

Estimating Long-Term Average Particulate Air Pollution Concentrations: Application of Traffic Indicators and Geographic Information Systems

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Background. As part of a multicenter study relating traffic-related air pollution with incidence of asthma in three birth cohort studies (TRAPCA), we used a measurement and modelling procedure to estimate long-term average exposure to traffic-related particulate air pollution in communities throughout the Netherlands; in Munich, Germany; and in Stockholm County, Sweden.

Methods. In each of the three locations, 40–42 measurement sites were selected to represent rural, urban background and urban traffic locations. At each site and fine particles and filter absorbance (a marker for diesel exhaust particles) were measured for four 2-week periods distributed over approximately 1-year periods between February 1999 and July 2000. We used these measurements to calculate annual average concentrations after adjustment for temporal variation. Traffic-related variables (eg, population density and traffic intensity) were collected using Geographic Information Systems and used in regression models predicting annual average concentrations. From these models we estimated ambient

air concentrations at the home addresses of the cohort members.

Results. Regression models using traffic-related variables explained 73%, 56% and 50% of the variability in annual average fine particle concentrations for the Netherlands, Munich and Stockholm County, respectively. For filter absorbance, the regression models explained 81%, 67% and 66% of the variability in the annual average concentrations. Cross-validation to estimate the model prediction errors indicated root mean squared errors of 1.1–1.6 $\mu\text{g}/\text{m}^3$ for $\text{PM}_{2.5}$ and 0.22–0.31 $\times 10^{-5}\text{m}^{-1}$ for absorbance.

Conclusions. A substantial fraction of the variability in annual average concentrations for all locations was explained by traffic-related variables. This approach can be used to estimate individual exposures for epidemiologic studies and offers advantages over alternative techniques relying on surrogate variables or traditional approaches that utilize ambient monitoring data alone.

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Recent interest has focused on traffic-related air pollution and the potential health effects associated with exposure.¹ Several studies have indicated relations between air pollutants originating from

traffic sources and health impacts.^{2–4} Additional population studies have indicated that potential surrogate measures of exposure to traffic-related pollution are associated with a variety of respiratory health endpoints.^{5–15} A major deficiency in these cohort, case-control and panel studies involves the estimation of exposure from outdoor sources, and specifically the ability to apply individual exposure estimates to large study populations. The development of methods to estimate individual exposures of study populations within a single urban area allows for the assessment of chronic exposures and health impacts associated with within-city variability in air pollution. To date, most assessments of the health impacts of long-term exposure have involved between-city comparisons using a limited number of monitors within each city, or the use of surrogates to estimate within-city variability in exposure. Such between-city comparisons are subject to exposure misclas-

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TABLE 1. Description of Measurement Sites in the Netherlands, Munich and Stockholm County*

	The Netherlands	Munich	Stockholm County
Number of sites†	40	40	42
Regional background (%)	12 (30)	0 (0)	9 (21)
Urban/suburban background (%)	16 (40)	23 (57)	21 (40)
Traffic (%)	12 (30)	17 (43)	12 (29)
Street canyon (%)‡	2 (5)	8 (20)	9 (21)
Distance to major street (m)§	100 (3–450)	55 (2–930)	90 (5–450)

* Traffic sites and street canyon designations are based on the criteria described in the Methods section. In the Netherlands, urbanization degree 1 and 2 of the municipality are considered as urban. In Stockholm County, sites in Solna are suburban, sites in Järfälla are rural and those in Stockholm are considered urban.

† Number of sites refers to the primary measurement sites in each location and does not include the continuous measurement site that was used for temporal trend adjustment.

‡ Defined by the field worker.

§ median (min–max).

sification as they rely on a small number of monitors. Surrogate variables for exposure to outdoor air pollution originating from traffic have not been directly validated for their use as exposure measures in epidemiologic studies. A further difficulty in the assessment of exposure to traffic-related air pollution is the inability of existing monitoring networks to assess the variability of air pollution concentrations within urban areas. Several studies have indicated that particle concentrations exhibit substantial spatial variability within urban areas, with higher concentrations found in city centers^{16,17} or near major roads.¹⁸ A potential solution to these problems is the use of Geographic Information Systems (GIS) in which geographic data can be combined with concentration measurements to estimate exposures for individual members of large study populations.¹⁹

Geographic-modeling approaches make feasible the application of models to large study populations because the geographic information is typically readily available, in contrast to spatially detailed air pollution concentration information. However, even exposure assessments based on geographic models may be inadequate unless these models have been validated as surrogates of exposure to air pollutants from outdoor sources. Alternatively, exposures can be estimated with dispersion²⁰ or air pollution and time-activity models that may also have the ability to incorporate indoor sources of exposure.²¹ Although such models may be useful, they are seldom validated with actual measurements and they require input data, specifically for emissions, that may not be readily available. A third approach to assessment of exposure to ambient air pollutants involves interpolating concentrations based on measurements conducted by monitoring networks.^{22,23} These methods are useful for assessing regional air pollution patterns, but they cannot identify small-scale variations in concentrations, given the density of most monitoring networks and given the spatial distribution of traffic sources.

Here we describe the application of a combined monitoring and GIS methodology to assess exposures to air

pollutants originating from traffic—a methodology that can be applied to large study populations. This approach extends the methods used in the recent “Small-Area Variations in Air Quality and Health” (SAVIAH) studies in England, the Netherlands, and the Czech Republic.²⁴ As part of an international collaborative study on the risks of development of childhood asthma and other allergic diseases (“Risk assessment of exposure to traffic-related air pollution for the development of inhalant allergy, asthma and other chronic respiratory conditions in children” or TRAPCA), outdoor air exposures to traffic-related pollutants were estimated for birth cohorts in the Netherlands, Germany and Sweden. For all three locations a common exposure assessment approach was used.

