a close relation between the diurnal variation of total traffic and the difference between upwind and downwind NO_x concentrations measured at 100 m distance from a motorway. In the present study, no significant association between NO_2 concentrations and distance to the motorway was found. Roorda-Knape et al. (1998) found a significant correlation between indoor NO_2 concentrations in classrooms and distance of the school to the motorway. Furthermore, outdoor NO_2 measurements at various distances from the same motorway showed a clear gradient. Two other studies in open terrain downwind of a motorway also found a decline in NO_2 concentrations with distance (Rodes and Holland, 1981; Kuhler et al., 1988). Two Japanese studies also documented associations between NO_2 concentrations and distance to major roads (Nitta et al., 1993; Nakai et al., 1995). Possibly, the relationship between NO_2 and distance in our study was obscured by the influence of local traffic within the city districts. Local traffic will have less influence on the associations with distance for the more to truck traffic related components $PM_{2.5}$ and "soot", since the number of trucks within a city district is probably negligible compared to the number of trucks on the motorway. In a more extensive NO_2 monitoring study conducted in 3 of the 24 schools, Rijnders et al. (2001) found a significant decrease of home outdoor and personal NO_2 concentrations with distance of 1.3–1.4 $\mu g\,m^{-3}$ per log(m), which is smaller than the non-significant value of $-1.9\,\mu g\,m^{-3}$ per log(m) found in the present study. The observed decrease with distance ($<3\,\mu g\,m^{-3}$ when expressed as the difference between 50 and 400 m distance) was small compared to the observed differences between NO_2 concentrations along the busiest highway (169,637 vehicles per 24 h) and least busy highway (45,129 vehicles per 24 h) of about 8 (personal) to 10 (outdoor) $\mu g/m^3$. This suggests that traffic intensity on a freeway is a more important determinant of NO_2 exposure than distance of home or school to the freeway (Rijnders et al., 2001).

Outdoor benzene concentrations were significantly associated with truck traffic intensity and, after excluding a school along a highway with a high truck traffic intensity that was also situated close to a gasoline station, also with car traffic intensity. Although the benzene emission of diesel vehicles is generally considered to be low, benzene has been found to be emitted both by gasoline and by diesel vehicles (Muzyka et al., 1998; Westerholm and Egebäck, 1994; Duffy et al., 1998). In a study in Sweden, Westerholm and Egebäck (1994) found an emission factor of 13 mg km^{-1} for a heavy-duty diesel truck using commercial, standard diesel fuel. The emission factors for a gasoline fuelled car without a catalyst was about 40 mg km^{-1}. For catalysed equipped cars, however, a much lower emission factor of 3–8 mg km^{-1} was found, depending on the test speed and type of catalyst. As benzene in outdoor air is also influenced by fugitive emissions, the possibility remains, however, that some of these associations were distorted by unrecognised fugitive emissions of benzene occurring near our monitoring stations.

Benzene concentrations indoors were not significantly associated with traffic intensity. Furthermore, benzene concentrations significantly declined with distance in outdoor air, whereas no such association was found in indoor air. Roorda-Knape et al. (1998) did not found a clear gradient between outdoor benzene concentrations and distance from the motorway. Significantly elevated benzene concentrations were only found at very close distance to the road (15 m). Clark et al. (1984) found about 60% higher benzene concentrations at 15 m distance from a motorway near London compared to a rural site in a park. Two Dutch studies reported about 2 times higher benzene concentrations at the facade or street-side balcony of homes along busy streets compared to homes along quiet streets (Bloemen et al., 1993; Fischer et al., 2000). In one of these studies (Bloemen et al., 1993), measurements were also conducted at the backsides of the same homes and no significant differences were found. These results suggest that elevated benzene concentrations, caused by traffic, mainly occur at short distances from the road. Also, as was already mentioned for NO_2 in the previous paragraph, the influence of local traffic could have blurred the effect of motorway emission on benzene concentrations.

