Environmental Science & Technology

Article

pubs.acs.org/est

Development of Land Use Regression Models for $PM_{2.5}$, $PM_{2.5}$ Absorbance, PM_{10} and PM_{coarse} in 20 European Study Areas; Results of the ESCAPE Project

Marloes Eeftens,[†,*] Rob Beelen,[†] Kees de Hoogh,[‡] Tom Bellander,[§] Giulia Cesaroni,[∥] Marta Cirach,[⊥,#,▽] Christophe Declercq,[○] Audrius Dėdelė,[◆] Evi Dons,[¶,∞] Audrey de Nazelle,[⊥,#,▽] Konstantina Dimakopoulou,[⊗] Kirsten Eriksen,[⋈] Grégoire Falq,[○] Paul Fischer,[✡] Claudia Galassi,[◎] Regina Gražulevičienė,[◆] Joachim Heinrich,[◘] Barbara Hoffmann,[☆,√] Michael Jerrett,[$] Dirk Keidel,[%,∀] Michal Korek,[§] Timo Lanki,[&] Sarah Lindley,[@] Christian Madsen,[+] Anna Mölter,[©] Gizella Nádor,[¥] Mark Nieuwenhuijsen,[⊥,#,▽] Michael Nonnemacher,[⧳] Xanthi Pedeli,[⊗] Ole Raaschou-Nielsen,[⋈] Evridiki Patelarou,[£] Ulrich Quass,[Я] Andrea Ranzi,[Å] Christian Schindler,[%,∀] Morgane Stempfelet,[○] Euripides Stephanou,[◪] Dorothea Sugiri,[☆] Ming-Yi Tsai,[%,∀,◪] Tarja Yli-Tuomi,[&] Mihály J Varró,[¥] Danielle Vienneau,[‡] Stephanie von Klot,[Σ] Kathrin Wolf,[Σ] Bert Brunekreef,[†,∯] and Gerard Hoek[†]

[†]Institute for Risk Assessment Sciences, Utrecht University, P.O. Box 80178, 3508 TD Utrecht, The Netherlands

[‡]MRC-HPA Centre for Environment and Health, Department of Epidemiology and Biostatistics, Imperial College London, London, United Kingdom

[§]Institute of Environmental Medicine, Karolinska Institutet, Stockholm, Sweden

[∥]Epidemiology Department, Lazio Regional Health Service, Rome, Italy

[⊥]Center for Research in Environmental Epidemiology (CREAL), Barcelona, Spain

[#]IMIM (Hospital del Mar Research Institute), Barcelona, Spain

[▽]CIBER Epidemiología y Salud Pública (CIBERESP), Spain

[○]French Institute for Public Health Surveillance, Saint-Maurice, France

[◆]Vytautas Magnus University, Kaunas, Lithuania

[¶]VITO-MRG (Flemish Institute for Technological Research), Environmental Risk and Health unit, Mol, Belgium

[∞]Hasselt University, Diepenbeek, Belgium

[⊗]Department of Hygiene, Epidemiology & Medical Statistics, National and Kapodistrian University of Athens, Medical School, Athens, Greece

[⋈]Danish Cancer Society Research Center, Copenhagen, Denmark

[✡]Centre for Environmental Health, National Institute for Public Health and the Environment, Bilthoven, The Netherlands

[◎]AOU San Giovanni Battista – CPO Piedmont, Turin, Italy

[◘]HMGU Institute of Epidemiology I, Neuherberg, Germany

[☆]IUF Leibniz Research Institute for Environmental Medicine, and [√]Medical Faculty, Heinrich-Heine, University of Düsseldorf, Düsseldorf, Germany

[$]School of Public Health, University of California, Berkeley, California, United States

[%]Department of Epidemiology and Public Health, Swiss Tropical & Public Health Institute, Basel, Switzerland

[∀]University of Basel, Basel, Switzerland

[&]Department of Environmental Health, National Institute for Health and Welfare, Kuopio, Finland

[@]School of Environment and Development (Geography), The University of Manchester, Manchester, England

[+]Division of Epidemiology, Norwegian Institute of Public Health, Oslo, Norway

[©]Centre for Occupational and Environmental Health, The University of Manchester, Manchester, England

[¥]Department of Environmental Epidemiology, National Institute of Environmental Health, Budapest, Hungary

[⧳]Institute for Medical Informatics, Biometry and Epidemiology, University of Duisburg-Essen, Essen, Germany

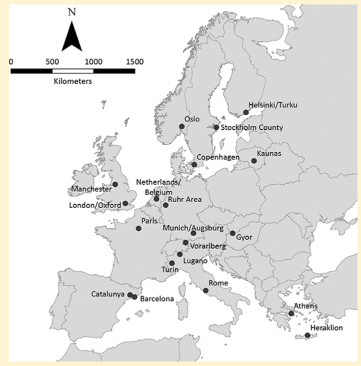
Received: May 22, 2012
Revised: August 30, 2012
Accepted: September 10, 2012
Published: September 10, 2012

 © 2012 American Chemical Society

[£]Department of Social Medicine, Medical School, University of Crete, Greece
[Я]Air Quality & Sustainable Nanotechnology, IUTA Institüt für Energie- und Umwelttechnik e.V., Duisburg, Germany
[Å]Regional Reference Centre on Environment and Health, ARPA Emilia Romagna, Modena, Italy
[¢]Environmental Chemical Processes Laboratory, University of Crete, Heraklion, Greece
[◪]Department of Environmental & Occupational Health Sciences, University of Washington, Seattle, Washington, United States
[Σ]HMGU Institute of Epidemiology II, Neuherberg, Germany
[∮]Julius Center for Health Sciences and Primary Care, University Medical Center Utrecht, Utrecht, The Netherlands

Supporting Information

ABSTRACT: Land Use Regression (LUR) models have been used increasingly for modeling small-scale spatial variation in air pollution concentrations and estimating individual exposure for participants of cohort studies. Within the ESCAPE project, concentrations of $PM_{2.5}$, $PM_{2.5}$ absorbance, PM_{10}, and PM_{coarse} were measured in 20 European study areas at 20 sites per area. GIS-derived predictor variables (e.g., traffic intensity, population, and land-use) were evaluated to model spatial variation of annual average concentrations for each study area. The median model explained variance (R^2) was 71% for $PM_{2.5}$ (range across study areas 35–94%). Model R^2 was higher for $PM_{2.5}$ absorbance (median 89%, range 56–97%) and lower for PM_{coarse} (median 68%, range 32– 81%). Models included between two and five predictor variables, with various traffic indicators as the most common predictors. Lower R^2 was related to small concentration variability or limited availability of predictor variables, especially traffic intensity. Cross validation R^2 results were on average 8–11% lower than model R^2. Careful selection of monitoring sites, examination of influential observations and skewed variable distributions were essential for developing stable LUR models. The final LUR models are used to estimate air pollution concentrations at the home addresses of participants in the health studies involved in ESCAPE.



1. INTRODUCTION

Epidemiological studies have shown adverse health effects of long-term exposure to air pollution.[1,2] Air pollution from motorized road traffic is a main public health concern in Europe.[3] Many studies have demonstrated large within-city contrasts in traffic related air pollutants in European and U.S. cities.[3−11] Land Use Regression (LUR) modeling has been used frequently to explain these spatial contrasts, using predictor variables derived from geographic information systems (GIS).[6,7,11] LUR models make use of a spatially dense network of measured air pollution concentrations. Each monitoring site is characterized by a set of potential predictors such as population density, land use and various traffic-related variables. Statistical modeling is used to determine which predictors best explain the pollution concentrations.[6,7,11] LUR modeling has generally been able to explain a large amount of spatial variability. An increasing number of epidemiological studies make use of LUR models for estimating outdoor air pollution concentrations at the home addresses of cohort subjects.[12,13]

Many LUR studies have used data on nitrogen oxides, usually because these can be easily obtained using low-cost passive samplers.[7] While health effects are probably more related to particles,[14,15] LUR models for particulate matter and absorbance are less numerous because they require a more intensive monitoring effort.[7] Routine monitoring networks often do not offer the required spatial density, do not measure all components of interest (e.g., soot) or do not measure at sites relevant for population exposure. Within Europe there is still a lack of $PM_{2.5}$ monitoring and PM monitoring is performed with continuous monitors that require correction factors and differ per country.[16]

So far, there are few LUR studies on the coarse fraction of particulate matter,[17] while there is increasing epidemiological evidence showing that coarse particles are associated with acute respiratory health effects.[18] Long-term effects of PM_{coarse} have not been studied extensively, partly because of a lack of spatially resolved data on coarse particle concentrations.[18]

The ESCAPE project (European Study of Cohorts for Air Pollution Effects, www.escapeproject.eu) was designed to study the effects of long-term air pollution exposure on health. ESCAPE makes use of health data from existing cohort studies. Exposures to air pollution were assessed for study participants' individual home address with LUR models based upon standardized specific PM monitoring campaigns in each of the study areas.