Methods

Air Sampling

Site Locations

In each of the three study locations (the Netherlands, Munich and Stockholm County) we selected 40–42 air pollution measurement sites according to common criteria. A detailed description of the site selection criteria is provided elsewhere.²⁵ Briefly, monitoring sites were selected to incorporate variation among the potential traffic-related predictor variables in the location of interest. In all locations two major types of sites were selected (Table 1). Urban/suburban background sites were those in which no more than 3000 vehicles per day typically pass through a circle with 50-meter radius around the site. Traffic sites were those urban/suburban sites with a greater number of vehicles per day and no other air pollution sources other than traffic nearby. These traffic sites were located in both “open” and “street canyon” streets in each country; a street canyon was defined according to the EuroAirnet criteria.²⁶ Regional background sites were also included in the Netherlands and in Stockholm County. In each of the three countries, the specific characteristics of the birth cohorts necessitated additional sampling site criteria as described in more detail below.

The cohort in the Netherlands (population = 16.0 million) includes individuals in both cities and smaller communities distributed within three regions of the country: the North, center and Southwest (Figure 1). The largest numbers of study subjects live in the cities of Rotterdam (population = approximately 590,000; 18% of cohort), Amersfoort (population = 120,000; 12% of cohort) and Spijkenisse (population = 70,000; 9% of

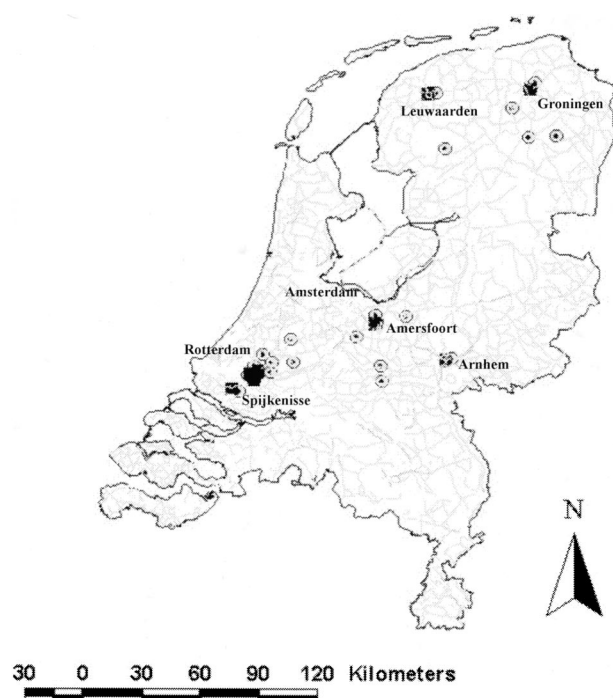


FIGURE 1. Location of air-sampling sites in the Netherlands. Circles denote background sites and squares denote traffic sites. Not all sites are visible because of overlapping of multiple sites within urban areas at the scale of the map.

cohort). The remaining subjects live in a large number of towns that differ widely in size. Regional background sites ($N = 12$) were those located in municipalities with less than 1000 addresses per km^2 . In the larger cities, urban/suburban background sites ($N = 16$) included sites in the city center and in suburban areas. The remaining sites were traffic sites ($N = 12$).

In Munich (population = 1.32 million) the 40 measurement sites were divided among urban/suburban background ($N = 23$) and traffic ($N = 17$) sites (Figure 2). These included 10 school-yard sites located <100 m, 100–300 m and >300 m from a major road, in which pollutants were measured in a previous study.²⁷ Traffic sites were located both at main roads and side roads to capture the maximum variability in air pollution concentrations within Munich. Urban/suburban background sites were distributed within the inner city ($N = 6$) and suburban areas ($N = 17$). Because of the distribution of the cohort study population in Munich, most of the urban/suburban background sites were located in the southwest and southeast suburbs of the city.

In the Stockholm County study area (population = 1.74 million), 42 sampling sites were selected within the catchment area of the birth cohort (Figure 3) according to the general criteria described above. Regional background sites were also included to characterize fully the variability of air pollution levels within the Stockholm County study area.

Measurements

Air pollution measurements were made between 1 March 1999 and 20 April 2000 in the Netherlands, 16 March 1999 and 21 July 2000 in Munich, and 9 February 1999 and 8 March 2000 in Stockholm (with 88% of measurements completed by 11 April 2000). Fourteen-day air samples for $\text{PM}_{2.5}$ were collected with Harvard impactors, as described in detail elsewhere.²⁵ Pump flow rates of 10 liter/minute were maintained with critical orifices, and sampling flows were measured before and after sampling with calibrated rotameters. Particles were collected on 37-mm 2- μm pore-size filters (Thermo Andersen, Smyrna, GA). To prevent filter overloading, timers were programmed to turn sampling pumps on for 15 minutes during each 2-hour period. In this way, sampling onto a single filter was conducted over 42 hours during each 14-day measurement period. At each location, we collected samples for four separate 14-day measurement periods distributed throughout the study period. After sample collection, filters were stored at 4°C and then transported to individual laboratories in each study location for weighing. Before weighing, samples were conditioned for at least 24 hours under controlled temperature (20–23°C) and relative humidity (30%–40%) conditions. Before and after sampling, filters were double weighed. If weights were not within 5 μg , filters were again weighed twice. Filter controls were used before and after sampling in all laboratories. Filter control weights had to be within 10 μg of their target weights, defined as the moving average of the past 10 weighing sessions for the control filters. Additionally, all filter weights were adjusted for the deviation of the control filters within a weighing session from the 10-day moving average. In addition, a laboratory weighing intercomparison was conducted with a set of five exposed and five blank filters.

Reflectance measurements were made by the Institute for Risk Assessment Sciences laboratory in the Netherlands, on the same $\text{PM}_{2.5}$ filters used for mass determination, with a Smoke Stain Reflectometer (Diffusion Systems Limited Model 43, Hanwell, London). Filter reflectance has previously been shown to be highly correlated with measurement of elemental carbon, a marker for particles produced by incomplete combustion.²⁸ We measured reflectance according to the Standard Operating Procedure²⁹ (a modification of ISO 9835, Determination of a Black Smoke Index) of the ULTRA study. Filters from Munich and Stockholm were sent to the Dutch laboratory in sealed Petri dishes. For each measurement period, a $\text{PM}_{2.5}$ field blank and duplicate were collected.

