Western European Land Use Regression Incorporating Satellite- and Ground-Based Measurements of NO₂ and PM₁₀

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ABSTRACT: Land use regression (LUR) models typically investigate within-urban variability in air pollution. Recent improvements in data quality and availability, including satellite-derived pollutant measurements, support fine-scale LUR modeling for larger areas. Here, we describe NO₂ and PM₁₀ LUR models for Western Europe (years: 2005–2007) based on >1500 EuroAirnet monitoring sites covering background, industrial, and traffic environments. Predictor variables include land use characteristics, population density, and length of major and minor roads in zones from 0.1 km to 10 km, altitude, and distance to sea. We explore models with and without satellite-based NO₂ and PM₂.₅ as predictor variables, and we compare two available land cover data sets (global; European). Model performance (adjusted R²) is 0.48–0.58 for NO₂ and 0.22–0.50 for PM₁₀. Inclusion of satellite data improved model performance (adjusted R²) by, on average, 0.05 for NO₂ and 0.11 for PM₁₀. Models were applied on a 100 m grid across Western Europe; to support future research, these data sets are publicly available.

1. INTRODUCTION

Land use regression (LUR) has rapidly become a standard approach for estimating spatial variability in air pollution, for example during exposure assessment in epidemiological studies. Since the inception of LUR,¹ many studies have explored how well LUR can estimate within-city spatial variability in pollutant concentrations.²,³ Recent attention has focused on comparing LUR to other methods such as interpolation and dispersion modeling,⁴ applying LUR to specific constituents (e.g., soot) and elements of PM₂.₅,⁶ and specific organic compounds (e.g., PAHs);⁶,⁷ and evaluating the transferability of models to other spatial and temporal contexts.⁹–¹⁴

LUR models are often derived from measurements made specifically to build the LUR. An alternative approach is to employ data from existing monitors; this approach is well suited to modeling broad geographic extents. Examples include individual European countries,¹¹,¹⁵ continental USA,¹⁶,¹⁷ Canada,¹⁸ and Western Europe.¹⁹

Here we develop NO₂ and PM₁₀ LUR models for Western Europe. Only one Europe-wide LUR has previously been published.¹⁹ We improve on that investigation by offering 2 orders of magnitude improvement in spatial resolution (1 km² [prior¹⁹] versus 0.01 km² [here]) and by including

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satellite-derived estimates of ground-level air pollution. Investigations with large populations and geographic extents, including epidemiological studies of air pollution and traffic-related air pollution, environmental injustice studies, and health risk assessment, would benefit from continental-scale models with a finer spatial resolution.

We investigate whether satellite-derived pollution measurements improve fine-scale concentration estimates in European-wide LURs. Our approach incorporates GIS-derived land use, topographic data, and satellite-derived estimates of ground-level concentrations for NO₂ and PM₂.₅. We benefit from the large number of regulatory monitoring stations (EuroAirnet) operating in Western Europe, facilitating independent evaluation with reserved sites.

2. METHODS

We develop land use regression (LUR) models for Western Europe (17 contiguous countries; Figure 1). Our dependent variables are ambient concentrations of NO₂ and PM₁₀ obtained from regulatory monitoring. Our independent variables include several GIS-derived measures of land use and topography (100 m grids) and satellite-derived estimates of surface concentrations of NO₂ and PM₂.₅ (not PM₁₀). Despite the availability of satellite-derived PM₂.₅ estimates, there is an insufficient number of ground-based monitoring sites to...
support modeling PM$_{2.5}$. We next describe the input data and then our modeling approach.

2.1. Data. 2.1.1. Ground-Based Monitoring Data. We use annual mean NO$_2$ and PM$_{10}$ concentrations (years 2005–2007) from EuroAirnet, the regulatory air pollution monitoring network in Europe. EuroAirnet comprises sites from national networks and is publicly reported in AirBase (version 5). Annual measurements are excluded if a site captured <75% of the total hours (NO$_2$) or <50 daily AOD measurements over the 6 years were removed. Satellite-derived humidity-corrected PM$_{2.5}$ estimates for 2001–2006 were aggregated to improve accuracy by enabling sufficient data capture; estimates for grid cells with <50 daily AOD measurements over the 6 years were removed.

