Western European land use regression incorporating satellite- and ground-1 based measurements of NO₂ and PM₁₀ 2

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- 29 per large table/figure.)
- 30

31 Abstract

- 32 Land use regression (LUR) models typically investigate within-urban variability in air pollution.
- 33 Recent improvements in data quality and availability, including satellite-derived pollutant
- 34 measurements, support fine-scale LUR modelling for larger areas. Here, we describe NO_2 and PM_{10}
- LUR models for Western Europe (years: 2005–2007) based on >1500 EuroAirnet monitoring sites
- 36 covering background, industrial, and traffic environments. Predictor variables include land use
- 37 characteristics, population density, and length of major and minor roads in zones from 0.1km to
- 10 km, altitude, and distance to sea. We explore models with and without satellite-based NO₂ and
- 39 PM_{2.5} as predictor variables and we compare two available land cover datasets (global; European).
- 40 Model performance (adjusted R^2) is 0.48–0.58 for NO₂ and 0.22–0.50 for PM₁₀. Inclusion of satellite
- 41 data improved model performance (adjusted R^2) by, on average, 0.05 for NO₂ and 0.11 for PM₁₀.
- 42 Models were applied on a 100m grid across Western Europe; to support future research, these datasets
- 43 are publicly available.

44

45 **1. Introduction**

- 46 Land use regression (LUR) has rapidly become a standard approach for estimating spatial variability
- 47 in air pollution, for example during exposure assessment in epidemiological studies. Since the
- 48 inception of LUR,¹ many studies have explored how well LUR can estimate within-city spatial
- 49 variability in pollutant concentrations.^{2, 3} Recent attention has focused on comparing LUR to other
- 50 methods such as interpolation and dispersion modelling;^{4, 5} applying LUR to specific constituents
- 51 (e.g., soot) and elements of $PM_{2.5}^{6,7}$ and specific organic compounds (e.g., PAHs);^{3, 8} and, evaluating
- 52 the transferability of models to other spatial and temporal contexts.⁹⁻¹⁴
- 53 LUR models are often derived from measurements made specifically to build the LUR. An alternative
- 54 approach is to employ data from existing monitors; this approach is well suited to modelling broad
- 55 geographic extents. Examples include individual European countries,^{11, 15} continental USA,^{16, 17}
- 56 Canada,¹⁸ and Western Europe.¹⁹

57 Here we develop NO₂ and PM₁₀ LUR models for Western Europe. Only one Europe-wide LUR has

- 58 previously been published.¹⁹ We improve on that investigation by offering two orders of magnitude
- improvement in spatial resolution $(1 \text{km}^2 \text{ [prior}^{19} \text{] versus } 0.01 \text{km}^2 \text{ [here]})$, and by including satellite-
- 60 derived estimates of ground-level air pollution. Investigations with large populations and geographic
- 61 extents, including epidemiological studies of air pollution and traffic-related air pollution,
- 62 environmental injustice studies, and health risk assessment, would benefit from continental-scale
- 63 models with a finer spatial resolution.
- 64 We investigate whether satellite-derived pollution measurements improve fine-scale concentration
- 65 estimates in European-wide LURs. Our approach incorporates GIS-derived land use, topographic
- 66 data, and satellite-derived estimates of ground-level concentrations for NO₂ and PM_{2.5}. We benefit
- 67 from the large number of regulatory monitoring stations (EuroAirnet) operating in Western Europe,
- 68 facilitating independent evaluation with reserved sites.

69 **2. Methods**

- 70 We develop land use regression (LUR) models for Western Europe (17 contiguous countries; Figure
- 1). Our dependent variables are ambient concentrations of NO_2 and PM_{10} , obtained from regulatory
- 72 monitoring. Our independent variables include several GIS-derived measures of land use and
- topography (100m grids) and satellite-derived estimates of surface concentrations of NO₂ and PM_{2.5}
- 74 (not PM₁₀; despite the availability of satellite-derived PM_{2.5} estimates, there is an insufficient number
- of ground-based monitoring sites to support modelling $PM_{2.5}$). We next describe the input data and
- then our modelling approach.