Measurements at the 40–42 sites in each country were not performed simultaneously, and therefore differences among the sites may have occurred because of temporal variation. As we intended these measurements

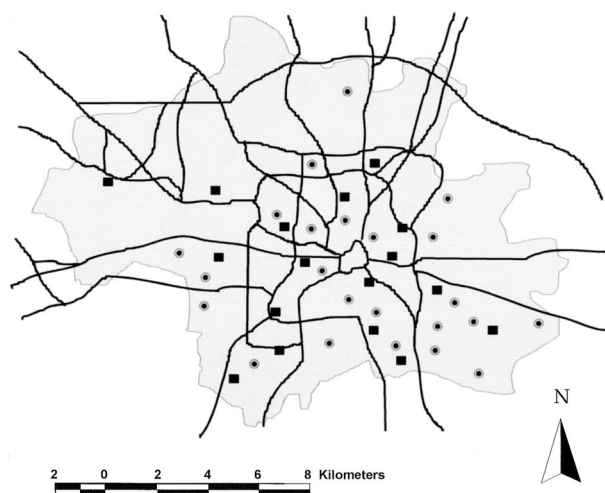


FIGURE 2. Location of air-sampling sites and major road network in Munich, Germany. Circles denote background sites and squares denote traffic sites.

to incorporate spatial variability only, the annual averages were adjusted for the impact of temporal variability using data from one site in each of the three study areas where 14-day integrated samples were made for the entire study period.²⁵ Measurements at these sites used procedures identical to the measurements made at the 40–42 sites in each country.

Geographic Data

The annual average concentrations calculated in the previous step were then related to predictor variables collected from GIS (Table 2). For the GIS variables, we also calculated different spatial scales (radius of buffer

around a measurement site). As with the sampling site selection, common criteria were followed by each center for the collection of geographic data and for the subsequent modeling. The core set of variables consisted of traffic intensity in buffers of 50, 250 and 1000 meters' radius; heavy vehicle traffic in the same buffers; distance to a major road; and household and population density in buffers of radius 300, 1000 and 5000 meters. Predictor variables were also calculated for buffers excluding the smaller spatial scales by subtracting the smaller scale buffers. Because of differences in data availability between the three countries, additional variables were collected, as described in detail below.

Geographic variables for the Netherlands were collected using ArcInfo (ESRI, Redlands, CA). For the road network in the Netherlands we used four ArcInfo coverages. Total vehicle and heavy vehicle (≥ 5.10 meters in length) traffic intensity coverages (50-m grid size) were obtained based on 1998 data from the Directorate-General of Public Works and Water Management.³⁰ These traffic intensities are counted and updated annually for all highways in the Netherlands. For provincial roads we used a coverage from the Emission Registration Collective.³⁰ Traffic intensities in this coverage were delivered by the Dutch provinces for the year 1990, and updated by the Emission Registration Collective for 1994. For the inner-city roads the Basisnetwerk (BASNET)³¹ coverage was used. This coverage does not contain traffic intensities, but categorizes the roads with the following levels: (1) highways, (3) access roads to areas with 25,000–50,000 inhabitants, (5) access roads to areas with 10,000–25,000 inhabitants, (7) access roads to areas with 5000–10,000 inhabitants, (9) access

TABLE 2. Description of Continuous Predictor Variables for the Measurement Sites

Variable	Buffer Radius (m)‡	The Netherlands		Munich		Stockholm County	
		Median	Range	Median	Range	Median	Range
Total traffic*	50	0	0–12,655	0	0–87,000	842	10–4,620
	250	138,523	0–510,753	35,500	0–278,000	17,099	192–56,880
	1,000	2,599,945	163,000–5,789,685	393,500	14,000–1,345,000	—	—
Heavy vehicles*	50	0	0–347	0	0–7,000	—	—
	250	0	0–22,218	1,900	0–18,000	—	—
	1,000	3,917	0–677,849	18,500	900–65,400	—	—
Traffic on nearest street*	NA	—	—	—	—	7,343	50–36,000
Distance from major road (m)	NA	455	4–5,221	50	2–930	112	5–450
Household density†	300	937	89–2,561	1,970	314–6,830	—	—
	1,000	7,134	1,132–15,643	13,773	2,740–45,996	—	—
	5,000	58,863	3,409–182,125	256,426	51,809–409,653	—	—
Population density†	300	1,893	412–6,166	3,673	765–10,761	2,938	15–7,538
	1,000	14,961	2,994–42,978	24,537	5,776–70,680	30,452	1,687–61,657
	5,000	132,157	8,377–430,796	454,727	112,479–699,913	711,651	56,549–1,034,529
Number of medium traffic roads	250	0	0–29	—	—	—	—
	1,000	90	0–228	—	—	—	—
	250	0	0–24	—	—	—	—
Number of high traffic roads	1,000	19.5	0–197	—	—	—	—

NA = not applicable; — = Data not calculated.

* Vehicles/day times street length (km).

† Number of addresses or persons within area of buffer.

‡ Buffer radius refers to the distance of the circular zone around each site for which the variables were calculated.

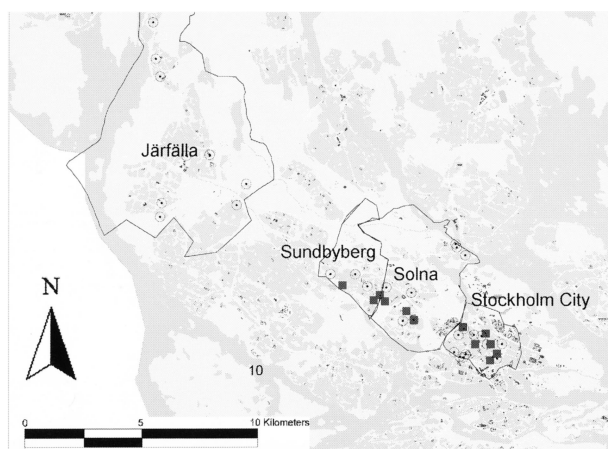


FIGURE 3. Location of air-sampling sites and major road network in Stockholm County, Sweden. Circles denote background sites and squares denote traffic sites.

roads to areas with 2000—5000 inhabitants, and (11) all other roads with flowing traffic.