This paper describes the development and performance of the LUR models of 20 European study areas for $PM_{2.5}$, $PM_{2.5}$ absorbance, PM_{10}, and PM_{coarse}. The ESCAPE database is currently the largest database of spatially resolved PM data in Europe, allowing development of LUR models. We will discuss issues in LUR model development, such as influential observations, which have not often been addressed in the LUR literature. Results of the ESCAPE PM pollution measurements were recently accepted for publication.[19]

2. MATERIALS AND METHODS

For 20 study areas across Europe (Figure 1), LUR models were developed for $PM_{2.5}$, $PM_{2.5}$ absorbance, PM_{10} and PM_{coarse} based upon measured annual average concentrations. LUR models were developed using a range of GIS-derived predictor variables, from consistent European data sets compiled through ESCAPE and local data sets. Models were developed using a supervised stepwise method that maximized model explained variance, with a priori specified signs of slopes (e.g., positive for traffic intensity). Models were optimized locally with no attempt to force a common model to all study areas. This decision was based on the diversity of study areas and differences in available GIS predictor variables. LUR models were developed locally at each center, following a common manual (http://www.escapeproject.eu/manuals/). A workshop was attended by all local centers to

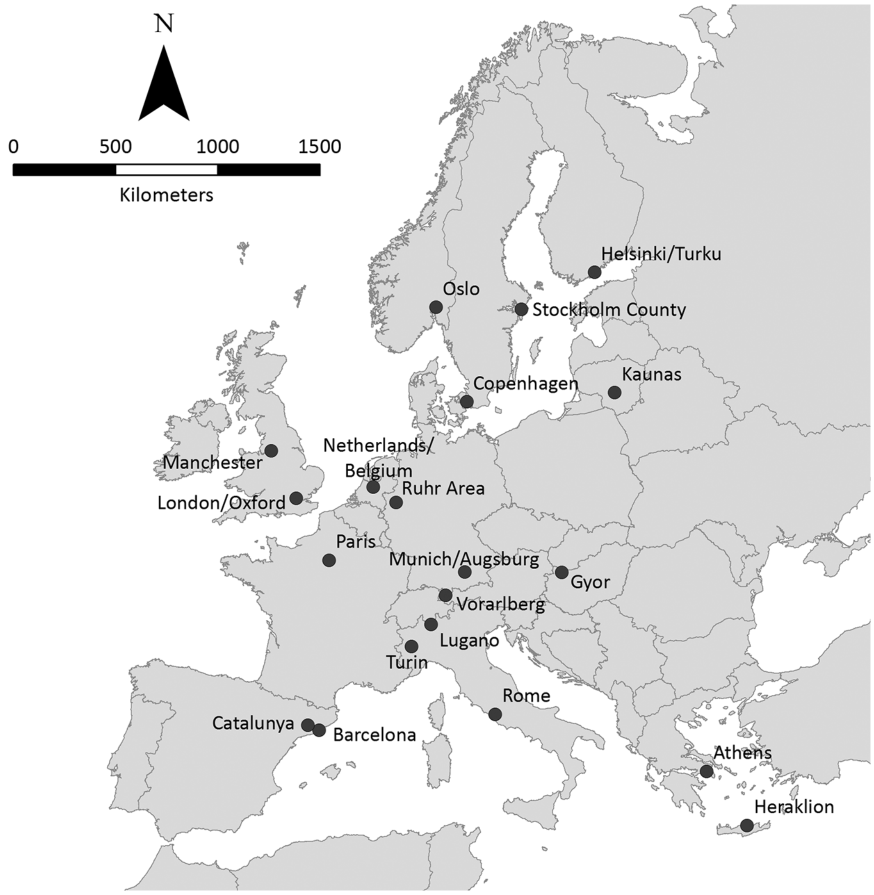


Figure 1. ESCAPE study areas.

standardize GIS analyses and LUR model development. Finalized LUR models were sent to the coordinating center for evaluation.

Air Pollution Measurement Data. The ESCAPE measurements and sampling site selection have been described previously.[19] Briefly, particulate matter (PM) was measured between October 2008 and April 2011. Twenty PM sampling sites were selected in each study area. In the larger study areas of The Netherlands and Belgium and Catalunya, forty sites were measured. Study areas were defined to represent the spatial distribution of the cohort addresses. We selected regional background, urban background and traffic sites. Traffic sites were overrepresented, and we selected a range of traffic intensities to limit outliers in modeling. Measurements in traffic sites (>10 000 vehicles.day^{-1}) were made at building façades, rather than the kerbside. A detailed description of each study area is given in the Online Supplement of Eeftens et al.[19] Most study areas comprised a major city and surrounding smaller towns. Each selected site was measured three times for 14 days, in the cold, warm and intermediate seasons. Two fractions of particulate matter (smaller than 2.5 μm ($PM_{2.5}$) and smaller than 10 μm (PM_{10})) were sampled using Harvard Impactors. The coarse fraction (PM_{coarse}) was calculated as the difference between PM_{10} and $PM_{2.5}$. Reflectance was measured on $PM_{2.5}$ filters and transformed into absorbance.[19] For each site, results from the three measurements were averaged to estimate the annual average, adjusting for temporal variation using a centrally located background reference site, which was operated for a whole year.[8,19] A temporal correction was calculated as the difference of each individual reference site measurement from the annual mean at the reference site. The calculated correction was then subtracted from all measurements that took place in that particular round.

GIS Predictor Data. *Positioning of Measurement Sites.* Multiple GPS measurements were taken at every site, but all positions were corrected manually to ensure an accurate position relative to roads on the detailed local road maps. This was done by someone who had personally visited the site.

Predictor Variables. Predictor variables were calculated for each site, using the site coordinates and digital data sets within a GIS. We used a combination of European data obtained centrally and local data. Local source data were collected because some data were not available on a European level or were more precise or more up-to-date. For traffic variables, we calculated circular buffers with radii of 25, 50, 100, 300, 500, and 1000 m around each monitoring site. For land use and population, we calculated buffers of 100, 300, 500, 1000, and 5000 m. A detailed description and an overview of all calculated variables, is shown in Supporting Information (SI) SI1.

The following GIS source data were available centrally:

1 Digital road network Road data were available at 1:10 000 resolution from Eurostreets version 3.1 digital road network, derived from the TeleAtlas MultiNet data set for the year 2008. The network includes road class but not traffic intensity.
2 Land use dataCORINE (COoRdination of INformation on the Environment) land cover data were available from the European Environment Agency (EEA) for the year 2000.[20,21] We used six land use categories: high density

residential land, low density residential land, industry, ports, urban green and natural land.[20,21]

3 Population density data Population data modeled at a 100 m grid were based upon land cover and the 2001 population density available from the EEA.[22,23]

4 Altitude Digital elevation data (SRTM 90 m) were obtained through the Shuttle Radar Topographic Mission, and available globally from CGIAR-CSI GeoPortal (http://srtm.csi.cgiar.org/). The map has a resolution of 90 m at the equator.

A detailed overview of the local GIS variables can be found in SI SI2. We required a spatial resolution of at least 100 m. Local GIS data included land use, population and household density, altitude and study-area specific variables such as distance to the sea. Detailed local road networks with linked traffic intensity were available for most areas. To account for variation in regional background in The Netherlands/Belgium, 10 regional background sites were measured, which allowed us to use an inverse distance weighted regional background concentration.[24] In the other (smaller) study areas, few regional background sites were measured as we anticipated little variation in regional background. We evaluated whether adding geographical coordinates to the final GIS model improved prediction, and if these trends were consistent with known pollution patterns.

LUR Model Development. Linear regression models were developed using a supervised stepwise selection procedure, first evaluating univariate regressions of the corrected annual average concentrations with all available potential predictors following procedures used before.[21] The predictor giving the highest adjusted explained variance (adjusted R^2) was selected for inclusion in the model if the direction of effect was as defined a priori. We then evaluated which of the remaining predictor variables further improved the model adjusted R^2, selected the one giving the highest gain in adjusted R^2, and the right direction of effect. Subsequent variables were not selected if they changed the direction of effect of one of the previously included variables. This process continued until there were no more variables with the right direction of effect, which added at least 0.01 (1%) to the adjusted R^2 of the previous model.