2.1.2. Satellite-Derived Estimates of Ground-Level Concentrations. We employ satellite-derived estimates of ground-level NO$_2$ and PM$_{2.5}$. Tropospheric NO$_2$ columns are from the OMI (Ozone Monitoring Instrument) instrument onboard the Aura satellite, Aerosol optical depth (AOD) retrieved from the MODIS (Moderate Resolution Imaging Spectroradiometer) and MISR (Multisite Imaging Spectroradiometer) instruments onboard the Terra satellite is used to estimate PM$_{2.5}$. As described elsewhere, satellite column-integrated retrievals were related to surface concentrations at 0.1° × 0.1° resolution (~10 km grid) using scaling factors interpolated from the GEOS-Chem chemical transport model (www.geos-chem.org) that account for the local vertical distribution and scattering properties of each pollutant. Annual satellite-derived estimates for NO$_2$ were made for years 2005, 2006, and 2007. Satellite-derived humidity-corrected PM$_{2.5}$ estimates for 2001–2006 were aggregated to improve accuracy by enabling sufficient data capture; estimates for grid cells with <50 daily AOD measurements over the 6 years were removed.

In Europe, PM$_{2.5}$ represents a large fraction (40–80%) of PM$_{10}$ mass in ambient air, motivating the use of satellite-derived PM$_{2.5}$ as an independent variable in a PM$_{10}$ LUR.

2.1.3. Predictor Variables. Predictor variables are integrated into a 100 m raster GIS database using ArcGIS10, employing the European reference grid (ETRS Lambert Azimuthal Equal Area S2 10). Satellite-derived pollution measurements and global land cover data are first resampled using nearest neighbor assignment; altitude is resampled using bilinear interpolation (used for continuous data). Variables, described below, are computed either as point estimates or zones. Zones of increasing radius (hereafter referred to as “buffers”) from 0.1 km to 10 km are computed using the Focalsum command with the circle option. Table 2 summarizes the predictor variables.

<table>
<thead>
<tr>
<th>data set</th>
<th>variable$^a$</th>
<th>code</th>
<th>buffer$^b$ or point estimate</th>
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<tbody>
<tr>
<td>OMI derived NO$_2$ (ppb): ~10 km</td>
<td>surface NO$_2$ concentration</td>
<td>SNO2</td>
<td>point</td>
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<tr>
<td>Terra derived PM$_{10}$ (μg/m$^3$): ~10 km</td>
<td>surface PM$_{10}$ concentration</td>
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<tr>
<td>Corine land cover (% area)</td>
<td>continuous urban fabric - high density</td>
<td>Hdr</td>
<td>buffer</td>
</tr>
<tr>
<td></td>
<td>discontinuous urban fabric - low density</td>
<td>Ldr</td>
<td></td>
</tr>
<tr>
<td></td>
<td>industry</td>
<td>Ind</td>
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<tr>
<td></td>
<td>ports</td>
<td>Port</td>
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<tr>
<td></td>
<td>urban green</td>
<td>Urbgr</td>
<td></td>
</tr>
<tr>
<td></td>
<td>total built up (Res + Ind + Port + transport infrastructure, airports, mines, dumps and construction sites)</td>
<td>Tbu</td>
<td></td>
</tr>
<tr>
<td></td>
<td>seminatural land</td>
<td>Nat</td>
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</tr>
<tr>
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<td>residential (Hdr + Ldr)</td>
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<tr>
<td>global land cover (% area)</td>
<td>impervious surface</td>
<td>Isurf</td>
<td>buffer</td>
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<td>tree canopy</td>
<td>Tree</td>
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<tr>
<td>EuroStreets roads (length in m)</td>
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<td>Majrd</td>
<td>buffer</td>
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<tr>
<td></td>
<td>minor roads</td>
<td>Mindr</td>
<td></td>
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<tr>
<td>modeled population (N)</td>
<td>population</td>
<td>Pop</td>
<td>buffer</td>
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<tr>
<td>topography: 90 m SRTM DTM</td>
<td>altitude - transformed$^d$</td>
<td>Talt</td>
<td>point</td>
</tr>
<tr>
<td>modeled distance to sea (m)</td>
<td>distance to sea - transformed$^e$</td>
<td>Tsea</td>
<td>point</td>
</tr>
<tr>
<td>coordinates (m)</td>
<td>X coordinates for 100 m cell centroids</td>
<td>Xcoord</td>
<td>point</td>
</tr>
</tbody>
</table>

$^a$Prespecified direction of effect is negative for: Urbgr, Nat, Tree, Talt, and Ycoord for both pollutants; and Tsea for PM$_{10}$. $^b$Buffer zones (m): 0; 100; 200; 300; 500; 600; 700; 800; 1000; 1200; 1500; 1800; 2000; 2500; 3000; 3500; 4000; 5000; 6000; 7000; 8000; 10000.

