

Performance of Multi-City Land Use Regression Models for Nitrogen Dioxide and Fine Particles

**Meng Wang, Rob Beelen, Tom Bellander, Matthias Birk,
Giulia Cesaroni, Marta Cirach, Josef Cyrus, Kees de Hoogh,
Christophe Declercq, Konstantina Dimakopoulou,
Marloes Eeftens, Kirsten T. Eriksen, Francesco Forastiere,
Claudia Galassi, Georgios Grivas, Joachim Heinrich,
Barbara Hoffmann, Alex Ineichen, Michal Korek, Timo Lanki,
Sarah Lindley, Lars Modig, Anna Mölter, Per Nafstad,
Mark J. Nieuwenhuijsen, Wenche Nystad, David Olsson,
Ole Raaschou-Nielsen, Martina Ragettli, Andrea Ranzi,
Morgane Stempfelet, Dorothea Sugiri, Ming-Yi Tsai,
Orsolya Udvardy, Mihaly J. Varró, Danielle Vienneau,
Gudrun Weinmayr, Kathrin Wolf, Tarja Yli-Tuomi, Gerard Hoek,
and Bert Brunekreef**

<http://dx.doi.org/10.1289/ehp.1307271>

Received: 24 June 2013

Accepted: 30 April 2014

Advance Publication: 2 May 2014

Performance of Multi-City Land Use Regression Models for Nitrogen Dioxide and Fine Particles

Meng Wang,¹ Rob Beelen,¹ Tom Bellander,² Matthias Birk,³ Giulia Cesaroni,⁴ Marta Cirach,⁵ Josef Cyrus,^{6,7} Kees de Hoogh,⁸ Christophe Declercq,⁹ Konstantina Dimakopoulou,¹⁰ Marloes Eeftens,¹ Kirsten T. Eriksen,¹¹ Francesco Forastiere,⁴ Claudia Galassi,¹² Georgios Grivas,¹³ Joachim Heinrich,³ Barbara Hoffmann,¹⁴ Alex Ineichen,¹⁵ Michal Korek,² Timo Lanki,¹⁶ Sarah Lindley,¹⁷ Lars Modig,¹⁸ Anna Mölter,¹⁹ Per Nafstad,^{20,21} Mark J. Nieuwenhuijsen,⁵ Wenche Nystad,²¹ David Olsson,¹⁸ Ole Raaschou-Nielsen,¹¹ Martina Ragettli,¹⁵ Andrea Ranzi,¹² Morgane Stempfelet,⁹ Dorothea Sugiri,¹⁴ Ming-Yi Tsai,^{15,22,23} Orsolya Udvardy,²⁴ Mihaly J. Varró,²⁴ Danielle Vienneau,^{8,15,22} Gudrun Weinmayr,²⁵ Kathrin Wolf,⁶ Tarja Yli-Tuomi,¹⁶ Gerard Hoek,¹ and Bert Brunekreef^{1,26}

¹Institute for Risk Assessment Sciences, Utrecht University, Utrecht, the Netherlands; ²Institute of Environmental Medicine, Karolinska Institutet, Stockholm, Sweden; ³Institute of Epidemiology I, Helmholtz Zentrum München, German Research Center for Environmental Health, Neuherberg, Germany; ⁴Epidemiology Department, Lazio Regional Health Service, Rome, Italy; ⁵Center for Research in Environmental Epidemiology (CREAL), Barcelona, Spain; ⁶Institute of Epidemiology II, Helmholtz Zentrum München, German Research Center for Environmental Health, Neuherberg, Germany; ⁷University of Augsburg, Environmental Science Center, Augsburg, Germany; ⁸MRC-PHE Centre for Environment and Health, Department of

Epidemiology and Biostatistics, Imperial College London, London, United Kingdom; ⁹French Institute for Public Health Surveillance, Saint-Maurice, France; ¹⁰Department of Hygiene, Epidemiology and Medical Statistics, National and Kapodistrian University of Athens, Medical School, Athens, Greece; ¹¹Danish Cancer Society Research Center, Copenhagen, Denmark; ¹²AO Citta' della Salute e della Scienza - Center for Cancer Prevention (CPO Piedmont) , Turin, Italy; ¹³School of Chemical Engineering, National Technical University of Athens, Athens, Greece; ¹⁴IUF Leibniz Research Institute for Environmental Medicine, University of Düsseldorf, Düsseldorf, Germany; ¹⁵Department of Epidemiology and Public Health, Swiss Tropical and Public Health Institute, Basel, Switzerland; ¹⁶Department of Environmental Health, National Institute for Health and Welfare, Kuopio, Finland; ¹⁷School of Environment and Development (Geography), University of Manchester, Manchester, England, United Kingdom; ¹⁸Department of Public Health and Clinical Medicine, Umeå University, Umeå, Sweden; ¹⁹Centre for Occupational and Environmental Health, The University of Manchester, Manchester, England, United Kingdom; ²⁰Institute of Health and Society, University of Oslo, Oslo, Norway; ²¹Norwegian Institute of Public Health, Oslo, Norway; ²²University of Basel, Basel, Switzerland; ²³Department of Environmental and Occupational Health Sciences, University of Washington, Seattle, USA; ²⁴Department of Air Hygiene, National Institute of Environmental Health, Budapest, Hungary; ²⁵Institute of Epidemiology and Medical Biometry, Ulm University, Ulm, Germany; ²⁶Medical Faculty, Heinrich-Heine University of Düsseldorf, Düsseldorf, Germany; ²⁶Julius Center for Health Sciences and Primary Care, University Medical Center Utrecht, Utrecht, the Netherlands

Address correspondence to Meng Wang, Institute for Risk Assessment Sciences, Division Environmental Epidemiology, Utrecht University, PO Box 80178 3508 TD Utrecht, the Netherlands. Telephone: +31 (0)6 16697117. Fax: +31 (0)30 2539499. E-mail: M.Wang@uu.nl

Running title: Multi-cities land use regression modeling

Acknowledgments: We thank all those who participated and were responsible for air pollution measurements, data management, model building and project supervision in all the ESCAPE study areas. 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. The funding source had no role in the study design; in the collection, analysis and interpretation of data; in the writing of the report; and in the decision to submit the manuscript for publication.

Competing financial interests: The authors declare they have no competing financial interest.

Abstract

Background: Land use regression (LUR) models have mostly been developed to explain intra-urban variations in air pollution based on often small local monitoring campaigns. Transferability of LUR models from city to city has been investigated, but little is known about the performance of models based on large numbers of monitoring sites covering a large area.

Objectives: To develop European and regional LUR models and to examine their transferability to areas not used for model development.

Methods: We evaluated LUR models for nitrogen dioxide (NO₂) and Particulate Matter (PM_{2.5}, PM_{2.5} absorbance) by combining standardized measurement data from 17 (PM) and 23 (NO₂) ESCAPE study areas across 14 European countries for PM and NO₂. Models were evaluated with cross validation (CV) and hold-out validation (HV). We investigated the transferability of the models by successively excluding each study area from model building.