4.2. Estimation of yearly averaged air pollution concentrations

The air pollution measurements were also aimed to assess long-term exposure to traffic related air pollution in an epidemiological study on respiratory health of the children attending the 24 schools. Since background concentrations during the measurements varied from school to school, measured air pollution concentrations were standardised by multiplication with the ratio of the yearly average background concentrations to the background concentration during the measurement. Other studies have used similar approaches (Schindler et al., 1998; Hitchins et al., 2000). Average standardised concentrations were highly correlated with the results of two alternative approaches that were used to classify the exposures at the different schools and can therefore be considered robust measures of the long-term exposures at the schools.

Almost all schools were situated in different city districts. We did not study the spatial variation within

the city districts. In the Netherlands, however, children usually go to primary schools that are relatively close to their homes. Furthermore, because traffic flows are highest during daytime, an important part of exposure to air pollution from traffic will occur when the children are at school. We therefore hypothesised that exposure to traffic related air pollution varies less between children from the same school than between children from schools along different motorways with varying traffic densities. To test this hypothesis, Rijnders et al. (2001) measured personal and outdoor NO_2 concentrations of about 110 children from 3 of the 24 participating schools. These 3 schools were located along motorways with varying total traffic intensities (169,637, 126,115 and 45,129 vehicles/24 h, respectively). Personal NO_2 concentrations as well as outdoor concentrations at the backsides of the children's homes significantly differed between the 3 schools, with the highest average concentrations occurring along the busiest motorway and the lowest average concentrations along the least busy motorway. The average standardised concentrations at the schools are therefore relevant measures of exposure to traffic related air pollution that can be used in the epidemiological part of the study. In addition, to further increase the strength of the exposure variables, we will restrict the epidemiological analysis to children living within 1000 m of the motorway.

For "soot" and benzene in both indoor and outdoor air and for NO_2 in indoor air there was an about 2.5 fold range in average standardised concentrations between the 24 schools. For $PM_{2.5}$ (indoor + outdoor) and NO_2 outdoors the range is smaller (1.4–1.7). In the US Six City Study, an about 2.5 fold range in long-term average particle concentrations was significantly associated with mortality (Dockery et al., 1993). Other studies with similar contrasts in long-term average air pollution concentrations have demonstrated both significant as non-significant effects on respiratory health of children (Dockery et al., 1989; Raizenne et al., 1996). The contrast in concentrations in our study should therefore be sufficient to study potential health effects of long-term exposure to air pollution on the children attending the 24 schools.

The percentage of time that the schools were downwind during the measurement did not necessarily equal the yearly average percentage of time downwind. For the individual schools the difference between these percentages ranged from −21 to 21% (average 0%). This is not corrected for in the calculation of the yearly averaged concentrations. Although significant associations were found between concentrations of "soot" and NO_2 and the percentage of time downwind, a 20% higher or lower percentage of time downwind only results in a 3–5% higher or lower concentration. These differences therefore cannot have substantially influenced the yearly average concentrations.

4.3. Relation between absorption coefficients and elemental carbon concentrations

Absorption coefficients, derived from the reflectance of $PM_{2.5}$ filters, were highly correlated with simultaneously measured EC concentrations. In a study in New York, the absorption coefficient of $PM_{2.5}$ Teflon filters, measured on the sidewalk of 3 traffic impacted and 1 background site for 5 days, was also highly correlated ($R = 0.95$) with simultaneously measured EC concentrations (Kinney et al., 2000). In a study in Berlin, outdoor black smoke concentrations at a traffic-impacted site were highly correlated with EC concentrations ($R = 0.98$) (Ulrich and Israël, 1992). Since EC is a major part of diesel soot, these results show that measurements of the reflectance of PM filters can be used as a surrogate for exposure to diesel exhaust particles.

5. Conclusions

This study has shown that air pollution concentrations in and outside 24 schools near different motorway segments are significantly associated with distance of the school to the motorway, traffic density and percentage of time downwind. These variables can therefore be used to assess exposures to traffic related air pollution of subjects living near motorways. Furthermore, the yearly averaged concentrations of PM2.5, "soot", NO_2 and benzene can be used as a more direct measure of long-term exposure in epidemiological studies of the children attending the 24 schools.