77 **2.1. Data**

78 2.1.1. Ground-based monitoring data

- We use annual mean NO_2 and PM_{10} concentrations (years 2005–2007) from EuroAirnet, the
- 80 regulatory air pollution monitoring network in Europe. EuroAirnet comprises sites from national
- 81 networks²⁰ and is publicly reported in AirBase (version 5).²¹ NO₂ is monitored by chemiluminescence.
- 82 PM₁₀ is monitored by various methods including Tapered Element Oscillating Microbalance (TEOM),
- 83 Beta Attenuation, and Gravimetric methods.²² The network includes "background", "industrial", and
- 84 "traffic" sites; all site types are included here. Urban background sites are representative of the
- 85 exposure of the general urban population while rural background are sited away from major sources
- 86 of air pollution.²³ Annual measurements are excluded if a site captured <75% of the total hours (NO₂)
- 87 or days (PM₁₀). Table 1 presents summary statistics for retained monitoring sites. For each year,
- 88 monitoring data are randomly stratified (by country and site type) into five groups, each with 20% of
- sites. Subset 1 (20%) is used for model evaluation; the remaining four subsets (80%) are combined
- 90 and used for model building. As a sensitivity analysis, we apply a five-fold cross-validation procedure
- 91 in which the 20% evaluation subset is rotated, thereby creating four additional models. We *a priori*
- 92 designate the first subset to model evaluation, reverting to the next subset only if spatial
- autocorrelation is detected. We further evaluate models developed using 100% of the monitoring sites,
- and undertake a sensitivity analysis including country to investigate potential differences in the
- 95 national networks comprising AirBase.
- 96 <<Table 1. Summary statistics for mean annual concentrations ($\mu g/m^3$) at all monitoring sites with 97 \geq 75% annual data capture >>
- 98 2.1.2. Satellite-derived estimates of ground-level concentrations
- 99 We employ satellite-derived estimates of ground-level NO_2^{17} and $PM_{2.5}^{24}$ Tropospheric NO_2 columns
- are from the OMI (Ozone Monitoring Instrument) instrument onboard the Aura satellite.²⁵ Aerosol
- 101 optical depth (AOD) retrieved from the MODIS (Moderate Resolution Imaging Spectroradiometer)²⁶
- and MISR (Multiangle Imaging Spectroradiometer)²⁷ instruments onboard the Terra satellite is used to
- estimate $PM_{2.5}$. As described elsewhere, ^{17, 24, 28} satellite column-integrated retrievals were related to
- 104 surface concentrations at $0.1^{\circ} \times 0.1^{\circ}$ resolution (~10km grid) using scaling factors interpolated from the
- 105 GEOS-Chem chemical transport model (www.geos-chem.org) that account for the local vertical
- 106 distribution and scattering properties of each pollutant. Annual satellite-derived estimates for NO₂
- 107 were made for years 2005, 2006 and 2007. Satellite-derived humidity-corrected PM_{2.5} estimates for
- 108 2001–2006 were aggregated to improve accuracy by enabling sufficient data capture; estimates for
- 109 grid cells with <50 daily AOD measurements over the 6 years were removed.²⁴ In Europe, PM_{2.5}
- represents a large fraction (40–80%) of PM_{10} mass in ambient air,^{29, 30} motivating the use of satellite-
- 111 derived $PM_{2.5}$ as an independent variable in a PM_{10} LUR.

112 2.1.3. Predictor variables

Predictor variables are integrated into a 100m raster GIS database using ArcGIS10, employing the European reference grid (ETRS Lambert Azimuthal Equal Area 52 10). Satellite-derived pollution measurements and global land cover data are first resampled using nearest neighbour 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 0.1km to 10km are computed using the Focalsum command with the circle option. Table 2 summarises the predictor variables.