Corine classes: Hdr: class 111; Ldr: class 112; Ind: class 121; Port: class 123; Urbgr: class 141; Nat: class 311–423; Res: class 111–112. $^d$Transformed altitude is calculated as $\sqrt{(nalt/\max(nalt))}$, where $nalt = \text{altitude} – \min(\text{altitude})$. $^e$Transformed distance to sea is calculated as $\sqrt{\text{minimum distance}/\max(\text{minimum distance})}$. 


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basis of the 44 land classes available in Corine, we define six main groups, represented by individual classes (Hdr, Ldr, Ind, Port; see Table 2) or aggregations of classes (Urbrg, Nat). We define two additional classes based on further aggregation of the urban classes (Res, Tbu). For both data sets (European; global), the percent area within in each buffer is computed for each land cover category. Population counts per grid cell are based on the European Environment Agency 1 km² population density grid.34,35

We use the 1:10,000 EuroStreets digital road network (version 3.1, based on TeleAtlas MultiNet TM for year-2008) to derive road density variables. EuroStreets includes 9 road classes, which we aggregate into major roads (motorways, main roads, and other major roads) and minor roads (secondary and four types of local roads). Nonmotorized tracks and paths are excluded. We intersect the road data with a 100 m base polygon and then calculate total length per grid cell and for each buffer. Consistent traffic-volume data are not available for Europe.

We use altitude data from the SRTM Digital Elevation Database version 4.1.36 The resolution of the SRTM data is 3 arc second (approximately 90 m), with vertical error <1 m. SRTM is available for most of the study area, up to 60°N latitude. For northern Scandinavia we use 1 km resolution Topo30 data. Distance to sea, a measure of continentality, differentiates coastal from inland areas which are not, for example, influenced by coastal recirculation patterns and particulates from sea spray. We compute this variable as the distance between centroids of a 1 km grid and the open ocean 25 km offshore as defined by Corine land cover. Distance (in m) is then assigned to the 100 m grid using inverse distance weighed (1/d) interpolation. Interpolated distance was validated against direct calculation of distance to sea, using NEAR, at the monitoring sites (r = 99). Following Beelen et al.,19 we apply a nonlinear transformation to altitude and distance to sea (see Table 2). We also include X and Y coordinates for the cell centroids to reflect broad scale trends in background air pollution concentrations.6,11

2.2. Modeling Approach. LUR model development follows the ESCAPE supervised stepwise selection to derive the multiple linear regression equation.37,38 Monitoring data (dependent variable), which are log-normally distributed, are log-transformed prior to modeling. We exclude potential predictor variables with ≥90% null values. Univariate regressions of the natural logarithm (LN) of annual mean concentrations and all available potential predictors variables are first developed, and the predictor with the highest adjusted \( R^2 \) retained. In subsequent steps, the remaining predictor variables are evaluated in turn; the variable offering the highest increase in adj-\( R^2 \) is retained if (1) the coefficient conforms to the prespecified direction of effect (see Table 2), (2) each additional predictor variable increases the adj-\( R^2 \) by at least 0.01, and (3) the direction of effect for predictors already included in the model does not change. Post hoc variables with p-value >0.10 or variance inflation factor (VIF) >5 are removed.17 When required, post hoc “ring” (i.e., annulus) variables are calculated by differencing the component buffers, and the model is rerun to derive the final coefficients.11,38 We apply standard diagnostic tests for ordinary least-squares regression, including checks on the normality of residuals, heteroscedasticity, spatial autocorrelation of residuals using Moran’s I, and influential observations using Cook’s D.