References

Bloemen, H.J.T., Balvers, T.T.M., van Scheindelen, H.J., Lebret, E., 1993. Benzene study South-Kennermerland. National Institute of Public Health and Environmental Protection Report, Bilthoven (in Dutch).

Brunekreef, B., Janssen, N.A.H., de Hartog, J., Harssema, H., Knape, M., van Vliet, P., 1997. Air pollution from traffic and lung function in children living near motorways. Epidemiology 8, 298–303.

Ciccone, G., Forastiere, F., Agabiti, N., Biggeri, A., Bisanti, L., Chellini, E., Corbo, G., Dell'Orco, V., Dalmasso, P., Volante, T.F., Galassi, C., Piffer, S., Renzoni, E., Rusconi, F., Sestini, P., Viegi, G., 1998. Road traffic and adverse respiratory effects in children. Occupational and Environmental Medicine 55, 771–778.

Clark, A.I., McIntyre, A.E., Lester, J.N., Perry, R., 1984. Ambient measurements of aromatic halogenated hydrocarbons at urban, rural and motorway locations. Science of the Total Environment 39, 265–279.

Dockery, D.W., Arden Pope III, C., Xu, X., Spengler, J.D., Ware, J.H., Fay, M.E., Ferris Jr, B.G., Speizer, F.E., 1993.

An association between air pollution and mortality in six US cities. New England Journal of Medicine 329, 24–30.

Dockery, D.W., Speizer, F.E., Stram, D.O., Ware, J.H., Spengler, J.D., Ferris, B.G., 1989. Effects of inhalable particles on respiratory health of children. American Review of Respiratory Disease 139, 587–594.

Duffy, B.L., Nelson, P.F., Ye, Y., Weeks, I.A., Galbally, I.E., 1998. Emissions of benzene, toluene, xylenes and 1,3 butadiene from a representative portion of the Australian car fleet. Atmospheric Environment 32, 2693–2704.

Fischer, P.H., Hoek, G., van Reeuwijk, H., Briggs, D.J., Lebret, E., van Wijnen, J.H., Kingham, S., Elliot, P.E., 2000. Traffic related differences in outdoor and indoor concentrations of particles and volatile organic compounds in Amsterdam. Atmospheric Environment 34, 3713–3722.

Hildemann, L.M., Markowski, G.R., Jones, M.C., Cass, G.R., 1991. Submicrometer aerosol mass distributions of emissions from boilers, fireplaces, automobiles, diesel trucks, and meat-cooking operations. Aerosol Science and Technology 14, 138–152.

Hitchins, J., Morawska, L., Wolff, R., Gilbert, D., 2000. Concentrations of submicrometre particles from vehicle emissions near a major road. Atmospheric Environment 34, 51–59.

Horvath, H., Kreiner, I., Norek, C., Preining, O., Georgi, B., 1988. Diesel emission in Vienna. Atmospheric Environment 22, 1255–1269.

ISO 9835, 1993. Ambient air—Determination of a black smoke index.

Janssen, N.A.H., van Mansom, D.F.M., van der Jagt, K., Hoek, G., 1997. Mass concentration and elemental composition of airborne particles at street and background locations. Atmospheric Environment 31, 1185–1193.

Kinney, P.L., Aggarwal, M., Northridge, M.A., Janssen, N.A.H., Shepard, P., 2000. Personal exposures to $PM_{2.5}$ and diesel exhaust particles on Harlem sidewalks. Environmental Health Perspectives 108, 213–218.

Krämer, U., Koch, T., Ranft, U., Ring, J., Behrendt, H., 2000. Traffic-related air pollution is associated with atopy in children living in urban areas. Epidemiology 11, 64–70.

Kuhler, M., Kraft, J., Koch, W., Windt, H., 1988. Dispersion of car emissions in the vicinity of a highway. Environmental Meteorology. Kluwer Academic Publishers, Dordrecht, pp. 39–47.

Lamoree, D.P., Turner, J.R., 1999. PM emissions emanating from limited-access highways. Journal of the Air and Waste Management Association 49, 85–94.