120 <-<Table 2. GIS predictor variables>>

121 Two land cover datasets are available: the 100m European Corine Land Cover³¹ and coarser global

datasets including 500m tree canopy³² and 1km impervious surfaces.³³ On the basis of the 44 land

123 classes available in Corine, we define six main groups, represented by individual classes (Hdr, Ldr,

124 Ind, Port; see Table 2) or aggregations of classes (Urbgr, Nat). We define two additional classes based

125 on further aggregation of the urban classes (Res, Tbu). For both datasets (European; global), the

126 percent area within in each buffer is computed for each land cover category. Population counts per

127 grid cell are based on the European Environment Agency 1km² population density grid.^{34,35}

128 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

131 (secondary and four types of local roads). Non-motorised tracks and paths are excluded. We intersect

the road data with a 100m base polygon, then calculate total length per grid cell and for each buffer.

133 Consistent traffic-volume data are not available for Europe.

134 We use altitude data from the SRTM Digital Elevation Database version 4.1.³⁶ The resolution of the

135 SRTM data is 3 arc second (approx. 90m), with vertical error <16m. SRTM is available for most of

the study area, up to 60°N latitude. For northern Scandinavia we use 1km resolution Topo30 data.

137 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 1km grid and the open ocean 25km offshore as

140 defined by Corine land cover. Distance (in m) is then assigned to the 100m grid using inverse distance

- 141 weighed (1/d) interpolation. Interpolated distance was validated against direct calculation of distance
- 142 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.^{9, 11}

145 **2.2. Modelling Approach**

- 146 LUR model development follows the ESCAPE supervised stepwise selection to derive the multiple
- 147 linear regression equation.^{37, 38} Monitoring data (dependent variable), which are log-normally
- 148 distributed, are log-transformed prior to modelling. We exclude potential predictor variables with
- 149 >=90% null values. Univariate regressions of the natural logarithm (LN) of annual mean
- 150 concentrations and all available potential predictors variables are first developed, and the predictor
- 151 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
- 153 conforms to the pre-specified 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)
- 156 >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
- 158 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
- 160 influential observations using Cook's D.
- 161 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,
- 165 and bias¹⁷ are reported here.

166 **3. Results**

167 3.1. Measured concentrations from ground-based monitoring

- 168 Variability in annual mean NO_2 and PM_{10} concentrations measured at the Airbase monitoring sites is
- 169 relatively consistent across the three years (Table 1). For both pollutants, the number of sites available
- for modelling (\geq 75% annual data capture) increases each year, owing to network growth,
- 171 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 $(23\% [17\%] less for NO_2 [PM_{10}],$
- 173 relative to 2005). Given the longer temporal period of the $PM_{2.5}$ satellite data, we also include LUR
- 174 models based on the 3-year average concentrations. For both pollutants, the largest share of
- 175 monitoring sites, with ~100–400 each, are in Austria, Italy, Spain, Germany and France (see
- 176 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 NO₂ (PM_{10})
- 178 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 NO₂
- 180 (PM_{10}) sites from 2005 to 2007. For both pollutants, Pearson's correlation between the ground- and
- 181 satellite-based measurements ranges from 0.33-0.37. The agreement between observed PM₁₀ and
- satellite-derived 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 Figure S1 and S2.