For models testing the inclusion of satellite-based measurements, that predictor variable is forced into the model as the first variable, and the model is built according to the procedure above. Partial \( R^2 \) values are recomputed and reported after the final model is derived. Models are evaluated against the independent subset of 20% sites reserved for this purpose; \( R^2 \), root mean squared error (RMSE), error, and bias17 are reported here.

3. RESULTS

3.1. Measured Concentrations from Ground-Based Monitoring. Variability in annual mean \( \text{NO}_2 \) and \( \text{PM}_{10} \) concentrations measured at the Airbase monitoring sites is relatively consistent across the three years (Table 1). For both pollutants, the number of sites available for modeling (≥75% annual data capture) increases each year, owing to network growth, improvements in data capture, or both. The number of sites measuring continuously over the 3-year period is lower than the number of sites for any individual year (23% [17%] less for \( \text{NO}_2 \) [\( \text{PM}_{10} \), relative to 2005). Given the longer temporal period of the \( \text{PM}_{2.5} \) satellite data, we also include LUR models based on the 3-year average concentrations. For both pollutants, the largest share of monitoring sites, with ~100–400 each, are in Austria, Italy, Spain, Germany, and France (see Supplementary Table S1). Most countries have either a consistent number or experienced an increase in number of sites by year. Great Britain is an exception, with a 60% (30%) reduction in \( \text{NO}_2 \) (\( \text{PM}_{10} \)) site number in year-2007 relative to 2006. Spain also exhibits a dip in monitor numbers for both pollutants in 2006. Expansion in the network is greatest for Italy, with a 65% (86%) increase in \( \text{NO}_2 \) (\( \text{PM}_{10} \)) sites from 2005 to 2007. For both pollutants, Pearson’s correlation between the ground- and satellite-based measurements ranges from 0.33–0.37. The agreement between observed \( \text{PM}_{10} \) and satellite-derived \( \text{PM}_{2.5} \) is likely decreased by differences in sampling period, spatial representation, and aerosol size but is sufficient to suggest applicability as a LUR predictor. Correlation is higher with background sites, which are expected to be more representative of the larger area covered by each satellite grid cell. Scatterplots are in Supplementary Figures S1 and S2.

3.2. Model Comparison. Table 3 compares the models on the basis of coefficient of determination (\( R^2 \)), mean error, and bias. For both pollutants, models with satellite data outperformed the respective model without satellite data, achieving higher model building and evaluation \( R^2 \) and lower error and bias. Increases in adj-\( R^2 \) attributable to including satellite estimates are 0.02–0.06 for \( \text{NO}_2 \) and 0.07–0.13 for \( \text{PM}_{10} \). Selection of land cover data set (Corine vs global) yielded modest (at most 0.04) impacts to adj-\( R^2 \).

The addition of satellite data did not substantially alter the structure of the \( \text{NO}_2 \) models (Table 3): road and land cover variables remain largely unchanged; other variables (altitude, population density, and distance to sea) only enter the models when satellite data is not included. By comparison, the \( \text{PM}_{10} \) model structure is less stable both across and within years; a consistent pattern in variables entering models with and without satellite data is not apparent.

Model results are mapped in Figures 1 and 2 (models with satellite-derived pollution estimates) and Figures 3 and 4 (models without satellite-derived pollution estimates). For both pollutants, the models generally resolve expected patterns in air pollution, with higher concentrations in urban areas and near roadways. There are detectable differences, however, in the specific spatial patterns for cities (see map insets and profiles).
Table 3. Comparison of All Models<sup>d</sup>