For noise-monitoring purposes, the National Institute of Public Health and the Environment (RIVM) estimated the traffic intensities for the 1996 BASNET data, but only for inner-city roads. The estimation is based on the above road classification, information on traffic intensities for a limited number of streets in larger municipalities from municipal data, and the number of people living in a city. As these estimated traffic intensities have no separate estimation for heavy traffic and because there was a clear overlap in traffic intensities between categories, we also used the BASNET 2000 coverage for the entire country of the Netherlands. We transformed this vector data into raster data for the buffer calculations of distance to roads of different traffic categories, in the Grid environment of ArcInfo. Population- (1999) and address- (number of addresses, 1998) density data were obtained for 100-m grids from the National Institute of Public Health and the Environment based on data from the Central Bureau of Statistics.³²

Several additional categorical variables were used to describe the measurement sites in the Netherlands. First, we included an indicator to characterize regional differences in the concentrations of air pollutants. Ten sites (25%) were located in the north of the country, 12 (30%) in the middle, and 18 (45%) in the west. Because the traffic intensity data in the Netherlands included estimates for inner city roads only, alternative traffic indicators were developed based on the number of roads of different BASNET categories in the respective buffer zones. Sites in the Netherlands were also categorized according to their distance to medium-traffic (access roads to areas with $\geq 10,000$ inhabitants) or high-traffic (highways and access roads to areas with $\geq 25,000$ in-

habitants) roads according to the BASNET classification. Eight (20%) of the sites were within 50 m of such roads, slightly lower than the 12 sites that were designated as traffic sites according to our site criteria.

Geographic variables for Munich were determined using ArcView version 3.2 (ESRI, Redlands, CA). The intensity (vehicles/day) of total traffic and of heavy vehicles (weight > 7.5 tonnes) was based on 1997 traffic counts for the main road network (755 streets with > 1000 cars/day) obtained from the Planungreferat, Munich Municipal Authority. Address-density (number of accommodation units) and population-density data, both in 100-m raster grid format, were based on 1999 data from Planungreferat, Munich Municipal Authority.

The geographic data used for the Swedish locations were measured by a combination of GIS and manual measurements using detailed maps. Total traffic counts (calculated as cars/day times km street length), in circles with radii of 50 and 250 m around the measurement sites, were measured based on the most recent (1987–1999) municipal traffic-count data for Stockholm,³³ Solna, Sundbyberg and Järfälla (unpublished municipal traffic count data for Solna, Sundbyberg and Järfälla). If no counts were available for specific roads in the vicinity of a monitoring site, the numbers were estimated by a person with local information about the traffic conditions based on comparison with roads on which data were available. Traffic counts on the nearest street and nearest major street relative to the sampling sites were based on the municipal traffic data or estimation, as described above. Traffic-count data were available for approximately 40% of the roads nearest to cohort addresses, including all major roads. The distance to the nearest streets was observed at the sampling site, whereas the distance to the nearest major street was calculated manually from printed maps. The population counts were calculated in three circles with radii of 300, 1000 and 3000 m, based on 1995 population-registry data³⁴ with a variable grid size (100- to 2000-m grids). For each circle, the average was taken for the population density of all grids with centroids inside the circle. When there was no centroid in the circle, the value of the nearest grid was applied.

Additional variables not readily available in GIS were also added to the regression models to determine the extent to which predictions could be improved if such information were available for the entire study population. These were mostly variables determined manually for the measurements sites and recorded on the measurement-site questionnaire (sampling height, street type, canyon, type of sampling site; street, rural background, urban background). These variables are not as easy to obtain for all cohort addresses, so they mostly serve the purpose of illustrating the potential to explain the variability of air pollution.

TABLE 3. Annual Average Concentrations of PM_{2.5} Mass ($\mu\text{g}/\text{m}^3$) and PM_{2.5} Filter Absorbance (10^{-5}m^{-1}) Measurements

	PM _{2.5} ($\mu\text{g}/\text{m}^3$)			PM _{2.5} Filter Absorbance (10^{-5}m^{-1})		
	Netherlands	Munich	Stockholm County	Netherlands	Munich	Stockholm County
Minimum	13.7	11.2	7.9	0.82	1.37	0.70
10th%	14.3	11.9	8.8	0.94	1.44	0.89
25th%	15.3	12.4	9.4	1.26	1.60	1.04
50th%	17.7	13.3	10.2	1.57	1.71	1.21
Mean	17.5	13.6	10.3	1.64	1.84	1.28
75th%	18.4	14.3	11.0	1.83	2.00	1.45
90th%	21.4	16.0	12.0	2.45	2.36	1.69
Maximum	25.7	19.7	16.0	3.21	3.23	2.28

Exposure Modeling

The relation between the geographic variables (independent variables) and the annual average air pollution concentrations (dependent variables) for the 40–42 sites was analyzed by multiple linear regression. Geographic variables and air pollution measurements were available for all measurement sites in each country. Because several of the variables included multiple spatial scales, we selected the most relevant spatial scale by separately entering all of the available spatial scales for a specific variable, and then evaluating the percentage of the explained variation. The scale with the highest adjusted R^2 was selected. Next, we added the other spatial scales to the first selected scale separately and the change in adjusted R^2 was evaluated. This procedure was applied starting with the most influential predictor variable based on univariate regression. Next, we assessed multiple regression models including the most influential variable and the different spatial scales of the same variable in separate models. If the adjusted R^2 was higher for the multiple regression model, that model was used. Finally, the effect of adding another variable to the core model was evaluated using the adjusted R^2 until a model with the highest adjusted R^2 was obtained.