Results: The European model explained 56% of the concentration variability across all sites for NO₂, 86% for PM_{2.5} and 70% for PM_{2.5} absorbance. The HV R²s were only slightly lower than the model R² (NO₂: 54%, PM_{2.5}: 80%, absorbance: 70%). The European NO₂, PM_{2.5} and PM_{2.5} absorbance models explained a median of 59%, 48% and 70% of within-area variability in individual areas. The transferred models predicted a modest to large fraction of variability in areas which were excluded from model building (median R²: 59% NO₂; 42% PM_{2.5}; 67% PM_{2.5} absorbance).

Conclusions: Using a large dataset from 23 European study areas, we were able to develop LUR models for NO₂ and PM metrics that predicted measurements made at independent sites and areas reasonably well. This finding is useful for assessing exposure in health studies conducted in areas where no measurements were conducted.

Introduction

Many studies have documented adverse health effects associated with long-term exposure to air pollutants (Brunekreef and Holgate 2002). With the improvement of the accuracy of geographical data, air pollution models incorporating data from geographical information system (GIS) are of increasing interest in exposure assessment (Hoek et al. 2008; Jerrett et al. 2005). Land use regression (LUR) modeling is a popular method used for exposure assessment in health studies (Cesaroni et al. 2013; Estarlich et al. 2011; Gehring et al. 2011). LUR modeling is a GIS and statistics based method that exploits land use, geographic and traffic characteristics (e.g. traffic intensity, road length, population density) to explain spatial concentration variations at monitoring sites.

Land use regression models were mostly constructed and utilized to predict concentrations within metropolitan areas (Hoek et al. 2011; Madsen et al. 2007; Marshall et al. 2008) or small regions (Brauer et al. 2003; Henderson et al. 2007). Often, models have been based on measurements made at a relatively small number of sampling sites (20~80 sites). Our recent study showed a positive association between the number of sampling sites and the prediction capability of models for NO₂ based on 144 sites in the Netherlands (Wang et al. 2012), in agreement with observations for Girona, Spain (Basagaña et al. 2012). At least for some of the reported studies, there is still room to improve the model performances if more sampling sites were selected (Hoek et al. 2008). Several studies have reported the possibilities of building models in large areas in Europe, United States and Canada (Beelen et al. 2009; Hart et al. 2009;

Hystad et al. 2011; Vienneau et al. 2009; Vienneau et al. 2013). With a large number of sites, these models explained large fractions of NO₂ variability (61%~90%) and a modest fraction of the variability of PM (40%~50%) across all sites. The large-area studies were all based upon routine monitoring data. National routine monitoring networks may include only a small number of sites within individual cities. Therefore it may be difficult to evaluate how well a large-area model explains within-city variability. This is relevant for epidemiological studies based in individual cities. A study in Switzerland based upon study-specific monitoring suggested that a country wide model did not perform well within six of the eight geographically diverse study areas (Liu et al. 2012).

The applicability of LUR models can be increased by transferring them to adjacent areas with similar geography and GIS databases where no or few measurements were conducted. The transferability of models has been investigated for local and national models (Allen et al. 2011; Poplawski et al. 2009; Vienneau et al. 2010). Most of the earlier studies recommended using the locally built models, even though transferred models explained variations in concentrations fairly well. This is because all the transferred models were city-city or country-country transfers for which local specific variables were not available and there was no advantage in the number of sampling sites as compared to the locally developed models.

So far, few studies attempted to explore the performance of LUR models with combined geographical areas in terms of prediction ability and transferability at independent sites and areas mainly due to lack of sufficient, comparable measurement data. In the context of the European

Study of Cohorts for Air Pollution Effects (ESCAPE 2013), we applied a standardized approach for measurements, GIS variable collection and model development for NO₂ and Particulate Matter (PM) in 36 study areas in Europe (Beelen et al. 2013; Cyrus et al. 2012; Eeftens et al. 2012a; Eeftens et al. 2012b). We recently published LUR models developed within individual study areas for nitrogen dioxide (NO₂) and particulate matter (Beelen, et al. 2013; Eeftens et al. 2012a). The ESCAPE database provides a unique opportunity to address important questions regarding application of LUR models developed for even larger areas. Therefore, the aims of this study are 1) to develop LUR models for NO₂, PM_{2.5} and PM_{2.5} absorbance based on combining the ESCAPE study areas across Europe and across four regions of Europe; 2) to evaluate the model performances systematically in terms of the model fitting and prediction ability; and 3) to investigate the transferability of the regional and European models to monitoring sites and areas not included in the model building.

Methods

Study areas and air pollution measurements

Details of the ESCAPE study design and the measurement campaign have been described previously (Cyrus et al. 2012; Eeftens et al. 2012b). Briefly, an intensive monitoring campaign was conducted in 36 European study areas between October 2008 and May 2011. ESCAPE included 20 areas with simultaneous measurements of both PM and NO₂ at 20 sites per area, and 20 sites where only NO₂ was measured. In an additional 16 areas, where PM measurements were not available, only NO₂ measurements were conducted at 40 sites per area. The number of

measurement sites was doubled in the large study area of the Netherlands and Belgium. In each area, we chose sampling sites at street, urban background and regional background locations. Sites were also selected to cover locally important variation e.g. presence of a port or altitude. These sites were selected to represent the spatial distribution of air pollution and residential addresses of participants of cohort studies in these areas. The background sites have been carefully selected to the locations not influenced by local traffic and other local emissions (e.g. industry and port) (Beelen et al. 2013; Eeftens et al. 2012a). Annual average concentrations were calculated from three two-week samples in the cold, warm and intermediate seasons. Due to limited number of samplers, five sites and the reference site were measured simultaneously. The measured values were adjusted for temporal trends with data from the continuous reference site in each area by calculating absolute differences between concentrations at monitoring sites and reference sites and using that as adjustment factor (Cyrys et al. 2012; Eeftens et al. 2012b). In this paper, we selected the 23 areas (Figure 1) in which traffic intensity variables were available for LUR model building in line with the importance of traffic intensity variables in model development (Beelen et al. 2013). This included 17 of the 20 PM/NO₂ areas and 6 of the 16 NO₂ only areas. We allocated the areas to 4 regions according to the geographical location, the characteristics of the climate, the traffic intensity levels and the configuration of the cities/country. These regions included five areas in north Europe (Oslo, Stockholm, Copenhagen, Helsinki/Turku, Umea), seven in the west (Netherlands and Belgium, London, Manchester, Bradford, Ruhr area, Erfurt, Paris), six in the center (Munich, Vorarlberg, Győr, Lugano,

Grenoble, Lyon) and five in the south (Turin, Rome, Athens, Barcelona, Marseille) (Figure 1 and Table 1).

For this study we selected NO₂ and PM_{2.5} absorbance to represent traffic-related air pollution and PM_{2.5} for a more complex mixture of sources. NO₂ was measured using Ogawa badges following the Ogawa analysis protocol (Ogawa&Co V 3.98, USA, Inc.). PM_{2.5} samples were collected on pre-weighted filters using Harvard Impactors, and were then used to measure absorbance (Cyrus et al. 2012; Eeftens et al. 2012b).