As final steps, variables with a p-value above 0.10 were removed from the LUR model. If the Variance Inflation Factor (VIF) was higher than 3 −indicating collinearity-, the variable with the highest VIF was removed and the model re-evaluated. Cook's D statistics were used to detect influential observations. Cook's D values above 1 were further examined by assessing the changes in model coefficients on excluding the responsible site. If removal of this site caused large changes in a specific variable's coefficient, the modeling procedure was repeated using all sites, but now without offering this variable.

Overall model performance was evaluated by leave-one-out cross validation (LOOCV): each site was sequentially left out from the model while the included variables were left unchanged. The Moran's I statistic was calculated to indicate spatial autocorrelation of the model residuals.

3. RESULTS

Within-Area Concentration Contrasts. Pollutant ranges are shown in Tables 1−3 for each study area and in more detail in Eeftens et al.[19] For most areas, substantial variation was present within the area. Within-area contrasts were largest for PM_{coarse} and $PM_{2.5}$ absorbance. Within-area contrasts differed between areas, for example, for $PM_{2.5}$ lower contrasts were found in Manchester, Ruhr Area, Gyor and Turin.

Available Predictor Variables. In 18 of the 20 study areas local traffic intensity data was collected. Exceptions were Heraklion and Catalunya. In many study areas, few sites were within 100, 300, or 500 m of a port, forest or industrial area, resulting in many 0-values. Similarly, for several areas a large number of 0-values occurred for major roads in small buffer (25 or 50 m). Generally, variables with less than 4−5 nonzero values were not offered in the modeling, but we evaluated the stability of parameter estimates for each model.

Land Use Regression Modeling. The LUR models for $PM_{2.5}$, $PM_{2.5}$ absorbance, and PM_{coarse} are described in Tables 1−3 and those for PM_{10} in SI SI3, Table 1. Descriptive statistics of the predictor variables used in the models can be found in SI SI4. In four areas, one site was excluded from modeling because only one successful measurement was available (Lugano, Oslo) or the site was too influential *and* was considered a non-representative site (Stockholm County, Manchester), further discussed in the modeling experiences section in the Discussion.

$PM_{2.5}$ Models. In most study areas, a substantial fraction of the measured spatial variability was explained by the available GIS predictor variables (Table 1). The median model explained variance (R^2) was 71% and ranged from 35% (Manchester) to 94% (Stockholm County).The variation in R^2 is partly related to the limited availability of relevant predictors, especially local traffic intensity data. The two areas without local or limited traffic intensity data (Heraklion, Catalunya) both had R^2 below the median. In Barcelona (part of the Catalunya study area), local traffic data was available and a much better model could be developed. Small variation of measured concentrations may have contributed to lower R^2 in some areas, such as Manchester, but overall the association is not strong (Table 1). There was no clear geographical pattern of the magnitude of R^2 across Europe.

For most models, the differences between the model R^2 and the leave-one-out cross validation R^2 was less than 15%, indicating stable models. Models included two to five predictor variables. Traffic indicators were included in 18 of the 20 models, with traffic intensity in various buffer sizes included in most models. Less often included predictors were residential land use, population density, industrial/port and natural land use.

$PM_{2.5}$ Absorbance Models. Model R^2 was higher for $PM_{2.5}$ absorbance (median 89%) than for $PM_{2.5}$, probably related to the larger spatially variability (Table 2). In Manchester, R^2 was high, whereas no reliable model could be developed for $PM_{2.5}$. Explained variance differed across areas from 56% (Heraklion) to 97% (Ruhr Area). The low value in Heraklion is likely due to the lack of traffic intensity data. Differences between model R^2 and LOOCV R^2 were generally lower than 10%, indicating stable models. The models included two to five predictors. In all models traffic variables were present. With the exception of Heraklion, all models included small-scale traffic variables, such as traffic intensity in the nearest street, the product of traffic intensity on the nearest major street and inverse distance and small buffers ($\leq$100 m) of traffic intensity. Models also included traffic in larger buffers and land use predictors.

PM_{coarse} Models. The median model R^2 was 68%, with a range from 32% (Kaunas) to 81% (Munich/Augsburg) (Table 3). Model R^2 was the lowest from the modeled PM metrics. Differences between model explained variance and cross validation were generally larger for PM_{coarse} than for the other PM metrics. PM_{coarse} models generally included two to three predictor variables, fewer than for the other PM metrics. In all areas except

Table 1. Description of Developed LUR Models for $PM_{2.5}$, Including Descriptive Statistics of the Measured Concentrations

study area	LUR model[a]	R^2 of model	R^2 validation	RMSE (validation) ($\mu g/m^3$)	number of sites[b]	Moran's I (*p*-value)	measured concentration ($\mu g/m^3$)[c]
Oslo, Norway	8.08 + 1.30 × 10^{-3} × HHOLD_500 + 9.28 × 10^{-5} × TRAFNEAR − 5.95 × 10^{-8} × NATURAL_5000	74%	68%	1.2	19	−0.05 (0.56)	8.6 [5.0−12.9]
Stockholm County, Sweden	7.95 − 8.96 × 10^{-6} × WATER_500 − 1.48 × 10^{-7} × WATER_500_5000 + 1.37 × 10^{-5} × HEAVYTRAFLOAD_50 + 3.66 × 10^{-4} × ROADLENGTH_500	87%	78%	0.8	19	−0.02 (0.28)	8.3 [4.4−11.3]
Helsinki/Turku, Finland	9.25 − 6.75 × 10^{-6} × NATURAL_500[d] + 6.34 × 10^{-7} × TRAFMAJORLOAD_50	67%	53%	1.0	20	−0.30 (0.03)	8.6 [5.3−12.3]
Copenhagen, Denmark	9.12 + 1.96 × 10^{-4} × ROADLENGTH_500 − 2.20 × 10^{-3} × GREEN_100[d]	62%	55%	1.1	20	−0.02 (0.68)	11.1 [8.4−14.0]
Kaunas, Lithuania	14.74 + 1.92 × 10^{-2} × POP_100 + 1.67 × 10^{-4} × TRAFMAJOR	60%	45%	2.6	20	−0.05 (0.45)	21.1 [16.6−30.3]
Manchester, UK	9.41 + 1.24 × 10^{-6} × HDRES_1000	35%	21%	0.8	19	−0.08 (0.50)	9.8 [8.1−11.9]
London/Oxford, UK	7.19 + 1.38 × 10^{-3} × INTMAJORINVDIST + 2.65 × 10^{-4} × ROADLENGTH_500	82%	77%	1.4	20	−0.19 (0.20)	11.2 [7.0−21.2]
Netherlands/Belgium	9.46 + 0.42 × REGIONALESTIMATE + 0.01 × MAJORROADLENGTH_50 + 2.28 × 10^{-9} × TRAFMAJORLOAD_1000	67%	61%	1.2	40	0.02 (0.77)	17.7 [12.7−21.5]
Ruhr Area, Germany	81.73 + 5.61 × 10^{-8} × HEAVYTRAFLOAD_1000 + 1.20 × 10^{-7} × INDUSTRY_5000 + 1.04 × 10^{-4} × POP_1000 − 2.57 × 10^{-5} × XCOORD	88%	79%	0.9	20	−0.02 (0.64)	18.5 [15.5−21.6]
Munich-Augsburg, Germany	11.90 + 1.94 × 10^{-2} × MAJORROADLENGTH_50[d] + 4.95 × 10^{-4} × ROADLENGTH_300[d] − 14.30 × URBGREEN_5000[d] + 7.41 × 10^{-9} × TRAFMAJORLOAD_1000[d]	78%	62%	1.0	20	−0.13 (0.49)	14.3 [9.7−17.6]
Vorarlberg, Austria	25.44 + 0.11 × BUILDINGS_100 − 0.65 × SQRALT	57%	42%	1.5	20	0.09 (0.06)	13.3 [8.8−17.3]
Paris, France	10.38 + 5.34 × 10^{-4} × MAJORROADLENGTH_500 + 2.75 × 10^{-7} × INDUSTRY_5000 + 1.46 × 10^{-4} × TRAFMAJOR	89%	73%	1.8	20	−0.11 (0.83)	16.0 [11.9−30.6]
Gyor, Hungary	23.98 − 1.71 × 10^{-2} × URBGREEN_5000 + 7.52 × 10^{-5} × ROADLENGTH_1000 + 5.90 × 10^{-8} × TRAFMAJORLOAD_500	64%	46%	1.2	20	−0.25 (0.05)	22.6 [20.6−26.2]
Lugano, Switzerland	46.30 + 2.25 × 10^{-4} × HEAVYTRAFLOAD_50 − 0.57 × SQRALT − 6.90 × 10^{-7} × NATURAL_5000	83%	77%	1.1	19	−0.12 (0.10)	17.2 [13.7−22.5]
Turin, Italy	24.90 − 7.03 × 10^{-6} × NATURAL_1000 + 9.40 × 10^{-7} × TRAFMAJORLOAD_50 + 1.63 × 10^{-7} × LDRES_5000	71%	59%	2.0	20	−0.09 (0.45)	29.3 [22.7−36.3]
Rome, Italy	16.08 + 4.56 × 10^{-6} × TRAFLOAD_25 + 3.81 × 10^{-3} × ROADLENGTH_100	71%	60%	1.9	20	0.02 (0.30)	19.8 [14.2−27.0]
Barcelona, Spain	16.21 − 4.08 × 10^{-6} × GREEN_1000 + 2.04 × 10^{-7} × TRAFLOAD_100 + 6.82 × 10^{-3} × INTINVDIST2	83%	71%	2.1	20	0.01 (0.46)	16.3 [8.4−24.4]
Catalunya, Spain	14.88 + 9.91 × 10^{-4} × INTMAJORINVDIST − 3.27 × 10^{-6} × GREEN_1000 + 5.36 × 10^{-7} × PORT_5000	62%	51%	2.4	40	−0.06 (0.38)	15.6 [8.4−24.4]
Athens, Greece	13.98 + 2.04 × 10^{-8} × TRAFLOAD_500 − 1.77 × 10^{-7} × NATURAL_5000 + 0.017 × ROADLENGTH_25 + 1.52 × 10^{-5} × INDUSTRY_300 + 1.80 × 10^{-2} × MAJORROADLENGTH_50	86%	69%	1.7	20	−0.10 (0.30)	20.9 [13.7−25.7]
Heraklion, Greece	12.95 + 0.03 × ROADLENGTH_25 + 9.06 × 10^{-6} × HDRES_300	49%	25%	2.1	20	−0.07 (0.98)	14.7 [11.3−21.0]