The precision of the exposure models developed based on the measurements and geographic variables for the 40 sites was evaluated by a cross-validation procedure. This involved using the regression model for 39 of the measurement sites to predict the concentration at the remaining site. This procedure was conducted for each of the 40 sites and these results were compared with the measured annual average concentrations determined for each of the sites. The root mean squared error (RMSE) was calculated as the square root of the sum of the squared differences of the observed concentration at site i and the predicted concentration at site i from a model developed without site i .

Results

Table 1 presents summary statistics of the breakdown of sampling sites in each location by the various classification criteria. As described above, the distribution of

sampling sites differed between locations because of the different characteristics of the cohorts. Most notably, the sites in Munich were all urban, whereas those in Stockholm County and the Netherlands included rural sites. The Netherlands contained a much lower percentage of street canyon sites than the other two locations because of the more suburban and smaller cities that were used for sampling. The proportion of traffic sites and the mean distance to the nearest major streets were similar for the three locations, demonstrating adherence to the common original site location criteria.

Air Pollution

Descriptive statistics of the air pollution measurements are shown in Table 3. Detection limits for PM_{2.5} mass concentrations (three times the standard deviation of field blanks) were 1.6, 3.4 and 0.7 $\mu\text{g}/\text{m}^3$ for the Netherlands, Munich and Stockholm, respectively (Table 3). For filter reflectance, detection limits were 0.1, 0.2 and 0.06 10^{-5}m^{-1} . The laboratory intercomparison indicated that the laboratories in Munich and Stockholm reported higher blank and control filter weights than did the laboratory in the Netherlands. These differences corresponded to PM_{2.5} concentration differences of 0.1 for Munich and 1.5 $\mu\text{g}/\text{m}^3$ for Stockholm. Although these differences cannot be explained by filter handling or transport, they may be partially explained by differences in weighing-room conditions.²⁵ As expected, concentrations measured at traffic sites were higher than those measured at the corresponding background sites. Urban sites also had higher concentrations than regional background sites for both PM_{2.5} and absorbance. Somewhat greater differences were found for absorbance than for PM_{2.5} when comparing across the different site types. This was expected given that traffic sources are thought to be the major contributor to filter absorbance but only one of several contributors to urban PM_{2.5} concentrations.

Geographic Variables

Descriptive statistics of the geographic predictor variables are presented in Table 2. Traffic intensities in the 50-m buffer for Munich and the Netherlands were zero

for more sites than expected based upon our prior expectation of the classification of sites, probably attributable to inaccurate estimations and incomplete coverage used to determine intensities in the GIS databases. Household and population densities were larger in Munich than in the Netherlands and Stockholm County (both the median and the maximum). This is to be expected because rural and suburban sampling sites were included in the Netherlands and Stockholm County. In both centers, the range across sites was substantial.

Many of the predictor variables were highly correlated. In Munich, heavy and total traffic intensity were highly correlated ($r > 0.9$) at all three spatial scales, although in the Netherlands heavy traffic intensity was uncorrelated with any of the other variables. The numbers of BASNET category 1–3 or 5–7 streets were correlated with estimated traffic intensity at the same spatial scale ($r \sim 0.6$ – 0.7). In Stockholm County the correlation between the nearest-street traffic intensity and the traffic intensity in the 50-meter buffer was high ($r = 0.8$).

Population and household density were highly correlated at all three spatial scales ($r > 0.9$) in all locations. The correlation of population and address density at different spatial scales was high as well in all three countries; for example, in Munich the correlation between household density in the 300-meter buffer and the 1000-meter buffer was 0.8. The correlation was decreased by calculating the difference between the 300- and 1000-meter buffers such that the additional contribution of the larger area buffer could be evaluated.

Traffic intensity and population density at the same spatial scale were only moderately correlated in Munich ($r \sim 0.1$ – 0.5) with the exception of traffic intensity in the 1000-meter buffer and household density in the 5000-meter buffer ($r = 0.75$). In the Netherlands, total traffic intensity was correlated ($r = 0.6$ – 0.8) with address and population density at the 1000- and 5000-meter scale. This was not the case for heavy vehicles traffic intensity. In Stockholm County there was a high correlation between population density in the 5000-meter buffer and traffic intensity in the 250- to 50-m buffer ($r = 0.7$).

Regression Models

The final models (GIS models in Tables 4 and 5) used for the calculation of cohort exposures are presented in Tables 4 and 5. These models include only variables that were also available for the cohort addresses. In all locations, a substantial fraction of the variability in annual average concentrations was explained by geographic variables. As anticipated, a larger fraction of the variability of the absorbance was explained than for $PM_{2.5}$.

To increase the comparability with the other two centers, we performed a sensitivity analysis in the Netherlands in which the data were restricted to the mea-

surements sites in the Rotterdam metropolitan area ($N = 18$), an urban area that more closely resembles the urban study areas in Munich and Stockholm. For the Rotterdam-only analysis, the regression model for $PM_{2.5}$ explained substantially less variability than the model developed for the entire country ($R^2 = 0.53$), whereas the absorbance model explained a similar amount of variability ($R^2 = 0.77$). This result is likely attributable to the lack of variability in $PM_{2.5}$ within a single urban area (Rotterdam) and suggests that the greater amount of explained variability for the $PM_{2.5}$ model in the Netherlands, relative to the other two locations, is a result of regional variation in $PM_{2.5}$ concentrations across the country (Table 4). When restricting the analysis to the Rotterdam sites, there was good agreement in the amount of explained variability in measured concentrations between the three locations.

In all three locations, the R^2 of the models could be further improved by including non-GIS variables ("best" models in Tables 4 and 5). This suggests that the available GIS variables, although predicting a substantial degree of variability, did not capture all explainable variability in the very small-scale differences and local characteristics affecting air pollution.