Predictor variables

We extracted values for the GIS predictor variables at the locations of sampling sites using ArcGIS (ESRI, Redlands, California). Details of the predictor variables have been described in previous papers (Beelen et al. 2013; Eeftens et al. 2012a). Briefly, the predictor variables were derived from both centrally available Europe-wide GIS databases and locally collected GIS data by the local centers using standard definitions.

Central GIS predictor variables included road network, land use, population density and altitude data. The digital road network was obtained from EuroStreets version 3.1 (EuroStreets 2013) for the year 2008. For all roads and major roads, the total lengths of roads were calculated within a buffer size of 25, 50, 100, 300, 500, 1000 meters. Traffic intensity data were not available for this road network. Land use variables were derived from the European Corine Land Cover (CLC 2000) database for the year 2000 for the buffer sizes of 100, 300, 500, 1000 and 5000 meters. Digital elevation data were obtained through the Shuttle Radar Topographic Mission (SRTM

2013). Detailed road network with linked traffic intensity for all road links were obtained from local sources for all 23 areas. Local land use, population density, altitude and other local variables were also locally extracted for modeling.

For the regional and European models, we pooled the data by including all the central GIS predictors and the local traffic variables with traffic intensity. We combined the centrally available land use variables high and low residence density, and the natural and urban green variables as not all the areas contained them separately. We made efforts to incorporate more local common variables for specific regions to capture regional variations. We included regional background concentrations of NO₂, PM absorbance and PM_{2.5} as the mean of the measured concentrations at (1 to 20) ESCAPE regional background sites in each local study area to characterize the spatial differences between study areas. In the Netherlands, regional background concentrations were interpolated from regional background sites throughout the country because background concentrations may vary at such a large scale. In total, 49 variables were evaluated at the European level and 54, 53, 54, 64 variables in the north, west, middle and south regions (see Supplemental Material, Table S1).

Model development

A total of 960 NO₂ sites and 356 PM sites (4 sites were missing due to failed campaign) were available for modeling from 23 and 17 study areas respectively. Detailed procedures of the NO₂ and PM model development have been published elsewhere (Beelen et al. 2013; Eeftens et al. 2012a). The regional and European models were developed using the same strictly standardized

approaches. Briefly, a supervised stepwise regression was used to develop the LUR model. We first evaluated univariate regression of the annual concentrations by entering all potential predictor variables. We forced the regional background concentration variable in the first step (for the European and regional models). Then the variable which produced the highest adjusted R^2 and which had the a priori defined direction of effect (e.g. positive for traffic intensity) was selected as the second predictor. Secondly, the remaining variables were added separately and we assessed whether the variable with the highest increase in adjusted R^2 improved the model by at least 1%. This process continued until no more variable with the a priori specified sign could increase the model adjusted R^2 by at least 1%. In the final step, we excluded the variables which had a p-value >0.1 . We checked whether the variance inflation factor (VIF) was lower than 3 in order to avoid multi-collinearity.

Model evaluations

We used three approaches for model evaluation:

1. We investigated the model fit at individual study areas by applying the European/regional model to the sites of each area that were used for modeling. The $\text{Model}_{\text{intra}} R^2$ shows the within area variations explained by the European/regional models which is directly comparable with the R^2 of city-specific models. The $\text{Model}_{\text{intra}} R^2$ is important for studies conducted within individual cities that use the European/regional model. The overall R^2 is relevant for multi-city studies that exploit both within and between city- variability of air pollution contrasts. The $\text{Model}_{\text{intra}} R^2$ is important for European studies such as ESCAPE as

cohorts were located within a city or small area and cohort-specific epidemiological analyses were conducted.

2. Cross validation is an internal validation for testing the stability of model fit. We conducted leave-one-area-out-cross-validation (LOAOCV) by leaving out all observations from a complete area of M study areas ($N=23$ for NO_2 and 17 for PM), refitting the model based on the remaining $M-1$ areas, and investigating the agreement between predicted and observed concentrations for each area that was left out. This was iterated M times and the LOAOCV reflects the heterogeneity of model fit due to regional variations between study areas. We do not report leave-one-site-out cross-validation which was almost identical to the model R^2 probably because of the large training dataset.
3. The hold-out validation (HV) is an evaluation of model predictive power to independent sites not used for model building. In contrast with cross validation, it reflects the prediction ability of models to the cohort addresses within the areas on which the models had been established. As a test, we divided the full set into two parts, the training sets were used for modeling and the remaining test sets were used for external evaluation. For NO_2 , we developed models using the PM/ NO_2 sites with 20-40 sites per area (480 sites in total) as training sets and the remaining 480 NO_2 -only sites as test sets. For $\text{PM}_{2.5}$ and $\text{PM}_{2.5}$ absorbance, a randomly selected 25% of the PM sites stratified by study area were used for validation purpose as we had fewer sites available for PM model building than for NO_2 model building. The HV R^2 is the squared Pearson correlation between predictions and observations at the independent sites throughout the whole study area. We calculated the HV

R^2 by truncating the values of predictors in the test data sets that were outside the range of the values observed in the data set for model development, to prevent unrealistic predictions based on model extrapolations (Wang et al. 2012). Prediction errors were estimated by root mean squared error (RMSE). In our previous study, the same NO_2 training and test sets were used for the ESCAPE city-specific model evaluations individually in each study area (Wang et al. 2013). Therefore, a fair comparison of prediction ability (HV R^2) between the European model and the city-specific models can be conducted using the same test sets for hold-out validation. The comparison was available only for NO_2 due to relatively large number of sampling sites.

Transferability of LUR models

To evaluate the prediction abilities of the regional/ European models to independent individual study areas, we developed the regional and European models by excluding one area at a time and applied the transferred models directly to the sites of the area that was left out. Therefore, 23 NO_2 models and 17 PM models were built respectively until each of the study areas had been excluded once from model building.

The $\text{TRANS}_{\text{intra}} R^2$ is the squared Pearson correlation between observed and predicted values in each of the remaining area that was excluded from modeling. The $\text{TRANS}_{\text{intra}} R^2$ is different from the $\text{Model}_{\text{intra}}$ and $\text{LOAOCV } R^2$ as the measurements conducted in the respective validation areas were completely left out from model development.

Results

NO₂ and PM concentrations

Table 2 shows the concentration distributions of NO₂ and PM metrics across the study areas by site types. Substantial spatial variations were found for all the pollutants across Europe. The variability was larger for NO₂ than for PM_{2.5}. The spatial variability for PM_{2.5} absorbance was intermediate between PM_{2.5} and NO₂. Concentration contrasts were larger at the street sites for NO₂ and PM_{2.5} absorbance than at the urban and rural background sites. Concentration contrasts for PM_{2.5} were more similar at all the site types, suggesting an influence of multiple sources in addition to traffic.