[a]See SI SI1 and Table 1 for detailed explanation of the variable names. Some variables are buffers with _X indicating the radius of the buffer in meters. The following predictors were derived for all sites: the surface area (m^2) of high density residential land (HDRES_X), low density residential land (LDRES_X), all residential land (HLDRES_X), industry (INDUSTRY_X), port (PORT_X), urban green space (URBGREEN_X), natural land (NATURAL_X), urban green and natural land combined (GREEN_X), water (WATER_X), the number (N) or surface area (m^2) of buildings (BUILDINGS_X), population (N) (POP_X) or number (N) of households (HHOLD_X), the square root of altitude (SQRALT), a regional concentration estimate ($\mu g/m^3$ or $10^{-5}m^{-1}$), X-coordinate (XCOORD), Y-coordinate (YCOORD), total length (m) of all road and all major road segments (ROADLENGTH_X, MAJORROADLENGTH_X), inverse distance (m^{-1}) and inverse squared distance (m^{-2}) to the nearest road of the central road network (DISTINVNEARC1, DISTINVNEARC2) and the nearest major road in the central network (DISTINVMAJORC1, DISTINVMAJORC2), traffic intensity on the nearest road (TRAFNEAR) and nearest major road (TRAFMAJOR), heavy traffic intensity on the nearest (HEAVYTRAFNEAR) and nearest major road (HEAVYTRAFMAJOR), inverse distance (m^{-1}) and inverse squared distance (m^{-2}) to the nearest road of the local network (DISTINVNEAR1, DISTINVNEAR2) and the nearest major road in the local network (DISTINVMAJOR1, DISTINVMAJOR2), the product of inverse/inverse squared distance to the nearest road and the traffic intensity on this road (vehicles·$day^{-1}m^{-1}$/vehicles·$day^{-1}m^{-2}$) (INTINVDIST, INTINVDIST2), equivalent for major roads (INTMAJORINVDIST, INTMAJORINVDIST2), and for heavy traffic (HEAVYINTINVDIST, HEAVYINTINVDIST2), the sum of (traffic intensity × the length of all road segments) within a buffer (vehicles·day^{-1}·m) for all roads (TRAFLOAD_X), for major roads (TRAFMAJORLOAD_X), for heavy traffic (HEAVYTRAFLOAD_X) and heavy traffic on major roads (HEAVYTRAFMAJORLOAD_X. See SI SI4 for description of distributions of included variables. [b]Number of sites that have been used for model development. Failed measurements explain fewer than 20 sites for Oslo and Lugano. Two sites in Stockholm County and Manchester were excluded from model building, see also SI SI6 for details. [c]Mean [min − max] [d]Local data, SI SI1 and SI2.

Table 2. Description of Developed LUR Models for $PM_{2.5}$ Absorbance, Including Descriptive Statistics of the Measured Concentrations