Marple, V.A., Rubow, K.L., Turner, W., Spengler, J.D., 1987. Low flow rate sharp cut impactors for indoor air sampling: design and calibration. Journal of the Air Pollution Control Association 37, 1303–1307.

Morawska, L., Thomas, S., Gilbert, D., Greenaway, C., Rijnders, E., 1999. A study of the horizontal and vertical profile of submicrometer particles in relation to a busy road. Atmospheric Environment 33, 1261–1274.

Muzyka, V., Veimer, S., Shmidt, N., 1998. Particle-bound benzene from diesel engine exhaust. Scandinavian Journal of Work and Environmental Health 24, 481–485.

Nakai, S., Nitta, H., Maeda, K., 1995. Respiratory health associated with exposures to automobile exhaust II. Personal NO_2 exposure levels according to distance from the roadside. Journal of Exposure Analysis and Environmental Epidemiology 5, 125–136.

Nitta, H., Sato, T., Nakai, S., Maeda, K., Aoki, S., Ono, M., 1993. Respiratory health associated with exposure to automobile exhaust I. Results of cross-sectional studies in 1979, 1982, and 1983. Archives of Environmental Health 48, 53–58.

Palmes, E.D., Gunnison, A.F., DiMattio, J., Tomczyk, C., 1976. Personal sampler for nitrogen dioxide. American Industrial Hygiene Association Journal 37, 570–577.

Raizenne, M., Neas, L.M., Damokosh, A.I., Dockery, D.W., Spengler, J.D., Koutrakis, P., Ware, J.H., Speizer, F.E., 1996. Health effects of acid aerosols on North American children: pulmonary function. Environmental Health Perspectives 104, 506–514.

Rijnders, E., Janssen, N.A.H., van Vliet, P.H.N., Brunekreef, B., 2001. Personal and outdoor nitrogen dioxide concentrations in relation to degree of urbanisation and traffic density. Environmental Health Perspectives in press.

Rodes, C.E., Holland, D.M., 1981. Variations of NO, NO_2 and O_3 concentrations downwind of a Los Angeles freeway. Atmospheric Environment 15, 243–250.

Roorda-Knape, M.C., Janssen, N.A.H., de Hartog, J.J., van Vliet, P.H.N., Harssema, H., Brunekreef, B., 1998. Air pollution from traffic in city districts near major motorways. Atmospheric Environment 32, 1921–1930.

Schindler, S., Ackerman-Liebrich, U., Leuenberger, P., Monn, C., Rapp, R., Bolognini, G., Bongard, J.P., Brandli, O., Domenighetti, E., Karrer, W., Keller, R., Medici, T.E., Perruchaud, A.P., Schoni, M.H., Tschopp, J.M., Villiger, B., Zellweger, J.P., 1998. Associations between lung function and estimated average NO_2 in eight areas of Switzerland. Epidemiology 9, 405–411.

Ulrich, E., Israël, G.W., 1992. Diesel soot measurement under traffic conditions. Journal Aerosol Science 1 (suppl 23), S925–928.

VDI 2465, 1995. Messen von Ruß (Immission) Chemisch-analytische Bstimmung des elementaren Kohlenstoffes nach extraction und Thermodesorption des organischen Kohlenstoffes.

van Vliet, P., Knape, M., de Hartog, J., Janssen, N.A.H., Harssema, H., Brunekreef, B., 1997. Air pollution from road traffic and chronic respiratory symptoms in children living near motorways. Environmental Research 74, 122–132.

Weiland, S.K., Mundt, K.A., Ruckman, A., Keil, U., 1994. Self-reported wheezing and allergic rhinitis in children and traffic density on street of residence. Annals of Epidemiology 4, 243–247.

Westerholm, R., Egebäck, K., 1994. Exhaust emissions from light- and heavy duty vehicles: chemical composition, impact of exhaust after treatment and fuel parameters. Environmental Health Perspectives 102 (suppl 4), 13–23.

Wolff, G.T., 1985. Characteristics and consequences of soot in the atmosphere. Environment International 11, 259–269.