Models in combined areas

Table 2 shows the model details of NO₂, PM and PM_{2.5} absorbance combining all the European study areas. The NO₂, PM_{2.5} and PM_{2.5} absorbance models explained 56%, 86% and 70%, respectively, of the variation across all sites, which includes both within and between area variations (overall model R²). The LOAOCV R² was 5% and 6% lower than the model R² for NO₂ and PM_{2.5}, and was identical to the model R² for PM_{2.5} absorbance. The hold-out validation R²s (50% training vs. 50% test sites for NO₂, 75% training vs. 25% test sites for PM metrics) were slightly smaller than or nearly identical to the model R²s, explaining 54%, 80% and 70% for NO₂, PM_{2.5} and PM_{2.5} absorbance at the independent validation sites respectively (see Supplemental Material, Table S2). The HV R² did not change if the predictor range was not truncated as only 1 site for NO₂ model was truncated. The HV RMSE values were close to the

values of LOAOCV RMSE for NO₂ and PM metrics. The RMSE values were relatively small compared to the range of measurements as shown in Table S2 (see Supplemental Material). The median HV R² of the European NO₂ model at individual study areas was identical to those of the city-specific models reported in Wang et al. (2013) (see Supplemental Material, Figure S1). In e.g. the Turin and Paris areas with a low hold-out evaluation R², the HV R²s of the European model were considerably larger than those of the city-specific models.

All the models in Table 3 included traffic intensity variables. The regional background concentration explained a large fraction (71%) of variation in PM_{2.5} documenting the importance of between-area differences for PM_{2.5} as compared to that for the more traffic-related pollutants NO₂ and PM_{2.5} absorbance.

The regional models performed equally well as the European models in all regions except Southern Europe, where none of the models performed well in terms of the predictions to the independent sites (HV R²: 0-0.23) (see Supplemental Material, Table S3). Reassigning Turin from south Europe to central Europe region only slightly changed the results.

As shown in Table 3, the median within-area variability (Model_{intra} R²) explained by the European model for NO₂ and absorbance at individual study areas was similar to the overall model R², suggesting predominant sources of local emissions. For PM_{2.5}, the median Model_{intra} R² was much lower than the overall model R² (0.48 vs. 0.86). Figure 2 and Figure S2 (see Supplemental Materials) present the correlation between predicted and measured PM_{2.5}, PM_{2.5}

absorbance and NO₂ by study areas. As the figures show, the variation of PM_{2.5} between areas was substantial compared to the within areas variation (e.g. low PM_{2.5} values in northern European cities such as Stockholm and high PM_{2.5} values in southern European cities such as Rome). On the contrary, for NO₂ and PM_{2.5} absorbance, variation within areas was substantial compared to the variation between areas (Supplemental Material, Figure S2). The observations are more under-predicted within individual areas for PM metrics (median regression slope: 0.47 PM_{2.5}; 0.57 PM_{2.5} absorbance; 0.56 NO₂) than across the whole European study areas (regression slope: 0.85 PM_{2.5}; 0.70 PM_{2.5} absorbance; 0.57 NO₂).

Transferability

Table 4 shows the performance of the models which used all monitoring data excluding one area at the time. These models explained on average 57%, 84% and 69% variability of NO₂, PM_{2.5} and PM_{2.5} absorbance respectively. The model structures and R²s were similar to the models in Table 3 which were based on all study areas. They included the same variable categories but with to some extent different buffer sizes. The models predicted the spatial variations of NO₂ and PM_{2.5} absorbance well in the areas not used for model building with median TRANS_{intra} R²s of 0.59 for NO₂ and 0.67 for PM_{2.5} absorbance. Transferability was less for PM_{2.5} with a median R² of 0.42. The same pattern was found for the model R² focusing on within-area variability only (Model_{intra}). The variation in prediction R²s was relatively small for NO₂ with an IQR of 0.09, but larger for PM_{2.5} (IQR 0.17) and PM_{2.5} absorbance (IQR 0.21) showing that predictions were less comparable for the two PM metrics. The variation is shown in Figure 3 and Figure S3 (see

Supplemental Materials). Interestingly, this did not depend so much on area as on the specific combination of area and component. For example, the areas in Hungary (Gyor), Germany (Munich) and Austria (Vorarlberg) showed decent model fit and predictability for NO₂ and PM_{2.5} absorbance, but almost no model fit and predictability for PM_{2.5}. The transferred regional models showed similar characteristics as those of the European models, while the median TRANS_{intra} R² was slightly lower (see Supplemental Material, Table S4).

Discussion

In this study we developed LUR models for NO₂, PM_{2.5} and PM_{2.5} absorbance with combined measurement data from 23 study areas across Europe. For NO₂ and PM_{2.5} absorbance, these models predicted spatial variations in areas not used for model building well. For PM_{2.5}, prediction R²s were moderate for intra-urban variation though in some areas in central Europe prediction R²s were low. The overall R² including both between and within study area variability was high for PM_{2.5} and PM_{2.5} absorbance and more moderate for NO₂.

Comparisons with other large area studies

Our European models performed comparable or even better in predictions of NO₂ and PM_{2.5} than other large area studies (Table S5) (Beelen et al. 2009; Beckerman et al. 2013; Bergen et al. 2013; Hystad et al. 2011; Novotny et al. 2011; Sampson et al. 2013; Vienneau et al. 2013). For PM_{2.5} absorbance, this is the first report of LUR models in such a large geographical area. Model R²s are difficult to compare as studies differed in study area, model development strategies, scale of prediction, offered predictor variables and number of sites. Consistent across studies, the

regional background predicted a small fraction of variability for NO₂ and a large fraction for PM_{2.5}. For intra-city model R², our NO₂ European model exhibited comparable performance (Model_{intra} R²=0.59) to the Canadian national model in seven specific areas (Edmonton, Montreal, Sarnia, Toronto, Victoria, Vancouver and Winnipeg) with Model_{intra} R² of 0.43 (Hystad et al. 2011). We observed no heterogeneity of model fit across study areas in the European model (LOAOCV R²s were close to the model R²).

Our European and regional models have several strengths compared to previous European models which modeled concentration in 1×1 km grids (Beelen et al. 2009; Vienneau et al. 2009) or more recently 100×100m (Vienneau et al. 2013): 1) We modeled small scale variation using sampling sites which were selected according to a standard method to cover intra-urban concentration contrasts, 2) We included multiple pollutants (PM_{2.5}, PM absorbance,) which were much less available or measured with different methods from routine monitoring networks in Europe. 3) We incorporated local traffic intensity data not available in Europe-wide databases (land use and road length data only). All the models included traffic intensity variables, improving prediction ability (HV R²) over models not having local traffic intensity data (but potentially road length) e.g. from 0.46 to 0.54 for NO₂.

The poor performance of the south European model may be attributed to the large heterogeneity of model fit (low LOAOCV R²) across south European study areas in which the concentrations in Athens were overestimated more than those of the other study areas. More formal methods

such as hierarchical cluster analysis to define regions could be explored to improve comparability of regions.

Our PM_{2.5} European model explained a median of 48% within-area variations as compared to the overall model R² of 86% which was largely explained by substantial differences in regional background concentrations. This was consistent with the R²s of the Canadian and American PM_{2.5} model (46% and 63%) of which the satellite data alone explained 41% and 52% of the variability respectively (Hystad et al. 2011; Beckerman et al. 2013) (see Supplemental Material, Table S5). PM_{2.5} is well known to be a regional pollutant with a large fraction of secondary aerosol, not explained well by the local GIS and traffic variables typically available for LUR model building. This suggested that for pollutants (e.g. PM_{2.5}) with much larger overall than within-city R², joint analyses of cohorts including between-city exposure components might be advisable. This does require the assumption of sufficient comparability of cohorts across Europe. Other methods such as Partial Least Squares regression may help to increase the prediction ability of models (Sampson et al. 2013).