study area	LUR model[a]	R^2 of model	R^2 validation	RMSE (validation) ($\mu g/m^3$)	number of sites[b]	Moran's I (p-value)	measured concentration (10^{-5} m^{-1})[c]
Oslo, Norway	1.17 + 3.01 × 10^{-12} × TRAFLOAD_1000 + 2.69 × 10^{-3} × MAJORROADLENGTH_50 − 2.40 × 10^{-2} × SQRALT + 3.75 × 10^{-3} × ROADLENGTH_25 − 1.24 × 10^{-6} × NATURAL_300	95%	91%	0.2	20	−0.05 (0.85)	1.3 [0.6–2.1]
Stockholm County, Sweden	0.51 + 5.59 × 10^{-5} × ROADLENGTH_500 + 2.55 × 10^{-6} × HEAVYTRAFLOAD_50 − 8.38 × 10^{-9} × WATER_5000	89%	85%	0.1	19	−0.46 (0.08)	0.8 [0.3–1.3]
Helsinki/Turku, Finland	1.15 + 2.09 × 10^{-7} × TRAFLOAD_50 − 1.15 × 10^{-6} × NATURAL_500[d]	65%	47%	0.3	20	−0.25 (0.07)	1.1 [0.6–2.3]
Copenhagen, Denmark	0.60 + 1.98 × 10^{-4} × MAJORROADLENGTH_300 + 3.56 × 10^{-6} × HHOLD_5000 + 5.64 × 10^{-4} × TRAFNEAR + 2.90 × 10^{-8} × INDUSTRY_5000	92%	86%	0.1	20	−0.06 (0.97)	1.2 [0.7–1.8]
Kaunas, Lithuania	1.32 + 4.24 × 10^{-8} × TRAFLOAD_50 + 1.31 × 10^{-6} × LDRES_300 + 5.77 × 10^{-4} × POP_100 + 6.62 × 10^{-6} × TRAFMAJOR	87%	69%	0.2	20	−0.05 (0.30)	2.0 [1.4–3.0]
Manchester, UK	1.31 + 1.22 × 10^{-3} × MAJORROADLENGTH_100 − 8.70 × 10^{-9} × NATURAL_5000 − 2.38 × 10^{-7} × URBGREEN_1000 + 3.06 × 10^{-4} × ROADLENGTH_100	91%	81%	0.1	19	−0.09 (0.45)	1.3 [0.9–2.1]
London/Oxford, UK	0.81 + 1.12 × 10^{-7} × HEAVYTRAFLOAD_500 + 8.00 × 10^{-9} × HLDRES_5000[d] + 125.41 × DISTINVMAJORC2	96%	92%	0.2	20	−0.21 (0.16)	1.6 [0.9–4.7]
Netherlands/Belgium	0.07 + 2.95 × 10^{-9} × TRAFLOAD_500 + 2.93 × 10^{-3} × MAJORROADLENGTH_50 + 0.85 × REGIONALESTIMATE + 7.90 × 10^{-9} × HLDRES_5000 + 1.72 × 10^{-6} × HEAVYTRAFLOAD_50	92%	89%	0.2	40	−0.16 (0.42)	1.7 [0.9–3.0]
Ruhr Area, Germany	0.97 + 1.80 × 10^{-6} × HEAVYTRAFLOAD_100 + 2.31 × 10^{-8} × HEAVYTRAFLOAD_100_1000 + 1.64 × 10^{-8} × INDUSTRY_5000 + 2.21 × 10^{-5} × POP_1000	97%	95%	0.1	20	−0.02 (0.65)	1.6 [1.0–2.6]
Munich/Augsburg, Germany	1.34 + 1.77 × 10^{-7} × TRAFLOAD_50 + 1.84 × 10^{-3} × ROADLENGTH_50 + 2.16 × 10^{-4} × TRAFMAJORLOAD_1000[d]	91%	82%	0.2	20	−0.19 (0.21)	1.9 [1.4–2.8]
Vorarlberg, Austria	0.84 + 6.86 × 10^{-7} × TRAFLOAD_25 + 3.14 × 10^{-3} × BUILDINGS_300 + 2.12 × 10^{-8} × HLDRES_5000	81%	73%	0.2	20	−0.04 (0.89)	1.8 [1.1–2.4]
Paris, France	0.94 + 7.98 × 10^{-8} × INDUSTRY_5000 + 2.36 × 10^{-7} × HDRES_1000 + 1.68 × 10^{-7} × TRAFMAJORLOAD_100	91%	81%	0.4	20	−0.16 (0.97)	2.0 [0.8–5.1]
Gyor, Hungary	1.55 + 1.54 × 10^{-4} × MAJORROADLENGTH_300 + 2.30 × 10^{-5} × ROADLENGTH_500 + 7.03 × 10^{-8} × TRAFLOAD_100 + 18.18 × DISTINVMAJORC2	80%	66%	0.2	20	−0.15 (0.34)	1.9 [1.5–2.8]
Lugano, Switzerland	2.77 + 1.08 × 10^{-4} × ROADLENGTH_300 + 3.38 × 10^{-5} × HEAVYTRAFLOAD_50 − 0.10 × SQRALT	79%	71%	0.3	19	−0.13 (0.09)	2.0 [1.2–3.0]
Turin, Italy	1.74 + 1.29 × 10^{-3} × MAJORROADLENGTH_100 + 2.43 × 10^{-5} × HLDRES_100 + 2.47 × 10^{-9} × TRAFLOAD_1000 − 1.01 × 10^{-6} × NATURAL_1000	88%	81%	0.3	20	−0.06 (0.82)	3.0 [1.6 − 4.2]
Rome, Italy	2.14 + 5.64 × 10^{-4} × INTMAJORINVDIST + 7.52 × 10^{-7} × POP_5000	84%	79%	0.3	20	0.02 (0.26)	2.9 [1.9–4.7]
Barcelona, Spain	1.01 + 7.46 × 10^{-6} × HDRES_300 + 2.66 × 10^{-3} × INTINVDIST2 + 1.11 × 10^{-7} × TRAFLOAD_50	86%	80%	0.4	20	−0.01 (0.64)	2.7 [0.9–4.9]
Catalunya, Spain	1.20 + 2.22 × 10^{-4} × INTMAJORINVDIST + 1.92 × 10^{-5} × ROADLENGTH_1000 − 1.70 × 10^{-8} × NATURAL_5000 + 3.74 × DISTINVMAJOR1	75%	69%	0.5	40	0.02 (0.68)	2.5 [0.9–4.9]
Athens, Greece	0.62 + 9.67 × 10^{-8} × TRAFMAJORLOAD_25 + 7.58 × 10^{-5} × ROADLENGTH_300 + 1.22 × 10^{-8} × HDRES_5000 + 1.59 × 10^{-9} × TRAFLOAD_500 + 0.002 × MAJORROADLENGTH_50	93%	81%	0.2	20	−0.16 (0.11)	2.4 [1.1–3.5]
Heraklion, Greece	0.77 + 1.22 × 10^{-3} × POP_100 + 8.43 × 10^{-5} × MAJORROADLENGTH_300	56%	40%	0.3	20	−0.97 (0.10)	1.2 [0.7–2.1]

[a]See note below Table 1 for explanation of the variable names. See SI Table 1 and SI1 for details. See SI SI4 for description of distributions of included variables. [b]Number of sites that have been used for model development. Failed measurements explain fewer than 20 sites for Lugano. Two sites in Stockholm County and Manchester were excluded from model building, see also SI SI6 for details. [c]Mean [min − max]. [d]Local data, SI SI1 and SI2.

Table 3. Description of Developed LUR Models for PM_{coarse}, Including Descriptive Statistics of the Measured Concentrations

study area	LUR model[a]	R^2 of model	R^2 validation	RMSE (validation) ($\mu g/m^3$)	number of sites[b]	Moran's I (p-value)	measured concentration ($\mu g/m^3$)[c]
Oslo, Norway	8.68 + 0.24 × MAJORROADLENGTH_25 − 0.37 × SQRALT	68%	53%	3.4	19	−0.06 (0.28)	6.5 [0−16.1]
Stockholm County, Sweden	0.70 + 4.32 × 10^{-6} × TRAFLOAD_50 + 2.73 × 10^{-5} × HLDRES_300	72%	65%	3.5	19	−0.06 (0.42)	10.0 [1.3−25.1]
Helsinki/Turku, Finland	3.82 + 2.52 × 10^{-2} × HHOLD_100 + 1.39 × 10^{-6} × TRAFLOAD_50	61%	33%	2.8	20	−0.36 (0.01)	6.2 [1.8−17.9]
Copenhagen, Denmark	6.01 + 1.84 × 10^{-7} × PORT_5000 − 4.78 × 10^{-8} × GREEN_5000[d] + 5.22 × 10^{-3} × MAJORROADLENGTH_50	71%	54%	1.0	20	−0.17 (0.12)	6.0 [2.1−8.7]
Kaunas, Lithuania	5.39 + 3.34 × 10^{-8} × TRAFMAJORLOAD_100 + 9.85 × 10^{-5} × LDRES_100	32%	3%	2.6	20	−0.05 (0.42)	8.4 [3.6−13.7]
Manchester, UK	6.46 − 1.55 × 10^{-6} × URBGREEN_1000 + 1.85 × 10^{-7} × INDUSTRY_5000 + 3.51 × 10^{-5} × HDRES_100 + 1.59 × 10^{-8} × HEAVYTRAFLOAD_1000	79%	56%	1.0	19	−0.03 (0.48)	7.6 [4.3−11.3]
London/Oxford, UK	5.36 + 33.08*DISTINVMAJOR1 + 7.98 × 10^{-4} × HEAVYTRAFMAJOR	68%	57%	1.3	20	−0.17 (0.29)	7.4 [4.4−10.3]
Netherlands/Belgium	7.59 + 5.02 × 10^{-9} × TRAFLOAD_1000 + 1.38 × 10^{-7}*PORT_5000 + 5.38 × 10^{-5} × TRAFNEAR	51%	38%	1.7	40	−0.08 (0.75)	9.3 [6.4−15.0]
Ruhr Area, Germany	7.42 + 2.14 × 10^{-4} × MAJORROADLENGTH_1000 − 2.42 × 10^{-5} × URBGREEN_300 + 1.36 × 10^{-7} × INDUSTRY_5000	66%	57%	1.2	20	−0.02 (0.73)	9.4 [7.1−12.8]
Munich/Augsburg, Germany	4.09 + 2.46 × 10^{-2} × MAJORROADLENGTH_50[d] + 4.20 × 10^{-6} × POP_5000 + 1.16 × 10^{-2} × ROADLENGTH_50[d]	81%	69%	1.6	20	−0.15 (0.36)	7.7 [4.9−15.9]
Vorarlberg, Austria	6.08 + 2.98 × 10^{-6} × TRAFLOAD_25 − 8.61 × 10^{-8} × NATURAL_5000 + 4.52 × 10^{-4} × ROADLENGTH_300	53%	31%	1.5	20	−0.11 (0.46)	7.3 [4.7−10.8]
Paris, France	7.54 + 7.54 × 10^{-4} × MAJORROADLENGTH_500 + 8.99 × 10^{-7} × TRAFLOAD_50 − 1.66 × 10^{-7} × NATURAL_5000	81%	73%	4.6	20	−0.08 (0.82)	9.6 [3.9−21.8]
Gyor, Hungary	7.27 + 9.23 × 10^{-3} × MAJORROADLENGTH_100	50%	49%	1.4	20	−0.14 (0.36)	8.0 [5.0−12.2]
Lugano, Switzerland	3.87 + 9.76 × 10^{-7} × TRAFLOAD_50 + 8.34 × 10^{-9} × TRAFLOAD_50_1000 + 2.29 × 10^{-6} × HDRES_500	77%	65%	1.1	18	−0.12 (0.18)	6.8 [3.8−9.9]
Turin, Italy	11.83 + 1.35 × 10^{-3} × POP_300 − 2.39 × 10^{-7} × NATURAL_5000	65%	58%	2.4	20	−0.10 (0.30)	13.8 [7.5−21.5]
Rome, Italy	7.77 + 6.34 × 10^{-6} × TRAFLOAD_25 + 1.74 × 10^{-2} × ROADLENGTH_50 + 8.68 × 10^{-5} × ROADLENGTH_50_1000	70%	57%	3.7	20	0.01 (0.28)	17.2 [11.3−32.0]
Barcelona, Spain	25.63 − 0.73 × SQRALT + 4.74 × 10^{-7} × TRAFMAJORLOAD_50	75%	70%	2.3	20	−0.09 (0.61)	21.0 [9.4−26.0]
Catalunya, Spain	38.42 − 2.33 × 10^{-7} × GREEN_5000 + 0.15 × MAJORROADLENGTH_25 + 1.95 × 10^{-6} × HDRES_1000 − 0.43 × SQRALT	76%	71%	4.1	40	−0.01 (0.70)	20.0 [9.4−30.3]
Athens, Greece	14.75 + 1.12 × 10^{-7} × TRAFMAJORLOAD_100 + 0.006 × ROADLENGTH_100	44%	23%	4.4	20	−0.20 (0.05)	21.9 [13.5−33.7]
Heraklion, Greece	18.27 + 108.67 × DISTINVMAJORC2 + 5.91 × 10^{-3} × ROADLENGTH_100	63%	56%	3.5	20	−0.38 (0.54)	23.6 [14.2−38.8]