The validity of the regression models used for estimation of cohort exposures was evaluated by cross-validation. Mean prediction errors (RMSE, calculated as the square root of the sum of the squared differences of the observed concentration at site i and the predicted concentration at site i from a model developed without site i) were very similar for the Netherlands ($1.59 \mu\text{g}/\text{m}^3$ for $PM_{2.5}$ and $0.31 \cdot 10^{-5} \text{m}^{-1}$ for absorbance), Munich ($1.35 \mu\text{g}/\text{m}^3$ for $PM_{2.5}$ and $0.31 \cdot 10^{-5} \text{m}^{-1}$) and Stockholm ($1.10 \mu\text{g}/\text{m}^3$ and $0.22 \cdot 10^{-5} \text{m}^{-1}$). The ratio of the RMSE to the range in concentration across sites ranged from 13% to 16%. The RMSE was lower at background sites (the Netherlands: $1.07 \mu\text{g}/\text{m}^3$ for $PM_{2.5}$ and $0.24 \cdot 10^{-5} \text{m}^{-1}$ for absorbance; Munich: 0.97 and 0.19; Stockholm: 0.83 and 0.17) than for traffic sites (the Netherlands: $2.39 \mu\text{g}/\text{m}^3$ for $PM_{2.5}$ and $0.43 \cdot 10^{-5} \text{m}^{-1}$ for absorbance; Munich: 1.87 and 0.45; Stockholm: 1.59 and 0.30). The reason for this difference in RMSE is probably that the actual concentration at traffic sites is determined by various factors that are difficult to characterize (traffic intensity, traffic speed, street configuration and distance to street).

Application of Models to Cohort Addresses

The regression models described above were then applied to the home addresses of the cohort. For all the addresses, we collected data on all geographic predictor variables that were used in the regression models. In the Netherlands, geographic coordinates could be found for

TABLE 4. Results of Regression Models for PM_{2.5} (μg/m³)

Variable	GIS Model‡			"Best" Model§		
	Slope	SE	R ² Full Model (Individual Variables)†	Slope	SE	R ² Full Model (Individual Variables)†
The Netherlands			0.73			0.78
Number of high-traffic roads (250 m)	0.178	4.92 × 10 ⁻²	0.35	0.127	4.84 × 10 ⁻²	0.35
Address density (300 m buffer)	1.69 × 10 ⁻³	4.46 × 10 ⁻⁴	0.17	1.25 × 10 ⁻³	4.35 × 10 ⁻⁴	0.17
Region West*	2.23	0.586	0.11	2.22	0.540	0.11
Region middle*	3.27	0.634	0.10	3.34	0.581	0.10
Traffic site	—	—	—	1.34	0.648	0.05
Distance to nearest major road (m)	—	—	—	-1.53 × 10 ⁻³	2.47 × 10 ⁻³	0.003
Munich			0.56			0.76
Traffic intensity (250–50 m)	1.35 × 10 ⁻⁵	3.28 × 10 ⁻⁶	0.29	8.69 × 10 ⁻⁶	2.87 × 10 ⁻⁶	0.29
Traffic intensity (50 m)	3.32 × 10 ⁻⁵	1.02 × 10 ⁻⁵	0.19	2.75 × 10 ⁻⁵	9.42 × 10 ⁻⁶	0.19
Address density (300 m)	3.26 × 10 ⁻⁴	1.27 × 10 ⁻⁴	0.08	2.78 × 10 ⁻⁴	1.09 × 10 ⁻⁴	0.08
Street canyon	—	—	—	0.846	0.499	0.07
Traffic site	—	—	—	2.01	0.607	0.05
Distance to nearest major road (traffic sites) (m)	—	—	—	-1.52 × 10 ⁻²	6.18 × 10 ⁻³	0.04
Distance to nearest road (traffic sites) (m)	—	—	—	-8.59 × 10 ⁻²	4.20 × 10 ⁻²	0.03
Stockholm County			0.50			0.63
Traffic flow on nearest road (vehicles/day)	7.27 × 10 ⁻⁵	1.91 × 10 ⁻⁵	0.37	7.32 × 10 ⁻⁵	1.83 × 10 ⁻⁵	0.37
Population density (5000–1000 m)	1.75 × 10 ⁻⁶	5.60 × 10 ⁻⁷	0.13	1.27 × 10 ⁻⁶	6.02 × 10 ⁻⁷	0.13
Street canyon	—	—	—	1.23	0.413	0.13

SE = standard error of the slope.

* Region North is the reference region.

† The R² for the full model is indicated for each region along with the additional variation explained with previously entered variables already in the model.

‡ GIS model refers to a model with variables that were also available for the cohort addresses.

§ The "best" model includes additional variables not available in GIS format or for cohort addresses.

|| Distances refer to the radius of the buffer zone (in meters) around the sampling site.

4135 of the 4146 subjects (99.7%). In both Munich and Stockholm 100% of the cohort addresses were successfully geocoded. Table 6 describes the estimated exposures for each of the cohorts as well as those measured at the measurement sites used to generate the regression models. The contrast in cohort exposures was smaller for PM_{2.5} than for absorbance. In all centers, the variability in estimated exposures was similar to the variability measured at the monitoring sites, although there were some differences for the various locations and metrics.

The range in exposures estimated for the cohort was only slightly larger than the range for the sampling sites. The minimum estimated exposure for PM_{2.5} in the Netherlands was 1.4% lower than the lowest value measured at one of the monitoring sites, whereas the highest estimated exposure for PM_{2.5} in Munich was 10% higher than the highest measured value. The maximum values estimated for filter absorbance were 15% higher than the highest value measured at one of the sampling sites in the Netherlands and 33% higher in Munich. The estimated exposures for Stockholm were within the range of measured concentrations for both PM_{2.5} and absorbance. Mean concentrations were similar for the estimates and measurements for all locations for both PM_{2.5} and absorbance. Based on these results, there was only limited extrapolation outside the concentrations measured at the sampling sites.