186 **3.2. Model comparison**

- 187 Table 3 compares the models on the basis of coefficient of determination (R^2) , mean error, and bias.
- 188 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
- 190 attributable to including satellite estimates are 0.02-0.06 for NO₂, 0.07-0.13 for PM₁₀. Selection of
- 191 land cover dataset (Corine vs. global) yielded modest (at most 0.04) impacts to $adj-R^2$.
- **192** The addition of satellite data did not substantially alter the structure of the NO₂ 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 PM_{10}
- model structure is less stable both across and within years; a consistent pattern in variables entering
- 196 models with and without satellite data is not apparent.
- 197 <-<Table 3. Comparison of all models>>
- 198 Model results are mapped in Figure 1 and 2 (models with satellite-derived pollution estimates) and
- 199 Figure S3 and S4 (models without satellite-derived pollution estimates). For both pollutants, the
- 200 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
- 202 (see map insets and profiles), because of differences in the overall structure of the models. At the
- 203 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
- 205 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.
- 208 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 NO₂ and <0.04 for PM₁₀; see Table
- S3). Table S3 also shows the evaluation subset used to derive the models presented in Table 3. All
- 211 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
- 213 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 $\sim 20\%$, for some of the PM₁₀ models. This
- 216 improvement in part is likely attributed to differences in PM monitoring equipment, but also reflects
- 217 differences in calibration²² and of site selection of the various countries.
- << Figure 1. Map and profile plots of NO₂ concentration in 2005 using satellite data; scatterplot of
 modelled vs. measured NO₂ at evaluation sites >>
- 220 << Figure 2. Map and profile plots of PM_{10} concentration in 2007 using satellite data; scatterplot of 221 modelled vs. measured PM_{10} at evaluation sites >>
- 222 **3.3. Final models**
- 223 NO₂
- The best-performing NO_2 models by year are in Table S6. The variables in each NO_2 model are
- 225 consistent across years: in addition to satellite-derived surface NO₂, all models include the length of
- 226 minor roads in an intermediate buffer (1500 or 1800m) and in the outer ring to 10km, major road
- length in a 100m buffer, and total built up land from Corine in a 300m buffer. The models also all
- contain Corine semi-natural land with a negative coefficient in a 500 or 600m 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 NO₂ at 17–23% (partial R^2 : 0.1). Those findings underscore the utility of
- satellite-based NO₂ concentrations for NO₂ LUR.
- 232 Overall, the final NO₂ models explain 55-60% of the variation in log-transformed NO₂ 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 $(-1.3 -1.8 \,\mu\text{g/m}^3)$; absolute error
- 236 $(8.1-8.5 \ \mu g/m^3)$; 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.

238 PM₁₀

- The best-performing model for PM_{10} by year is shown in Table S7. The variables in the final PM_{10}
- 240 models varied by year, with the global land cover models performing better than Corine in 2005 and
- 241 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 immediate buffer. As with NO_{2} ,
- for PM₁₀ 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 up areas and remote areas.
- 246 The structure of the 2007 model is rather different, and includes minor roads in both a local (200m)
- and intermediate (200–2500m) buffer, and percent tree canopy as the only land cover variable.
- 248 Satellite-based $PM_{2.5}$ and the Y coordinate each contribute ~30–35% to the model predictive power in
- each year (see Table S7; partial R^2 : 0.1–0.2).
- 250 Based on model R^2 , the year-2007 model explains ~47% of the variation in measured concentrations
- $(\sim 50\%)$ of the variation in log-transformed NO₂) at the sites reserved for model evaluation; models for
- earlier years explain 38–44% of variation in measured concentrations (Table S8; Figure S6). Error and
- bias are relatively similar across years, with lower error and bias in later years: error (-0.2 -1.2)
- $\mu g/m^3$; absolute error (4.4– 6.0 $\mu g/m^3$); mean bias (3–5%); absolute bias (17–22%).

255 **4. Discussion**

- LUR models given here explain 46–56% (36–48%) of the variation in annual mean NO_2 (PM₁₀)
- 257 concentration at independent sites. For both pollutants, satellite data are consistently the first or
- second variable into the model, and those data improve LUR model performance. Based on model R^2 ,
- satellite data contribute more to the PM_{10} models than the NO₂ models, despite the difference in
- 260 particle sizes (using $PM_{2.5}$ satellite data to model PM_{10} measurements). This finding is likely because
- the satellite data provide estimates averaged over a ~10km grid, and thus reflects regional background
- rather than local variations in concentrations. Compared to NO_2 , ambient concentrations of PM_{10} are
- much more affected by long-range transport; that transport is detected by the $PM_{2.5}$ satellite data.
- 264 The overall performance of the NO_2 model is better than for PM_{10} , 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 NO_2 (chemiluminescence) than PM_{10}
- 267 (multiple methods). Recent spatiotemporal LURs for the USA reported an R^2 of 0.78 for NO₂¹⁷ and
- 268 0.63 for PM_{2.5}.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 PM_{10} models. This perhaps points to the need for regional
- 271 models, especially for PM_{10} .
- 272 We expect that meteorological conditions also play a role in PM_{10} 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 work,^{19,39} we did not specifically include coarse-scale
- 275 meteorological variables. We took this *a priori* decision because the effects of meteorology are
- 276 generally captured by the satellite-derived air pollution data, yet at a higher spatial resolution than for
- 277 meteorological data. While daily meteorological variability is incorporated into the satellite-derived