[a]See note below Table 1 for explanation of the variable names note. See SI Table 1 and SI1 for details. See SI SI4 for description of distributions of included variables. [b]Number of sites that have been used for model development. Failed measurements explain fewer than 20 sites for Oslo and Lugano. Two sites in Stockholm County and Manchester were excluded from model building, see also SI SI6 for details. [c]Mean [min − max]. [d]Local data, SI SI1 and SI2.

Turin, traffic variables were included in the model, probably reflecting resuspended road dust from tire and break wear. In six models only traffic variables were included.

PM_{10} Models. Median model R^2 for PM_{10} was 77%, ranging from 50% (Kaunas) to 90% (Barcelona, London/Oxford). Differences between model R^2 and LOOCV R^2 were generally below 10% (SI SI3, Table 1). PM_{10} models included two to four predictors. Explained variance was slightly higher than for $PM_{2.5}$.

We found no evidence of spatial autocorrelation in the residuals as for the large majority of models, Moran's I was not significant ($p > 0.05$). The included predictor variables accounted sufficiently for the spatial variation within each area. For the few significant values, the Moran's I coefficients were negative and small.

4. DISCUSSION

LUR models were developed with moderate to good explained variance for three size fractions of particulate matter and $PM_{2.5}$ absorbance. Explained variance of the LUR models was highest for $PM_{2.5}$ absorbance (median 89%), followed by PM_{10} (77%), and was lower for $PM_{2.5}$ (71%) and PM_{coarse} (68%). Variability in R^2 was partly related to limited concentration contrast and availability of predictor data, in particular traffic intensity. During model development we encountered challenges including influential observations related to extreme concentrations or extreme predictor variables at a single site and predictor variables with a large fraction of zero values. ESCAPE adds significantly to a still small number of LUR models for particulate matter air pollution.

Comparison Across Particle Fractions. Model performance for $PM_{2.5}$ absorbance was higher than for $PM_{2.5}$, which was also found ina limited number of previous studies.[7] It likely reflects the major impact of motorized road traffic on PM absorbance as a marker of black carbon and the larger long-range transport component to $PM_{2.5}$.[3] In Europe, a substantial fraction of private cars and most middle and heavy duty vehicles use diesel. The lower model performance for coarse particles agrees with a study in Amsterdam, which reported model R^2 of 37% for PM_{coarse}, 76% for $PM_{2.5}$ absorbance and 57% for $PM_{2.5}$.[17] It is likely that the lack of specific GIS data on local coarse particle sources has contributed to the lower model performance, such as the wear of different road surface materials. PM_{coarse} is calculated by subtracting PM_{10} and $PM_{2.5}$ mass concentrations, so the precision of these measurements is lower than for the other components.

Selected Predictor Variables. Although all study areas used the same procedures for developing LUR models, final models included different variables. Most models included local in addition to centrally available variables. One or more traffic variables were selected in almost all models, reflecting the major impact of traffic emissions and the overrepresentation of traffic sites in our site selection. Buffer sizes of 50 m were most common, but also the largest buffer size of 1000 m was included in many models. The 25 m buffer size did not enter models frequently, likely because of limitations in accuracy of GIS road networks. Therefore, 25 m buffers were not considered in some areas with insufficient road network accuracy. This buffer size does not represent the impact of larger ring roads or motorways, but instead represents the impact of moderately busy inner-city streets where homes are close to the road. Factors related to population density and residential land-use were selected in many models, representing pollution from various activities including home heating. All buffer sizes appeared, but larger buffers (1000 and 5000 m) were most common for $PM_{2.5}$ absorbance. Urban green and natural land use were selected particularly in $PM_{2.5}$ and PM_{10} models, often in large buffer sizes (1000 or 5000 m). In areas with major ports (Netherlands and Belgium, Catalunya) and industrial areas, port and industrial land use were represented in the LUR models. As CORINE does not distinguish type of industry, these models can only be applied in the area where they were developed.[20] We did not include industry in models if local expertise indicated that there was no industry with significant emissions present. Altitude was present in models for Lugano, Barcelona and Catalunya, study areas which included a large variation in altitude among the selected sites.

Area-Specific versus Combined LUR Models. There were differences in available predictors between study areas. Using only central variables would have resulted in poorer models, as, for example, indicated by the poorer models in areas without local traffic intensity data. A study by Vienneau et al. evaluated a pooled LUR model for Great Britain and The Netherlands, but concluded that there was no benefit in forcing a model based on common variables compared to country-specific models.[21] Studies on the transferability of LUR models suggest that they are best developed locally, and perform less well when applied to other areas.[6,7,11] To evaluate the merits of combining study areas, we developed LUR models for the combined areas of London/Oxford, The Netherlands/Belgium and the Ruhr Area. We found high R^2's for $PM_{2.5}$, $PM_{2.5}$ absorbance and PM_{10}, and a moderately high R^2 for PM_{coarse}, (SI SI5). Indicator variables for study area were important predictors in all models, representing the large between-area variance.[19] We found significant differences in the effect of the traffic variable across the study areas for $PM_{2.5}$ and $PM_{2.5}$ absorbance. This suggests there are true differences between parameter estimates for different areas, supporting the ESCAPE strategy to develop separate models for each area. Furthermore, in the combined-area model for $PM_{2.5}$, the well-established regional background variation in The Netherlands was not included, indicating that we may miss predictors which are only of local importance. However, without external validation data it is not possible to conclude which approach is more valid.

Modeling Experiences. In LUR papers generally little detail is presented on model diagnostics. We experienced that especially the use of the Cook's D influence statistic to identify influential observations was useful. Influential observations were due to extreme concentrations at a single site, extreme predictor variables at a single site and predictor variables with a large fraction of zero values. The latter two problems were resolved by not offering these variables as predictor variables. In Stockholm County and Manchester, we excluded a site with extreme concentrations from model development. A site was only removed from the modeling procedure if (1) the site was very influential (e.g., model parameters changed heavily, that is lost significance, changed direction depending on whether that site was included or excluded), (2) all possible models identified this site as very influential, and (3) both the local partners and the ESCAPE exposure working group agreed that in retrospect, the site was selected at a location which was not representative for residential exposures. If a site was considered unsuitable, it was removed from the modeling procedure of all components. A more detailed description of the sites which were removed from the modeling procedure is given in SI SI6. Only two sites (out of a total of 440) were excluded because of the above criteria. We underline the importance of a careful selection of measurement sites, representative for the outdoor concentrations at the home addresses. Exclusion of sites was considered defensible as the goal is to develop stable models that can be applied at residential addresses.