Comparison with ESCAPE city-specific models

NO₂ and PM models based on small training sets and a large number of predictor variables overestimate predictive ability in independent test sets though still explaining fairly large fractions (50%~60%) of spatial variability (Wang et al. 2012; Wang et al. 2013). Hold Out validation R²s of the European models developed on a large number of sites were very similar to the model R². The average differences of the model R²s and HV R²s were just 2%, 6% and 0%

for NO₂, PM_{2.5} and PM_{2.5} absorbance. The slightly larger drop for PM_{2.5} could be due to more sources affecting PM_{2.5} compared to NO₂ and PM_{2.5} absorbance.

The ESCAPE city-specific models which have been published previously using local specific variables explained a median of 82%, 71% and 89% of the concentration variations for NO₂, PM_{2.5} and PM_{2.5} absorbance (Beelen et al. 2013; Eeftens et al. 2012a). This is higher than the R² of within-area variability explained by the European models in Table 2 (Model_{intra} R²: 59%, 48%, 70% respectively). The average differences between the individual city-specific model R² (Beelen et al. 2013; Eeftens et al. 2012a) and the intra-urban R² (see Supplemental Material, Figure S3) are 24%, 24% and 17% for NO₂, PM_{2.5} and PM_{2.5} absorbance respectively. As model R² overestimates predictive ability especially when developed for a relatively small number of sites (Wang, 2012; Wang, 2013), the comparison between local and European models should not be based on model R² but holdout evaluation R² at independent sites. Comparison of the prediction ability between the European and city-specific models is feasible only for NO₂ which suggested that the European and city-specific model had similar median prediction ability to the external sites not used for modeling. The HV R² in some cities e.g. Turin (ITU) and Paris (FPA) which had poor predictions by the city-specific model may be improved substantially by the European model. We cannot draw a firm conclusion about one or the other approach being more reliable because comparisons for PM models were infeasible. The European model may reduce bias in health estimates due to relatively large number of sampling sites and small number of variables as compared to the city-specific models (Basagaña et al. 2013).

Most of the combined models included traffic variables in both large ($\geq 500\text{m}$) and small buffers ($\leq 50\text{m}$), representing general area characteristics as well as localized influences. In contrast to the study-area specific ESCAPE models (Beelen et al. 2013; Eeftens et al. 2012a), none of our European models included population/residence density but instead selected road length in large buffers which likely also represents urban-rural difference in terms of population distributions (de Hoogh et al. 2013). In our GIS dataset, the squared correlation R^2 between road length and population density is 0.46 within 1000m buffer but is only 0.13 within 100m buffer. Road length variables in large buffers represent therefore various aspects of “total human activity” such as traffic, heating, population density, etc.

Transferability of combined models

Previous studies on the transferability of LUR models were mainly focusing on city-to-city or country-to-country transferability. Briggs et al. (2000) concluded that the SAVIAH (Small-Area Variations In Air Quality and Health) models could be applied to other UK cities after calibrating with data from a few monitoring sites. Poplawski et al. (2009) and Allen et al. (2011) observed that local calibration may improve the predictions of the Canadian city specific models to a few other comparable cities in Canada and the United States. Vienneau et al. (2010) found reasonable transferability of British and the Dutch models between these two countries. All the previous studies concluded that the performances of the transferred models were worse than those of the local source models. Our results show prediction capabilities for the traffic related pollutants NO_2 and $\text{PM}_{2.5}$ absorbance which are on par with those documented, in terms of Hold

Out validation R^2 s, with previous local exercises (Basagaña et al. 2012; Wang et al. 2012). This might be due to the fact that the ESCAPE study used highly standardized monitoring and GIS data for measurement, data collection and model building across all areas. This suggests that our combined models can be carefully applied to other areas in Europe with common predictors, similar geographies and availability of consistent regional background concentration within the region. Because the locations are well characterized, any candidate background location in a new area can be judged against the same criteria. Obviously, this will only work when the pollution characteristics or components are actually measured in the new area. In practice, this means that modeling of new areas will in most cases be restricted to NO_2/NO_x and PM_{10} , in fewer areas (in Europe) to $\text{PM}_{2.5}$ and PM absorbance. Satellite data have large spatial coverage and have improved NO_2 and PM_{10} European models based upon routine monitor data by 5% and 11% (Vienneau et al. 2013). Satellite data could be used in the future to estimate background concentrations in new locations.

In some individual areas of central Europe, the European model performed poorly for $\text{PM}_{2.5}$ however, probably due to lack of an important local predictor variable, e.g. residential density in Munich and Vorarlberg, industry in Hungary or altitude in Vorarlberg. Therefore, caution is needed when transferring the European models to cities for which the European model lacks predictor variables that are known to be important sources of variation locally. The poor performance in a few areas, suggests that the value of the European model is especially in multi-center analyses such as ESCAPE compared to individual areas.

Applications in epidemiological studies

The overall R^2 of the European model was highest for $PM_{2.5}$ and lowest for NO_2 . In contrast, for within-city variation the model had the lowest predictive ability for $PM_{2.5}$ though still fairly high (median $R^2=0.48$). The $PM_{2.5}$ absorbance model explained both large fractions of variability overall and within-city. The high overall R^2 suggests that the model can be used in pooled analyses of health data, exploiting exposure contrasts between study areas. Using between-city comparisons would especially be useful to increase $PM_{2.5}$ contrasts. For ESCAPE where the health findings based on these local exposure models are currently being published (Beelen et al. 2014; Raaschou-Nielsen et al. 2013), the model offers the possibility for pooled analyses. Pooled analyses have not been conducted so far, partly because of concerns of comparability of the diverse cohorts across Europe. There is also the perspective to include new study populations from areas where local measurements were never conducted but relevant predictor variables are available. For exposure assessment with LUR models, efforts are mainly in the sampling campaign and GIS data collection.

Conclusions

European LUR models for NO_2 , $PM_{2.5}$ and $PM_{2.5}$ absorbance were found to have reasonable power to predict spatial variations of these components in areas not used for model building.