- 278 PM_{2.5} estimates, year-to-year variability, however, is not captured by the long-term mean (2001–
- 279 2006) we use in the PM_{10} LUR models. If year-to-year model variation is in fact mainly driven by
- 280 meteorological factors, model performance may benefit from including meteorological variables in
- the LUR models or, like NO₂, using annual satellite data.
- 282 In general, the models described here exhibit comparable performance as previous LUR models at the
- European scale: Beelen et al.,¹⁹ report validation R^2 s of 0.61 (0.45) for NO₂ (PM₁₀) using a hybrid
- 284 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 R^2s for independent monitoring
- sites is very similar to the model R^2 , consistent with methodological work showing that model R^2 can
- exceed independent evaluation R^2s for small datasets, but less so for large datasets such as the ones
- $289 \qquad \text{we use here.}^{40, 41}$
- An important next step for this research would be to model $PM_{2.5}$, a pollutant which is subject to
- recent EU guideline limits²³ 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
- 293 PM_{2.5} 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
- 295 capture). A large fraction of the spatial variation of PM_{10} is related to variation of $PM_{2.5}$. The
- ESCAPE study reported an average R^2 between spatial variation of PM_{10} and $PM_{2.5}$ of 0.74 (range
- **297** 0.44–0.95).³⁰
- 298 Modelling 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
- 300 generally cannot be directly transferred to other spatial domains,^{10, 11, 14} our approach addresses a
- 301 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) μ g/m³ for NO₂ (PM₁₀).
- Furthermore, we estimate that 9% (NO₂) and 1% (PM₁₀) of the European population reside in areas
- 304 exceeding the annual guideline limit of 40 μ g/m³ (current annual guidelines are the same for NO₂ and
- PM_{10}).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
- 308 sites in EuroAirnet, which is an assembly of sites from existing country networks; use of different
- PM_{10} monitoring methods and correction factors by country) or discrepancies in the definition of land
- 310 cover or road classes across Europe.⁴³
- There are several challenges in producing suitable models for air pollution exposure assessmentacross 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 in pollution. Most studies evaluating exposures over large areas use a vector-LUR
- 315 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
- 317 continuous surface.^{17, 18, 44-46} A strength of our models is that we take a raster-based LUR approach,
- 318 which enables direct prediction at the 100m grid (Figure 1 and 2). We thus eliminate the need for
- interpolation which can over-smooth estimates. In this study, a 100m resolution is justified given the
- 320 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 R^2
- 322 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}
- 324
- As was previously demonstrated in Canada¹⁸ and the USA,^{16, 17, 47} we show here that combining LUR
- 326 models with worldwide, satellite-based pollution measurements can offer improved continental-scale
- 327 exposure models for Europe. To support future research, model results are publicly available.

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- for assistance in obtaining and processing the global land use data. We acknowledge the free use of
- tropospheric NO₂ column data from the OMI sensor via <u>www.temis.nl</u>.

334 Supporting Information Available

- Table S1: Number of monitoring sites by year. Table S2: Evaluation statistics by regions for best
- models (based on concentration ($\mu g/m^3$)). Table S3: Sensitivity analysis rotating 20% evaluation
- subsets. Table S4: Sensitivity analysis NO₂ models based on all monitoring sites. Table S5:
- 338 Sensitivity analysis PM₁₀ models based on all monitoring sites. Table S6: Final NO₂ models by year.
- Table S7: Final PM_{10} models by year. Table S8: Summary of model building and evaluation statistics.
- Figure S1: Measured ground-based NO₂ vs. satellite-derived NO₂, at all monitoring sites. Figure S2:
- 341 Measured ground-based PM_{10} vs. mean 2001-2006 satellite-derived $PM_{2.5}$, at all monitoring sites.
- 342 Figure S3: Map and profile plots of NO₂ concentration in 2005 without satellite data; scatterplot of
- modelled vs. measured NO₂ at evaluation sites. Figure S4: Map and profile plots of PM_{10}
- 344 concentration in 2007 without satellite data; scatterplot of modelled vs. measured PM₁₀ at evaluation
- sites. Figure S5: Modelled vs. measured NO₂ concentration ($\mu g/m^3$) at evaluation sites for final
- models. Figure S6: Modelled vs. measured PM_{10} concentration ($\mu g/m^3$) at evaluation sites for final
- 347 models. This information is available free of charge via the Internet at http://pubs.acs.org/.