Discussion

We were able to develop highly predictive regression models in all three locations. Geographic variables explained a greater proportion of the variability in measured concentrations for absorbance than for PM_{2.5}. This indicates that the geographic variables we used are more closely related to absorbance measurements than to PM_{2.5} measurements. This is expected as these geographic variables were selected specifically to predict traffic, for which filter absorbance is used as a surrogate measure of traffic-related air pollution. Previous work has shown that filter absorbance is more strongly related to road distance, and therefore presumably to traffic, than is PM_{2.5}.^{18,35} To explain a greater proportion of the variability in PM_{2.5} would require additional variables that relate to other contributors to urban PM_{2.5} concentrations, such as regional concentrations and industrial sources. Despite the greater proportion of explained variability in the absorbance models relative to the PM_{2.5} models, the mean (across all sites and countries) precision of the predicted concentrations (the measurement error as a percentage of the range in measured concentrations) was 14% for both PM_{2.5} and absorbance.

As indicated in Tables 4 and 5, there were differences in the predictive power of the regression models between the three locations. For both PM_{2.5} and filter absorbance,

TABLE 5. Results of Regression Models for PM_{2.5} Filter Absorbance (10⁻⁵m⁻¹)

Variable	GIS Model‡			"Best" Model§		
	Slope	SE	R ² Full Model (Individual Variables)†	Slope	SE	R ² Full Model (Individual Variables)†
The Netherlands			0.81			0.90
Number of high-traffic roads (250 m)	4.26×10^{-2}	1.13×10^{-2}	0.49	4.05×10^{-2}	8.10×10^{-3}	0.49
Address density (300 m)	3.71×10^{-4}	8.55×10^{-5}	0.17	2.45×10^{-4}	6.47×10^{-5}	0.17
Region West*	0.485	0.113	0.09	0.449	8.13×10^{-2}	0.09
Region middle*	0.534	0.122	0.03	0.515	8.73×10^{-2}	0.03
Minimum distance to nearest major road <50 m	0.295	0.135	0.03	3.84×10^{-2}	0.113	0.03
Traffic site	—	—	—	0.541	9.37×10^{-2}	0.09
Munich			0.67			0.83
Traffic intensity (250–50 m)	3.45×10^{-6}	7.50×10^{-7}	0.35	2.73×10^{-6}	6.38×10^{-7}	0.35
Traffic intensity (50 m)	1.10×10^{-5}	2.19×10^{-6}	0.30	1.04×10^{-5}	2.00×10^{-6}	0.30
Population density (5000–300 m)	4.50×10^{-7}	3.05×10^{-7}	0.03	1.57×10^{-7}	2.67×10^{-7}	0.03
Population density (300 m)	5.79×10^{-6}	1.86×10^{-5}	0.001	-1.26×10^{-5}	1.46×10^{-5}	0.001
Distance to nearest road (traffic sites) (m)	—	—	—	-2.47×10^{-2}	8.63×10^{-3}	0.05
Distance to nearest road (background sites) (m)	—	—	—	-3.64×10^{-3}	1.27×10^{-3}	0.04
Street canyon	—	—	—	0.139	0.111	0.04
Traffic site	—	—	—	0.457	0.124	0.03
Stockholm County			0.66			0.76
Traffic flow on nearest street	2.19×10^{-5}	3.71×10^{-6}	0.54	1.93×10^{-5}	4.33×10^{-6}	0.54
Population density (5000–1000 m)	4.12×10^{-7}	1.09×10^{-7}	0.12	3.01×10^{-7}	1.18×10^{-7}	0.12
Street canyon	—	—	—	0.228	8.69×10^{-2}	0.06
Traffic site	—	—	—	9.91×10^{-2}	9.36×10^{-2}	0.04

SE = standard error of the slope.

* Region North is the reference region.

† The R² for the full model is indicated for each region along with the additional variation explained with previously entered variables already in the model.

‡ GIS model refers to a model with variables that were also available for the cohort addresses.

§ The "best" model includes additional variables not available in GIS format or for cohort addresses.

|| Distances refer to the radius of the buffer zone (in meters) around the sampling site.

the regression models for the Netherlands had a higher proportion of explained variability than did the models for Munich or Stockholm. One explanation for this is that the sites in the Netherlands exhibited more variability than the other locations (Tables 1 and 2). Additionally, sites in the Netherlands were located in several regions of the country that differed substantially in their PM_{2.5} concentrations. When the analysis was restricted to the Rotterdam metropolitan area alone, the R² value for the PM_{2.5} model was quite similar to those obtained for Stockholm and Munich. In contrast, for absorbance the Rotterdam model still explained somewhat more variability than did the models for Stockholm and Munich. As the GIS databases were collected from external sources and were not developed specifically for this project, differences in the accuracy and

specificity of the GIS data may explain the observed differences in the regression models between locations. Despite the fact that the study locations were quite different in terms of air pollution concentrations, geographic scale and the variation of urbanization, similar regression variables were predictive in all three locations. For example, all PM_{2.5} models contained variables expressing population or address density and local traffic intensity. The variables in the absorbance models were quite similar to the PM_{2.5} models for all locations. This finding would imply that the measurement and modeling approach described here may be applied elsewhere with similar success. However, the developed quantitative models should not be used directly in locations other than the presented study areas. Differences in the vehicle fleet (age, type of fuel), street configurations

TABLE 6. Annual Average Air Pollutant Concentrations Measured at the Sampling Sites and Estimated for the Home Addresses of Cohort Members

	PM _{2.5} (μg/m ³)				PM _{2.5} Filter Absorbance (10 ⁻⁵ m ⁻¹)			
	Measurement Site		Cohort		Measurement Site		Cohort	
	Mean	Range	Mean	Range	Mean	Range	Mean	Range
Netherlands (N = 4,135)	17.5	13.7–25.7	16.9	13.5–25.2	1.6	0.8–3.2	1.7	0.8–3.7
Munich (N = 1,756)	13.6	11.2–19.9	13.4	11.9–21.9	1.8	1.4–3.3	1.8	1.4–4.4
Stockholm County (N = 579)	10.3	7.9–16.0	9.8	8.7–12.8	1.3	0.7–2.3	1.1	0.9–2.0

N = size of cohort with valid exposure estimates in each location.