Cross Validation. The performance of the models was evaluated using leave-one out cross validation, as in previous LUR studies.[21,25,26] The limited number of PM monitoring sites made it infeasible to set aside a sufficiently large part of the data set for hold-out validation. Two recent studies documented that LUR models based on a limited number of training sites perform well in leave-one-out cross-validation, but do worse in hold-out validation, using an independent external data set.[27,28] This is explained by a risk of overfitting when evaluating a large number of predictor variables to explain concentrations at relatively few sites.[29] While a minimum of 40–80[7], or 80 sites has been suggested,[27] there are more examples of LUR studies on fewer sites, especially for PM.[12,24] Most ESCAPE models are based on 20 sites, which may limit the robustness of the models. We further expect that the presented cross-validation R^2's of the models are an overestimation of the hold-out validation R^2's. We tried to minimize the problem of over fitting by a priori choices in model development.

Variable Selection. Within ESCAPE, we used a supervised stepwise selection of variables for the LUR models. This approach has been frequently used in previously published LUR models[7], and was motivated by our preference to select models which were plausible (direction of effect as defined a priori) and stable (coefficients not dependent on a single observation). We evaluated a large number of predictors, which were often highly correlated. While we selected the model with the highest adjusted R^2, we emphasize that there is no single absolute LUR model for a certain area. Different models with similar performance in explaining spatial variability in concentrations can be developed.

Other techniques for the selection of predictor variables are used in which combinations of candidate predictor variables are evaluated based on their performance in cross-validation.[30] This deletion/substitution/addition algorithm aims to maximize the cross-validation R^2, rather than optimizing the adjusted model R^2 in a stepwise manner.[30]

Su et al. also developed a method for selecting an optimal buffer size for each candidate predictor, thereby limiting the number of predictors considered for the LUR model.[31] Few comparisons have been made of the performance of different selection procedures. A recent comparison of the performance of the ESCAPE procedure, a procedure based upon maximizing the cross-validation R^2 and the deletion-substitution algorithm found very small differences in hold-out validation R^2.[27]

We considered a wide range of buffer sizes, based on known dispersion patterns, but little is known about buffers larger than 5000 m. Su et al. found no larger buffers than 3000 m, but the maximum distance of influence remains hard to define[31]. In large study areas including several metropolitan areas there may be merit to evaluate larger buffers.

Previous Studies in ESCAPE Areas. LUR models have previously been published for $PM_{2.5}$ and $PM_{2.5}$ absorbance for The Netherlands, Munich, Stockholm County, and Ruhr area.[25,26] $PM_{2.5}$ models were comparable in model explained variance for The Netherlands (73% in 2003 against 67% in this paper), while they improved for Munich (56–78%), Stockholm County (50–94%), and drastically for the Ruhr area (17–88%). $PM_{2.5}$ absorbance models improved for all areas, from 81% to 92% for The Netherlands, from 67% to 91% for Munich, from 66% to 90% for Stockholm County, and 82 to 97% for the Ruhr area. The improvements are likely explained by better available predictor variables, but might have also been affected by the selection of sites, which included more street sites in ESCAPE. In the past decade more and better GIS data sets have become available to derive potential predictor variables.[32] The resolution of GIS data sets has increased, traffic intensity data are linked to digital road networks, and some digital data are available through open sources.

Limitations. The main limitation is the restricted number of 20 monitoring sites available for the development of PM models. Although there are no strict rules for a minimum required number of sites, only few purpose-designed sampling networks including particulate matter have included 40 sites or more. LUR models have been developed successfully for studies including fewer measurement sites.[11] Because of the large number of study areas included in the ESCAPE project, twenty PM sites per study area was the maximum feasible. As discussed before, the risk of overfitting is greater when using smaller training sets in model building.[27,28] Our measurement campaign was restricted temporally, as previous PM sampling campaigns.[7] This issue was addressed by temporal adjustment using a continuous reference site where measurements were made for a 12-month period.

The ESCAPE project modeled long-term air pollution concentrations at the home and/or school address of cohort study subjects. The modeled individual concentrations do not take account of time activity patterns and indoor/outdoor differences and are therefore not equal to personal exposure.

ESCAPE measurements took place between October 2008 and April 2011, while resulting LUR models will be applied to estimate long-term exposure of cohorts recruited generally in the mid 1990s. There is limited evidence that LUR models can accurately estimate spatial pollution contrasts 10 years back in time.[33,34] A separate paper will address this issue in a subset of the ESCAPE study areas.

Information on the fraction of heavy traffic, average speed, and street configuration (e.g., canyons) was unavailable for many study areas. Several European data sets were somewhat older (2000 for land use and 2001 for population density). However, changes in land cover between 2000 and 2006 were small (1.24% of total surface area changed classes).[35]

■ ASSOCIATED CONTENT

Supporting Information

Supporting Information is available on (1) the derivation of predictor variables, (2) description of local GIS variables, (3) description of PM10 LUR models, (4) distribution of the predictor variables, (5) the development of combined LUR models for London/Oxford, The Netherlands/Belgium and Ruhr Area and (6) description of excluded sites. This material is available free of charge via the Internet at http://pubs.acs.org.

■ AUTHOR INFORMATION

Corresponding Author

*Phone: +31 30 253 94 74; fax: +31 30 253 94 99; e-mail: m.r.eeftens@uu.nl.

Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

The research leading to these results has received funding from the European Community's Seventh Framework Program (FP7/2007-2011) under grant agreement number: 211250. We thank all those who have been involved in exposure monitoring and modeling: Sònia Alvarez, Christophe Ampe, Chiara Badaloni, Laura Bouso, Simone Bucci, Glòria Carrasco, Francesco

Forastiere, Erwan Gremaud, Sally Liu, Danny Houthuijs, Nino Künzli, Jaume Matamala, Kees Meliefste, Marieke Oldenwening, Patrizio Pasquinelli, Meritxell Portella, Jordi Sunyer, Anna Ripoll, Marjan Tewis, Wim Swart, and Meng Wang.

ABBREVIATIONS:

CORINE: coordination of information on the environment
ESCAPE: European Study of Cohorts for Air Pollution Effects
GIS: Geographic information systems
LOOCV: leave one out cross validation
LUR: land use regression
NO_2: nitrogen dioxide
NO_X: nitrogen oxides
$PM_{2.5}$: mass concentration of particles less than 2.5 μm in size
PM_{10}: mass concentration of particles less than 10 μm in size
$PM_{2.5}$ absorbance: measurement of the blackness of $PM_{2.5}$ filters; a proxy for elemental carbon, which is the dominant light absorbing substance
PM_{coarse}: mass concentration of particles between 2.5 and 10 μm in size

REFERENCES

(1) Pope, C. A., 3rd; Dockery, D. W. Health effects of fine particulate air pollution: lines that connect. *J. Air Waste Manag. Assoc.* **2006**, *56*, 709−742.

(2) Brunekreef, B.; Holgate, S. T. Air pollution and health. *Lancet* **2002**, *360*, 1233−1242.

(3) HEI. *Report 17: Traffic-related Air Pollution: a Critical Review of the Literature on Emissions, Exposure, and Health Effects*; Health Effects Institute, 2010.

(4) Monn, C. Exposure assessment of air pollutants: a review on spatial heterogeneity and indoor/outdoor/personal exposure to suspended particulate matter, nitrogen dioxide and ozone. *Atmos. Environ.* **2001**, *35*, 1−32.

(5) Lebret, E.; Briggs, D.; van Reeuwijk, H.; Fischer, P.; Smallbone, K.; Harssema, H.; Kriz, B.; Gorynski, P.; Elliott, P. Small area variations in ambient NO_2 concentrations in four European areas. *Atmos. Environ.* **2000**, *34*, 177−185.

(6) Jerrett, M.; Arain, A.; Kanaroglou, P.; Beckerman, B.; Potoglou, D.; Sahsuvaroglu, T.; Morrison, J.; Giovis, C. A review and evaluation of intraurban air pollution exposure models. *J. Expo. Anal. Environ. Epidemiol.* **2005**, *15*, 185−204.

(7) Hoek, G.; Beelen, R.; de Hoogh, K.; Vienneau, D.; Gulliver, J.; Fischer, P.; Briggs, D. A review of land-use regression models to assess spatial variation of outdoor air pollution. *Atmos. Environ.* **2008**, *42*, 7561−7578.

(8) Hoek, G.; Meliefste, K.; Cyrys, J.; Lewné, M.; Bellander, T.; Brauer, M.; Fischer, P.; Gehring, U.; Heinrich, J.; van Vliet, P.; Brunekreef, B. Spatial variability of fine particle concentrations in three European areas. *Atmos. Environ.* **2002**, *36*, 4077−4088.