<table>
<thead>
<tr>
<th>Year</th>
<th>Model</th>
<th>Variables&lt;sup&gt;c&lt;/sup&gt;</th>
<th>Adj-R&lt;sup&gt;2&lt;/sup&gt;</th>
<th>R&lt;sup&gt;2&lt;/sup&gt;</th>
<th>ME</th>
<th>MAE</th>
<th>MB</th>
<th>MAB</th>
<th>Variables&lt;sup&gt;c&lt;/sup&gt;</th>
<th>Adj-R&lt;sup&gt;2&lt;/sup&gt;</th>
<th>R&lt;sup&gt;2&lt;/sup&gt;</th>
<th>ME</th>
<th>MAE</th>
<th>MB</th>
<th>MAB</th>
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<tr>
<td></td>
<td>Corine SNO2-05, Minrd-1800, Nat-600, Majrd-100, Tbu-300, Minrd-1800–10000</td>
<td>0.58</td>
<td>0.56</td>
<td>−1.8</td>
<td>8.1</td>
<td>13</td>
<td>37</td>
<td>Tbu-2000, Minrd-400–10000, Nat-600, Majrd-100, Minrd-400</td>
<td>0.55</td>
<td>0.54</td>
<td>−1.7</td>
<td>8.8</td>
<td>16</td>
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<td>Global SNO2-05, Minrd-400–1800, Majrd-100, Tree-300, Minrd-400, Isurf-800</td>
<td>0.56</td>
<td>0.57</td>
<td>−1.6</td>
<td>8.0</td>
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<td>Minrd-500–2500, Majrd-100, Tree-700, Minrd-2500–10000, Minrd-500</td>
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<td>9.1</td>
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<td>8.3</td>
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<td>Global SPM, Ycoord, Minrd-200–2500, Talt, Minrd-200, Majrd-100</td>
<td>0.45</td>
<td>0.47</td>
<td>−0.5</td>
<td>4.6</td>
<td>4</td>
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<td>Tree-1200, Ycoord, Minrd-200–6000, Talt, Majrd-200, Majrd-100</td>
<td>0.40</td>
<td>0.37</td>
<td>−0.8</td>
<td>5.1</td>
<td>3</td>
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<td>2005–2007</td>
<td>Corine SPM, Ycoord, Nat-12000, Tsea, Pop-1800, Tbu-800, Majrd-100</td>
<td>0.48</td>
<td>0.48</td>
<td>−0.2</td>
<td>4.4</td>
<td>3</td>
<td>17</td>
<td>Nat-2000, Ycoord, Tbu-300–10000, Talt, Tbu-300</td>
<td>0.36</td>
<td>0.31</td>
<td>−0.2</td>
<td>4.4</td>
<td>3</td>
<td>17</td>
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<tr>
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<td>Global SPM, Ycoord, Isurf-200, Tree-600, Tsea, Majrd-100</td>
<td>0.48</td>
<td>0.48</td>
<td>−0.3</td>
<td>4.4</td>
<td>3</td>
<td>17</td>
<td>Tree-1000, Ycoord, Minrd-10000, Talt, Majrd-100</td>
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<td>0.34</td>
<td>−0.8</td>
<td>4.9</td>
<td>3</td>
<td>18</td>
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<sup>d</sup>Model building using natural logarithm of concentration (LN concentration).<sup>e</sup>Model evaluation using concentration ($\mu g/m^3$): ME = mean error ($\mu g/m^3$); MAE = mean absolute error ($\mu g/m^3$); MB = mean bias (%); MAB = mean absolute bias (%). 'Variables listed by order of entry into models, with satellite forced into the model as the first variable<sup>f</sup>.' The best models are shown in boldface.
because of differences in the overall structure of the models. At the European scale, the maps show that known hotspots with frequently elevated regional background levels (e.g., the Ruhr area, Po valley, and western Netherlands) are better captured in models that include satellite-derived pollution estimates. Table S2 presents model evaluation by region. A striking example from that table is for the Italy + Greece region (PM$_{10}$, $n = 309$ monitors), $R^2$ is 0.07 without satellite data, 0.45 with satellite data.

The sensitivity analysis of 80:20 subsets for annual models reveals that models are robust to changes in the evaluation subset; differences in adj-$R^2$ are slight (<0.02 for NO$_2$ and <0.04 for PM$_{10}$: see Table S3). Table S3 also shows the evaluation subset used to derive the models presented in Table 3. All models, for both pollutants, show no spatial autocorrelation in the residuals. Models based on 100% sites were similar in structure and performance (Tables S4 and S5). Including country indicators generally improved models, although not all indicators were statistically significant. Furthermore, to avoid the introduction of step changes in concentrations at country borders, we do not use country in our final models. Improvement was marked, up to ∼20%, for some of the PM$_{10}$ models. This improvement, in part, is likely attributed to differences in PM monitoring equipment and may also reflect differences in calibration$^{22}$ and of site selection of the various countries.