TABLE 7. Results of Simple Regression Models of Road Distance and Annual Averages of PM_{2.5} ($\mu\text{g}/\text{m}^3$) and PM_{2.5} Filter Absorbance (10^{-5}m^{-1})

Variable	PM _{2.5} ($\mu\text{g}/\text{m}^3$)			PM _{2.5} Filter Absorbance (10^{-5}m^{-1})		
	Slope	SE	R ² Full Model (Individual Variables)*	Slope	SE	R ² Full Model (Individual Variables)*
The Netherlands			0.23			0.33
Distance to nearest major road	-8.13×10^{-3}	3.58×10^{-3}	0.20	-2.18×10^{-3}	7.50×10^{-4}	0.28
Distance to nearest road	-1.48×10^{-2}	1.15×10^{-2}	0.03	-3.82×10^{-3}	2.42×10^{-3}	0.05
Munich			0.19			0.23
Distance to nearest major road	-2.34×10^{-3}	1.26×10^{-3}	0.11	-6.88×10^{-4}	2.96×10^{-4}	0.15
Distance to nearest road	-2.13×10^{-2}	1.13×10^{-2}	0.08	-5.34×10^{-3}	2.66×10^{-3}	0.08
Stockholm County			0.33			0.39
Distance to nearest major road	-6.96×10^{-3}	1.96×10^{-3}	0.24	-1.83×10^{-3}	4.43×10^{-4}	0.29
Distance to nearest road	-2.67×10^{-2}	1.15×10^{-2}	0.09	-6.73×10^{-3}	2.60×10^{-3}	0.10

SE = standard error of the slope.

* The R² for the full model is indicated for each region along with the R² contribution of each individual variable for the model containing both listed variables.

and, potentially, altitude may result in different quantitative relations between geographic variables and air pollution. Also, differences in availability of GIS data between countries necessitate development of local regression models by conducting air pollution measurements.

Comparisons between the GIS and "best" models, as presented in Tables 4 and 5, indicate that the GIS models can be improved in all cases when data on very local conditions are available. For example, whether a location is designated as a traffic or background site and whether it can be categorized as a street canyon provided additional explanatory power to most of the models. Although such variables can be obtained by observation for a limited number of sites, they are not typically available in GIS databases nor can they be easily obtained for large sample sizes. This represents one limitation of the GIS based-modelling methodology. One explanation for the inability of GIS data to account for very local scale differences is that the grid size available for the modelling was typically 100 m, thus limiting the ability to detect differences in exposures for locations less than 100 m apart. Although limited air-monitoring information is available, several studies have suggested that traffic-related pollutants exhibit substantial variability at distances of 50 m or less from major roads.^{35,36} An additional limitation of the methodology is the restriction of exposure estimation to pollutants from outdoor sources. The use of outdoor concentrations estimated for the home addresses as proxies for at-home exposure to air pollutants originating from traffic is supported by previous studies indicating high correlations between indoor and outdoor NO₂ for homes without indoor combustion sources³⁷ and by studies associating indoor particulate matter and filter absorbance levels with traffic intensity.¹⁸ Recent work has shown that personal exposure to NO₂ is related to the degree of urbanization, the traffic density and distance to a nearby major road.³⁸

Despite these limitations, however, we have shown that this combined measurement and modelling methodology results in good predictions in all three locations. The models presented here can be compared with simpler exposure estimation approaches used in other studies, such as distance to nearest road,¹⁰⁻¹³ the intensity of traffic on the nearest road^{5,9,15} and self-reported traffic intensity.^{6,7,14} Regression models that include distance to the nearest road and distance to the nearest major road as predictors of PM_{2.5} concentrations are presented in Table 7. Compared with the models developed earlier (Tables 4 and 5), these simpler models explained a much lower proportion of the variability in measured concentrations. Because estimations of actual traffic intensity on the road adjacent to a monitoring site were available only for Stockholm, it was not possible to fully evaluate this simpler measure. For Stockholm, models including traffic intensity on the nearest road and the nearest major road had R² values of 0.37 for PM_{2.5} and 0.54 for filter absorbance. Again, these models have lower predictive power than those presented in Tables 4 and 5, which also include indicators of population density.

It is noteworthy that all of the more sophisticated models include some measure of address or population density in addition to various measures of traffic intensity and/or distance. Population or address density may serve as a surrogate for the general level of human activity (including traffic) in the vicinity of a monitoring site, whereas the more specific traffic variables describe the impact of nearby traffic. Population density has been associated with decreased driving speeds and increased emissions, suggesting that areas with higher population/address density may be subject to higher emissions per vehicle.³⁹ In general, variables describing traffic intensity appeared to have greater explanatory power than those describing distance to nearby roads. Thus, incorporation of variables in addition to road

distance or immediate traffic intensity provided additional explanatory power.

Because a substantial fraction of the variability in annual average concentrations for all locations was explained by GIS variables, the measurement and modeling approach described here can be used to predict exposures for epidemiologic studies. Three locations were included in the study. For all locations, similar model variables were found to be predictive of measured ambient concentrations. Given the general agreement between the models for the three locations and the fact that the locations differed in their degree of urbanization, this approach appears to be generalizable to other locations and possibly also beyond urban areas. Application of this methodology would require local measurements and model calibration. The exposure estimation approach we describe offers an advantage over traditional approaches that utilize ambient monitoring data alone, as individual exposure estimates can be computed for each member of the study population. It is therefore an attractive method for studying the health effects of long-term exposure to air pollutants that exhibit substantial within-community spatial contrasts.

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