References

- Allen RW, Amram O, Wheeler AJ, Brauer M. 2011. The transferability of NO and NO₂ land use regression models between cities and pollutants. *Atmos Environ* 45:369-378.
- Basagaña X, Rivera M, Aguilera I, Agis D, Bouso L, Elosua R, et al. 2012. Effect of the number of measurement sites on land use regression models in estimating local air pollution. *Atmos Environ* 54:634-642.
- Basagaña X, Aguilera I, Rivera M, Agis D, Foraster M, Marrugat J, et al. 2013. Measurement error in epidemiologic studies of air pollution based on land-use regression models. *Am J Epidemiol* 178:1342-1346.
- Beckerman BS, Jerrett M, Serre M, Martin RV, Lee SJ, van Donkelaar A, et al. 2013. A hybrid approach to estimating national scale spatiotemporal variability of PM in the contiguous United States. *Environ Sci Technol* 47: 7233-7241.
- Beelen R, Hoek G, Pebesma E, Vienneau D, de Hoogh K, Briggs DJ. 2009. Mapping of background air pollution at a fine spatial scale across the European Union. *Sci Total Environ* 407:1852-1867.
- Beelen R, Hoek G, Vienneau D, Eeftens M, Dimakopoulou K, Pedeli X, et al. 2013. Development of NO₂ and NO_x land use regression models for estimating air pollution exposure in 36 study areas in Europe - the ESCAPE project. *Atmos Environ* 72:10 - 23.
- Beelen R, Raaschou-Nielsen O, Stafoggia M, Andersen ZJ, Weinmayr G, Hoffmann B, et al. 2014. Effects of long-term exposure to air pollution on natural-cause mortality: An analysis of 22 european cohorts within the multicentre escape project. *Lancet* 383:785-795.
- Bergen S, Sheppard L, Sampson PD, Kim SY, Richards M, Vedal S, et al. 2013. A national prediction model for pm component exposures and measurement error-corrected health effect inference. *Environ Health Perspect* 121:1017-1025.
- Brauer M, Hoek G, van Vliet P, Meliefste K, Fischer P, Gehring U, et al. 2003. Estimating long-term average particulate air pollution concentrations: Application of traffic indicators and geographic information systems. *Epidemiology* 14:228-239.

- Briggs DJ, de Hoogh C, Guiliver J, Wills J, Elliott P, Kingham S, et al. 2000. A regression-based method for mapping traffic-related air pollution: Application and testing in four contrasting urban environments. *Sci Total Environ* 253:151-167.
- Brunekreef B, Holgate ST. 2002. Air pollution and health. *Lancet* 360:1233-1242.
- Cesaroni G, Badaloni C, Gariazzo C, Stafoggia M, Sozzi R, Davoli M, et al. 2013. Long-term exposure to urban air pollution and mortality in a cohort of more than a million adults in rome. *Environ Health Perspect* 121:324-331.
- CLC2000 (Corine land cover). 2000. Raster database (version 12/2009).
<http://www.eea.europa.eu/data-and-maps/data/corine-land-cover-2000-clc2000-100-m-version-12-2009> [accessed 20 May 2013].
- Cyrys J, Eeftens M, Heinrich J, Ampe C, Armengaud A, Beelen R, et al. 2012. Variation of NO₂ and NO_x concentrations between and within 36 European study areas: Results from the ESCAPE study. *Atmos Environ* 62:374 - 390.
- de Hoogh K, Wang M, Adam M, Badaloni C, Beelen R, Birk M, et al. 2013. Development of land use regression models for particle composition in twenty study areas in Europe. *Environ Sci Technol* 47:5778-5786.
- Eeftens M, Beelen R, de Hoogh K, Bellander T, Cesaroni G, Cirach M, et al. 2012a.
Development of land use regression models for PM_{2.5}, PM_{2.5} absorbance, PM₁₀ and PMcoarse in 20 European study areas; results of the ESCAPE project. *Environ Sci Technol* 46:11195-11205.
- Eeftens M, Tsai M, Ampe C, Anwander B, Beelen R, Cesaroni G, et al. 2012b. Spatial variation of PM_{2.5}, PM₁₀, PM_{2.5} absorbance and PMcoarse concentrations between and within 20 European study areas and the relationship with NO₂ - results of the ESCAPE project. *Atmos Environ* 62:303 - 317.
- ESCAPE (European Study of Cohorts for Air Pollution Effects). 2013. Manuals. Available:
<http://www.escapeproject.eu/manuals/> [accessed 20 May 2013]

- Estarlich M, Ballester F, Aguilera I, Fernandez-Somoano A, Lertxundi A, Llop S, et al. 2011. Residential exposure to outdoor air pollution during pregnancy and anthropometric measures at birth in a multicenter cohort in Spain. *Environ Health Perspect* 119:1333-1338.
- EuroStreets. 2013. EuroStreets digital road network version 3.1. Available: <http://www.geodan.com/products/geographic-data-catalogue/products/eurostreets/> [accessed 20 May 2013].
- Gehring U, Wijga AH, Fischer P, de Jongste JC, Kerkhof M, Koppelman GH, et al. 2011. Traffic-related air pollution, preterm birth and term birth weight in the piama birth cohort study. *Environ Res* 111:125-135.
- Hart JE, Yanosky JD, Puett RC, Ryan L, Dockery DW, Smith TJ, et al. 2009. Spatial modeling of PM₁₀ and NO₂ in the continental United States, 1985-2000. *Environ Health Perspect* 117:1690-1696.
- Henderson SB, Beckerman B, Jerrett M, Brauer M. 2007. Application of land use regression to estimate long-term concentrations of traffic-related nitrogen oxides and fine particulate matter. *Environ Sci Technol* 41:2422-2428.
- Hoek G, Beelen R, de Hoogh K, Vienneau D, Gulliver J, Fischer P, et al. 2008. A review of land-use regression models to assess spatial variation of outdoor air pollution. *Atmos Environ* 42:7561-7578.
- Hoek G, Beelen R, Kos G, Dijkema M, van der Zee SC, Fischer PH, et al. 2011. Land use regression model for ultrafine particles in Amsterdam. *Environ Sci Technol* 45:622-628.
- Hystad P, Setton E, Cervantes A, Poplawski K, Deschenes S, Brauer M, et al. 2011. Creating national air pollution models for population exposure assessment in Canada. *Environ Health Perspect* 119:1123-1129.
- Jerrett M, Arain A, Kanaroglou P, Beckerman B, Potoglou D, Sahuvaroglu T, et al. 2005. A review and evaluation of intraurban air pollution exposure models. *J Expo Anal Environ Epidemiol* 15:185-204.

- Liu LJS, Tsai MY, Keidel D, Gemperli A, Ineichen A, Hazenkamp-von Arx M, et al. 2012. Long-term exposure models for traffic related NO₂ across geographically diverse areas over separate years. *Atmos Environ* 46:460-471.
- Madsen C, Carlsen KCL, Hoek G, Oftedal B, Nafstad P, Meliefste K, et al. 2007. Modeling the intra-urban variability of outdoor traffic pollution in Oslo, Norway - a GA(2)LEN project. *Atmos Environ* 41:7500-7511.
- Marshall JD, Nethery E, Brauer M. 2008. Within-urban variability in ambient air pollution: Comparison of estimation methods. *Atmos Environ* 42:1359-1369.
- Novotny E V, Bechle MJ, Millet DB, Marshall JD. 2011. National satellite-based land-use regression: NO₂ in the United States. *Environ Sci Technol* 45:4407-4414
- Poplawski K, Gould T, Setton E, Allen R, Su J, Larson T, et al. 2009. Intercity transferability of land use regression models for estimating ambient concentrations of nitrogen dioxide. *J Expo Anal Environ Epidemiol* 19:107-117.
- Raaschou-Nielsen O, Andersen ZJ, Beelen R, Samoli E, Stafoggia M, Weinmayr G, et al. 2013. Air pollution and lung cancer incidence in 17 European cohorts: Prospective analyses from the European study of cohorts for air pollution effects (ESCAPE). *Lancet Oncol* 14:813-822.
- Sampson PD, Richards M, Szpiro AA, Bergen S, Sheppard L, Larson TV, Kaufman JD. 2013. A regionalized national universal kriging model using Partial Least Squares regression for estimating annual PM_{2.5} concentrations in epidemiology. *Atmos Environ* 75: 383-392.
- SRTM (Shuttle Radar Topographic Mission 90m). 2013. Digital Elevation Database version 4.1. Available: <http://srtm.csi.cgiar.org/> [accessed 20 May 2013].
- Vienneau D, de Hoogh K, Briggs D. 2009. A GIS-based method for modelling air pollution exposures across Europe. *Sci Total Environ* 408:255-266.
- Vienneau D, de Hoogh K, Beelen R, Fischer P, Hoek G, Briggs D. 2010. Comparison of land-use regression models between great Britain and the Netherlands. *Atmos Environ* 44:688-696.