3.3. Final Models. NO$_2$. The best-performing NO$_2$ models by year are in Table S6. The variables in each NO$_2$ model are consistent across years: in addition to satellite-derived surface NO$_2$, all models include the length of minor roads in an intermediate buffer (1500 or 1800 m) and in the outer ring to 10 km, major road length in a 100 m buffer, and total built up land from Corine in a 300 m buffer. The models also all contain Corine seminatural land with a negative coefficient in a 500 or 600 m buffer. Minor roads in the intermediate buffer contribute 59−65% to the model predictive power (partial $R^2$: 0.3−0.4), followed by satellite-based NO$_2$ at 17−23% (partial $R^2$: 0.1). Those findings underscore the utility of satellite-based NO$_2$ concentrations for NO$_2$ LUR.

Overall, the final NO$_2$ models explain 55−60% of the variation in log-transformed NO$_2$ at the more than 400 reserved evaluation sites distributed across Europe (Table S8; Figure S5). Expressed and mapped as concentrations ($\mu$g/m$^3$), the explained variation is 50−56%. Error and bias are relatively similar across years, with highest error + bias in year-2007: error ($-1.3 - -1.8 \mu$g/m$^3$); absolute error (8.1−8.5 $\mu$g/m$^3$); mean bias (11−18%); and absolute bias (34−41%). Minor road length and satellite estimates of NO$_2$ are consistently the two most important predictors.

PM$_{10}$. The best-performing model for PM$_{10}$ by year is shown in Table S7. The variables in the final PM$_{10}$ models varied by year, with the global land cover models performing better than Corine in 2005 and 2007. All models contain satellite-based PM$_{2.5}$, the Y coordinate indicating the general decreasing trend in concentrations from south to north, and major roads in the intermediate buffer. As with NO$_2$ for PM$_{10}$ the satellite measurement is consistently the first or second variable to enter the model. Distance to sea enters all but the 2007 model, which instead has the altitude variable. The 2005, 2006, and 2005−2007 models include land cover classes representing both built

Figure 2. Map and profile plots of PM$_{10}$ concentration in 2007 using satellite data; scatterplot of modeled vs measured PM$_{10}$ at evaluation sites.
4. DISCUSSION

LUR models given here explain 46–56% (36–48%) of the variation in annual mean NO2 (PM10) concentration at independent sites. For both pollutants, satellite data are consistently the first or second variable into the model, and these data improve LUR model performance. Based on model R2, satellite data contribute more to the PM10 models than the NO2 models, despite the difference in particle sizes (using PM10 satellite data to model PM10 measurements). This finding is likely because the satellite data provide estimates averaged over a ~10 km grid and thus reflects regional background rather than local variations in concentrations. Compared to NO2, ambient concentrations of PM10 are much more affected by long-range transport; that transport is detected by the PM2.5 satellite data.

The overall performance of the NO2 model is better than for PM10, perhaps owing to other more local predictor variables, consistent with observations in the ESCAPE study.37,38 Furthermore, in the EU methodological consistency of monitoring is greater for NO2 (chemiluminescence) than PM10 (multiple methods). Recent spatiotemporal LURs for the USA reported an R2 of 0.78 for NO2,17 and 0.63 for PM2.5.16 As indicated by our models with country indicators (Tables S4 and S5) and the evaluation by region (Table S2), however, there are differences between countries which cannot be explained by the variables in our final PM10 models. This perhaps points to the need for regional models, especially for PM10.

We expect that meteorological conditions also play a role in PM10 model performance. In Europe, for example, 2006 was a year with several air pollution episodes including that associated with the July heat wave. Here, unlike in our previous work19,39 we did not specifically include coarse-scale meteorological variables. We took this a priori decision because the effects of meteorology are generally captured by the satellite-derived air pollution data, yet at a higher spatial resolution than for meteorological data. While daily meteorological variability is incorporated into the satellite-derived PM2.5 estimates, year-to-year variability, however, is not captured by the long-term mean (2001–2006) we use in the PM10 LUR models. If year-to-year model variation is in fact mainly driven by meteorological factors, model performance may benefit from including meteorological variables in the LUR models or, like NO2, using annual satellite data.