(9) Jerrett, M.; Burnett, R. T.; Ma, R.; Pope, C. A., 3rd; Krewski, D.; Newbold, K. B.; Thurston, G.; Shi, Y.; Finkelstein, N.; Calle, E. E.; Thun, M. J. Spatial analysis of air pollution and mortality in Los Angeles. *Epidemiology* **2005**, *16*, 727−736.

(10) Cohen, M. A.; Adar, S. D.; Allen, R. W.; Avol, E.; Curl, C. L.; Gould, T.; Hardie, D.; Ho, A.; Kinney, P.; Larson, T. V.; Sampson, P.; Sheppard, L.; Stukovsky, K. D.; Swan, S. S.; Liu, L. J.; Kaufman, J. D. Approach to estimating participant pollutant exposures in the multi-ethnic study of atherosclerosis and air pollution (MESA Air). *Environ. Sci. Technol.* **2009**, *43*, 4687−4693.

(11) Ryan, P. H.; LeMasters, G. K. A review of land-use regression models for characterizing intraurban air pollution exposure. *Inhal. Toxicol.* **2007**, *19* (Suppl 1), 127−133.

(12) Henderson, S.; Beckerman, B.; Jerrett, M.; Brauer, M. Application of land use regression to estimate long-term concentrations of traffic-related nitrogen oxides and fine particulate matter. *Environ. Sci. Technol.* **2007**, *41*, 2422−2428.

(13) Beelen, R.; Hoek, G.; van den Brandt, P. A.; Goldbohm, R. A.; Fischer, P.; Schouten, L. J.; Armstrong, B.; Brunekreef, B. Long-term exposure to traffic-related air pollution and lung cancer risk. *Epidemiology* **2008**, *19*, 702−710.

(14) Pope, C. A., 3rd; Burnett, R. T.; Thun, M. J.; Calle, E. E.; Krewski, D.; Ito, K.; Thurston, G. D. Lung cancer, cardiopulmonary mortality, and long-term exposure to fine particulate air pollution. *JAMA, J. Am. Med. Assoc.* **2002**, *287*, 1132−1141.

(15) Sarnat, J. A.; Schwartz, J.; Catalano, P. J.; Suh, H. H. Gaseous pollutants in particulate matter epidemiology: confounders or surrogates? *Environ. Health Perspect.* **2001**, *109*, 1053−1061.

(16) European Environment Agency (EEA). *Spatial Assessment of PM_{10} and Ozone Concentrations in Europe (2005)*; EEA Technical report No 1, **2009**.

(17) Hoek, G.; Beelen, R.; Kos, G.; Dijkema, M.; van der Zee, S. C.; Fischer, P. H.; Brunekreef, B. Land use regression model for ultrafine particles in Amsterdam. *Environ. Sci. Technol.* **2011**, *45*, 622−628.

(18) Brunekreef, B.; Forsberg, B. Epidemiological evidence of effects of coarse airborne particles on health. *Eur. Respir. J.* **2005**, *26*, 309−318.

(19) Eeftens, M.; Tsai, M.; Ampe, C.; Anwander, B.; Beelen, R.; Bellander, T.; Cesaroni, G.; Cirach, M.; Cyrys, J.; de Hoogh, K.; De Nazelle, A.; de Vocht, F.; Declercq, C.; Dėdelė, A.; Eriksen, K.; Galassi, C.; Gražulevičienė, R.; Grivas, G.; Heinrich, J.; Hoffmann, B.; Iakovides, M.; Ineichen, A.; Katsouyanni, K.; Korek, M.; Krämer, U.; Kuhlbusch, T.; Lanki, T.; Madsen, C.; Meliefste, K.; Mölter, A.; Mosler, G.; Nieuwenhuijsen, M.; Oldenwening, M.; Pennanen, A.; Probst-Hensch, N.; Quass, U.; Raaschou-Nielsen, O.; Ranzi, A.; Stephanou, E.; Sugiri, D.; Udvardy, O.; Vaskövi, É.; Weinmayr, G.; Bert Brunekreef, B.; Hoek, G. Variation of $PM_{2.5}$, PM_{10}, $PM_{2.5}$ absorbance and PM_{coarse} concentrations between and within 20 European study areas − results of the ESCAPE project. *Atmos. Environ.* **2012**, accepted.

(20) Beelen, R.; Voogt, M.; Duyzer, J.; Zandveld, P.; Hoek, G. Comparison of the performances of land use regression modelling and dispersion modelling in estimating small-scale variations in long-term air pollution concentrations in a Dutch urban area. *Atmos. Environ.* **2010**, *44*, 4614−4621.

(21) Vienneau, D.; de Hoogh, K.; Beelen, R.; Fischer, P.; Hoek, G.; Briggs, D. Comparison of land-use regression models between Great Britain and the Netherlands. *Atmos. Environ.* **2010**, *44*, 688−696.

(22) Briggs, D. J.; Gulliver, J.; Fecht, D.; Vienneau, D. M. Dasymetric modelling of small-area population distribution using land cover and light emissions data. *Remote Sens. Environ.* **2007**, *108*, 451−466.

(23) Gallego, F. J. A population density grid of the European Union, Population and Environment. *Popul. Environ.* **2010**, *31*, 460−473.

(24) Beelen, R.; Hoek, G.; Fischer, P.; Brandt, P.A.v.d.; Brunekreef, B. Estimated long-term outdoor air pollution concentrations in a cohort study. *Atmos. Environ.* **2007**, *41*, 1343−1358.

(25) Brauer, M.; Hoek, G.; Van Vliet, P.; Meliefste, K.; Fischer, P.; Gehring, U.; Heinrich, J.; Cyrys, J.; Bellander, T.; Lewne, M.; Brunekreef, B. Estimating lonq-term average particulate air pollution concentrations: Application of traffic indicators and geographic information systems. *Epidemiology* **2003**, *14*, 228−239.

(26) Hochadel, M.; Heinrich, J.; Gehring, U.; Morgenstern, V.; Kuhlbusch, T.; Link, E.; Wichmann, H.-.; Krämer, U. Predicting long-term average concentrations of traffic-related air pollutants using GIS-based information. *Atmos. Environ.* **2006**, *40*, 542−553.

(27) Basagaña, X.; Rivera, M.; Aguileraa, I.; Agis, D.; Bouso, L.; Elosua, R.; Foraster, M.; de Nazelle, A.; Nieuwenhuijsen, M.; Vila, J.; Künzli, N. Effect of the number of measurement sites on land use regression models in estimating local air pollution. *Atmos. Environ.* **2012**, DOI: http://dx.doi.org/10.1016/j.atmosenv.2012.01.064.

(28) Wang, M.; Beelen, R.; Eeftens, M.; Meliefste, K.; Hoek, G.; Brunekreef, B. A systematic validation of land use regression models for NO_2. *Environ. Sci. Technol.* **2012**, *46*, 4481−4489.

(29) Babyak, M. A. What you see may not be what you get: a brief, nontechnical introduction to overfitting in regression-type models. *Psychosom. Med.* **2004**, *66*, 411−421.

(30) Sinisi, S. van der Laan, M. Loss-Based Cross-Validated Deletion/Substitution/Addition Algorithms in Estimation, 2004. http://biostats.bepress.com/cgi/viewcontent.cgi?article=1142&context=ucbbiostat (accessed September 25, 2012).

(31) Su, J. G.; Jerrett, M.; Beckerman, B. A distance-decay variable selection strategy for land use regression modeling of ambient air pollution exposures. *Sci. Total Environ.* **2009**, *407*, 3890−3898.

(32) Briggs, D. J. The use of GIS to evaluate traffic-related pollution. *Occup. Environ. Med.* **2007**, *64*, 1−2.

(33) Eeftens, M.; Beelen, R.; Fischer, P.; Brunekreef, B.; Meliefste, K.; Hoek, G. Stability of measured and modelled spatial contrasts in NO_2 over time. *Occup. Environ. Med.* **2011**, *68*, 765−770.

(34) Cesaroni, G.; Porta, D.; Badaloni, C.; Stafoggia, M.; Eeftens, M.; Meliefste, K.; Forastiere, F. Nitrogen dioxide levels estimated from land use regression models several years apart and association with mortality in a large cohort study. *Environ. Health* **2012**, *11*, 48−57.

(35) European Environment Agency. Implementation and chievements of CLC2006. European Environment Agency (EEA). http://www.eea.europa.eu/data-and-maps/data/corine-land-cover-2006-raster-2/clc-final-report/clc-final-report/at_download/file (accessed September 25, 2012).