Vienneau D, de Hoogh K, Bechle MJ, Beelen R, van Donkelaar A, Martin R, et al. 2013.

Western European land use regression incorporating satellite- and ground-based measurements of NO₂ and PM₁₀. *Environ Sci Technol* 47: 13555-13564.

Wang M, Beelen R, Eeftens M, Meliefste K, Hoek G, Brunekreef B. 2012. Systematic evaluation of land use regression models for NO₂. *Environ Sci Technol* 46:4481-4489.

Wang M, Beelen R, Basagaña X, Becker T, Cesaroni G, de Hoogh K, et al. 2013. Evaluation of land use regression models for NO₂ and particulate matter in 20 European study areas: The ESCAPE project. *Environ Sci Technol* 47:4357-4364.

Table 1. Study areas.

Code	Type	Region	Study area
NOS	PM/NO ₂	North	Oslo, Norway
SST	PM/NO ₂	North	Stockholm, Sweden
FIH	PM/NO ₂	North	Helsinki, Turku, Finland
DCO	PM/NO ₂	North	Copenhagen, Denmark
SUM	NO ₂	North	Umea, Sweden
UKM	PM/NO ₂	West	Manchester, UK
UKO	PM/NO ₂	West	London, Oxford, UK
BNL	PM/NO ₂	West	Netherlands and Belgium
GRU	PM/NO ₂	West	Ruhr area, Germany
GRE	NO ₂	West	Erfurt, Germany
UKB	NO ₂	West	Bradford, UK
FPA	PM/NO ₂	West	Paris, France
GMU	PM/NO ₂	Central	Munich, Germany
AUV	PM/NO ₂	Central	Vorarlberg, Austria
FLY	NO ₂	Central	Lyon, France
HUG	PM/NO ₂	Central	Györ, Hungary
SWL	PM/NO ₂	Central	Lugano, Switzerland
FGR	NO ₂	Central	Grenoble, France
ITU	PM/NO ₂	South	Turin, Italy
IRO	PM/NO ₂	South	Rome, Italy
SPB	PM/NO ₂	South	Barcelona, Spain
FMA	NO ₂	South	Marseille, France
GRA	PM/NO ₂	South	Athens, Greece

Table 2. Distributions of measured annual average NO₂ and PM concentrations across Europe.

Pollutant and site type^a	N^b	Min	P25	Median	P75	Max
NO ₂ (µg/m ³)						
S	454	11.80	25.48	33.98	49.90	109.00
UB	414	3.03	15.38	22.88	30.67	57.63
RB	92	1.53	9.56	15.48	17.98	32.87
PM _{2.5} (µg/m ³)						
S	166	7.87	12.03	17.18	21.17	36.30
UB	144	5.62	10.97	15.87	18.62	32.59
RB	47	4.42	11.20	13.86	16.64	23.24
PM _{2.5} absorbance (10 ⁻⁵ m ⁻¹)						
S	166	0.78	1.63	2.16	2.81	5.09
UB	144	0.53	1.23	1.67	2.01	3.03
RB	47	0.33	0.92	1.16	1.45	2.37

^aSite types: S-street sites; UB-urban background; RB-regional background. ^bTotal number of sites in the study areas.

Table 3. European models for NO₂, PM_{2.5} and PM_{2.5} absorbance.

Predictors	Partial R ²	Beta ^a	Model _{intra} ^b R ² /IQR	LOAOCV ^c R ² /RMSE
NO ₂ (N ^d =960, final model R ² =0.56)		2.63E-01	0.59/0.19	0.50/8.49 (µg/m ³)
Regional background concentration	0.08	2.44E-06		
Traffic load in 50m	0.35	2.74E-04		
Road length in 1000m	0.50	-2.84E-07		
Natural and Green in 5000m	0.55	2.21E-04		
Traffic intensity on the nearest road	0.56	1.38E+01		
Intercept		2.63E-01		
PM _{2.5} (N ^d =356, final model R ² =0.86)			0.48/0.16	0.81/2.38 (µg/m ³)
Regional background concentration	0.71	9.73E-01		
Traffic load between 50m and 1000m	0.81	4.75E-09		
Traffic load in 50m	0.84	5.28E-07		
Road length in 100m	0.86	2.12E-03		
Intercept		3.06E-01		
PM _{2.5} absorbance (N ^d =356, final model R ² =0.70)			0.70/0.19	0.70/0.45 (10 ⁻⁵ m ⁻¹)
Regional background concentration	0.28	9.06E-01		
Traffic load in 50m	0.58	2.07E-07		
Road length in 500m	0.67	2.90E-05		
Natural and Green in 5000m	0.69	-9.63E-09		
Traffic load between 50m and 1000m	0.70	4.20E-10		
Intercept		2.95E-01		

^aBeta: coefficients of predictor variables in the models. ^bThe Model_{intra} R²s show the median and Inter Quartile Range of the within-area variability explained by the European model in individual areas. ^cLOAOCV: Leave-One-Area-Out-Cross-Validation. ^dN: number of monitored sites available for model building.

Table 4. Transferability of European models to areas which were not used for model building [median (IQR)].

Pollutant	Model R²	Model_{intra}^a R²	TRANS_{intra}^b R²	TRANS_{intra}^b RMSE
NO ₂	0.57(0.01)	0.59(0.19)	0.59(0.09)	5.58(2.28)
PM _{2.5}	0.84(0.01)	0.48(0.16)	0.42(0.17)	1.14(0.58)
PM _{2.5} absorbance	0.69(0.01)	0.70(0.19)	0.67(0.21)	0.23(0.07)

^aModel_{intra} R²: R² of within-area variation explained by European model, the same data as in Table 2. ^bTRANS_{intra}: squared correlations and RMSE between the predictions and observations at independent areas.

Figure legends

Figure 1. Map of study areas including region indication.

Figure 2. Scatterplot of predicted and measured $PM_{2.5}$ with study areas color and symbol coded and two city-specific examples, Rome (IRO) and Stockholm (SST). See Table 1 for study area codes.

Figure 3. Transferability ($TRANS_{intra} R^2$) of the European models for NO_2 and PM in the 23 study areas. See Table 1 for study area codes.

Figure 1.