In general, the models described here exhibit comparable performance as previous LUR models at the European scale: Beelen et al.19 report validation R2s of 0.61 (0.45) for NO2 (PM10) using a hybrid Kriging-LUR approach. Our NO2 models may explain less of the variation in measured concentrations relative to the work of Beelen et al. in part because we model all site types, including traffic, rather than only background sites. We found that evaluation R2s for independent monitoring sites is very similar to the model R2, consistent with methodological work showing that model R2 can exceed independent evaluation R2s for small data sets, but less so for large data sets such as the ones we use here.40,41

An important next step for this research would be to model PM2.5, a pollutant which is subject to recent EU guideline limits43 and, based on the Global Burden of Disease estimates, is responsible for 3.2 million deaths and 76 million years of lost healthy life worldwide.42 Although site numbers for PM2.5 are slowly increasing, for this time period and study area, too few sites are available to derive reliable LUR models (146 and 195, respectively, in year-2005 and 2007 with sufficient annual data capture). A large fraction of the spatial variation of PM10 is related to variation of PM2.5. The ESCAPE study reported an average R2 between spatial variation of PM10 and PM2.5 of 0.74 (range 0.44–0.95).30

Modeling over large areas at fine spatial resolutions is an attractive solution for a variety of applications with large study populations, including health risk assessment. Given that LUR models generally cannot be directly transferred to other spatial domains,10,11,14 our approach addresses a particular need for reliable and consistent models at the continental level. From our models we estimate the mean population-weighted exposure in 2007 was 27 (25) μg/m3 for NO2 (PM10). Furthermore, we estimate that 9% (NO2) and 1% (PM10) of the European population reside in areas exceeding the annual guideline limit of 40 μg/m3 (current annual guidelines are the same for NO2 and PM10).23 Some caution is needed in interpretation of these results given differences in model performance by region (Table S2). These regional differences in model performance may in part be attributable to known deficiencies in the monitoring network (uneven distribution and clustering of sites in EuroAirenet, which is an assembly of sites from existing country networks; use of different PM10 monitoring methods and correction factors by country) or discrepancies in the definition of land cover or road classes across Europe.43

There are several challenges in producing suitable models for air pollution exposure assessment across large areas. We aim here for models at a spatial scale fine enough to estimate within-city and near-roadway contrasts in pollution while also accounting for long-range transport and other large scale variability. Most studies evaluating exposures over large areas use a vector-LUR approach whereby estimates are then made at census centroids, a coarse mesh, or home addresses; should a map or estimates at additional locations be required, interpolation is then used to produce a continuous surface.17,18,44−46 A strength of our models is that we take a raster-based LUR approach, which enables direct prediction at the 100 m grid (Figure 1 and 2). We thus eliminate the need for interpolation which can oversmooth estimates. In this study, a 100 m resolution is justified given the quality and resolution of the source information as well as the dense network of monitoring sites distributed in different exposure environments across Europe. Although not always reflected in the R2 as a performance measure, this attribute (large number of monitors, located in diverse environments) is an important advance over the previous models for Europe.19,39
As was previously demonstrated in Canada and the USA, we show here that combining LUR models with worldwide, satellite-based pollution measurements can offer improved continental-scale exposure models for Europe. To support future research, model results are publicly available.

## ASSOCIATED CONTENT

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<th>Supporting Information</th>
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<tr>
<td>Table S1: Number of monitoring sites by year. Table S2: Evaluation statistics by regions for best models (based on concentration (µg/m³)). Table S3: Sensitivity analysis - NO2 models based on all monitoring sites. Table S4: Sensitivity analysis - PM2.5 models based on all monitoring sites. Table S5: Summary of model building and evaluation statistics. Figure S1: Measured ground-based NO2 vs satellite-derived NO2, at all monitoring sites. Figure S2: Measured ground-based PM10 vs mean 2001–2006 satellite-derived PM2.5, at all monitoring sites. Figure S3: Map and profile plots of NO2 concentration in 2005 without satellite data; scatterplot of modeled vs measured NO2 at evaluation sites. Figure S4: Map and profile plots of PM2.5 concentration in 2007 without satellite data; scatterplot of modeled vs measured PM10 at evaluation sites. Figure S5: Modeled vs measured NO2 concentration (µg/m³) at evaluation sites for final models. Figure S6: Modeled vs measured PM10 concentration (µg/m³) at evaluation sites for final models. This material is available free of charge via the Internet at <a href="http://pubs.acs.org">http://pubs.acs.org</a>.</td>
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**Notes**

The authors declare no competing financial interest.

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