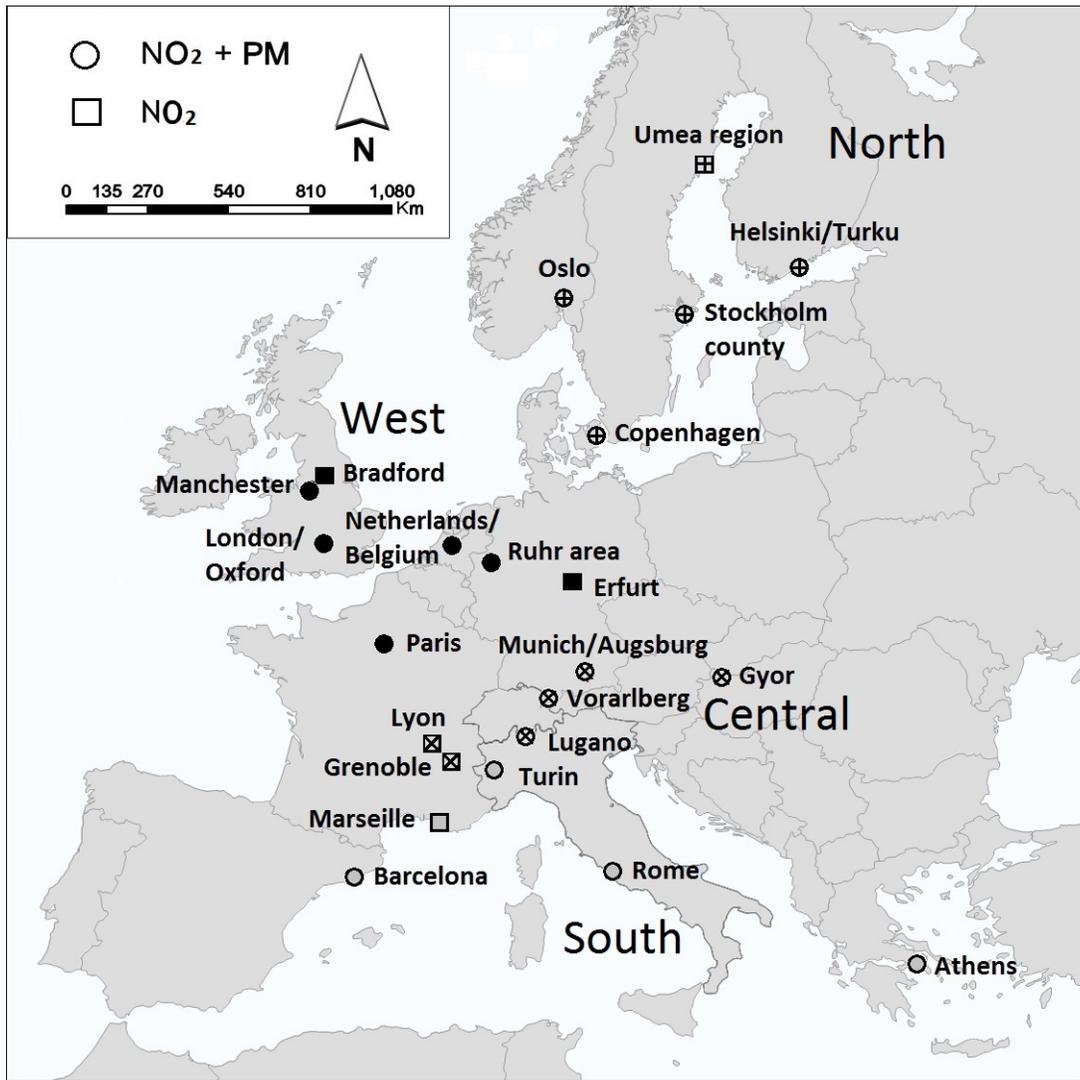


Figure 2.

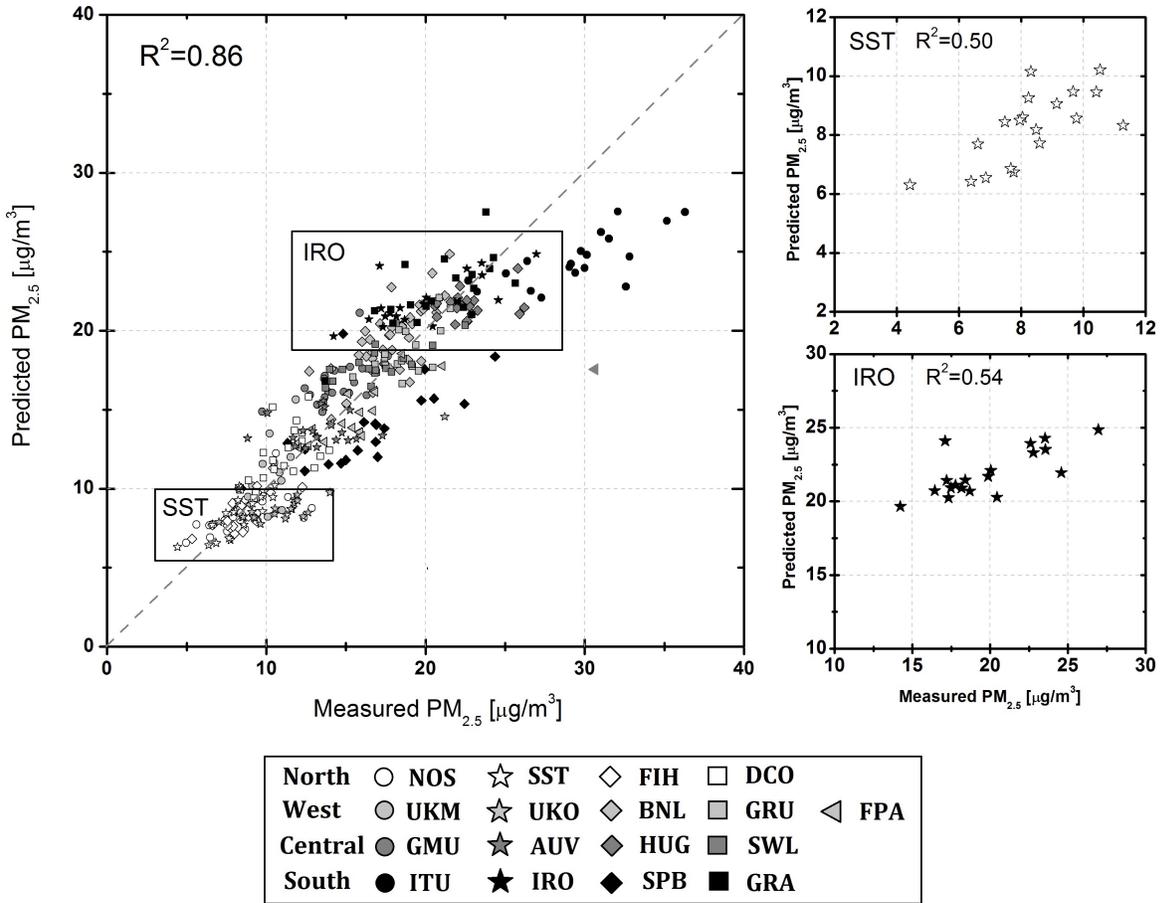


Figure 3.