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Table 1. Summary statistics for mean annual concentrations ($\mu g/m^3$) at all monitoring sites with $\geq 75\%$ annual data capture

Year	Ν	Min	5%	95%	Max	Mean	SD	GM	GSD			
NO ₂												
2005	2010	0.8	7.1	60.8	112.3	29.3	16.5	24.5	1.9			
2006	2099	0.9	7.9	61.8	121.3	29.8	16.8	25.1	1.9			
2007	2236	0.3	7.5	58.7	106.5	28.8	15.9	24.3	1.9			
2005-2007	1670	0.9	8.0	57.9	108.5	28.5	15.5	24.2	1.9			
PM ₁₀												
2005	1487	7.8	14.8	44.9	70.9	26.6	9.2	25.2	1.4			
2006	1584	7.7	15.7	45.7	71.7	27.7	9.2	26.3	1.4			
2007	1664	3.6	15.2	44.1	77.4	26.7	8.7	25.4	1.4			
2005-2007	1151	7.7	16.1	43.5	61.7	26.7	8.3	25.5	1.4			

GM = geometric mean; GSD = geometric standard deviation (unit less)

Table 2. GIS predictor variables

Dataset	Variable ^a	Code	Buffer ^b or
			point estimate
OMI derived NO ₂ (ppb): \sim 10km	Surface NO ₂ concentration	SNO2	Point
Terra derived PM _{2.5} (μ g/m ³): ~10km	Surface PM _{2.5} concentration	SPM	Point
Corine land cover ^c	Continuous urban fabric - high density	Hdr	Buffer
(% area)	Discontinuous urban fabric - low density	Ldr	
	Industry	Ind	
	Ports	Port	
	Urban green	Urbgr	
	Total built up (Res + Ind + Port + transport	Tbu	
	infrastructure, airports, mines, dumps and		
	construction sites)		
	Semi-natural land	Nat	
	Residential (Hdr + Ldr)	Res	
Global land cover	Impervious surface	Isurf	Buffer
(% area)	Tree canopy	Tree	
EuroStreets roads	Major roads	Majrd	Buffer
(length in m)	Minor roads	Minrd	
Modelled Population (N)	Population	Рор	Buffer
Topography: 90m SRTM DTM	Altitude - transformed ^d	Talt	Point
Modelled distance to sea (m)	Distance to sea - transformed ^e	Tsea	Point
Coordinates (m)	XY coordinates for 100m cell centroids	Xcoord	Point
		Ycoord	

a. Pre-specified direction of effect is negative for: Urbgr, Nat, Tree, Talt and Ycoord for both pollutants; and Tsea for PM_{10} b. "Buffer" zone distances (m): 0; 100; 200; 300; 400; 500; 600; 700; 800; 1000; 1200; 1500; 1800; 2000; 2500; 3000; 3500; 4000; 5000; 6000; 7000; 8000; 10000

c. Original Corine classes: Hdr: class 111; Ldr: class 112; Ind: class 121; Port: class 123; Urbgr: class 141-142; Tbu: class 111-133; Nat: class 311-423; Res: class 111-112

d. Transformed Altitude is calculated as $\sqrt{(nalt/max(nalt))}$, where nalt=altitude-min(altitude)

e. Transformed Distance to sea is calculated as $\sqrt{(minimum distance/max(minimum distance))}$

Table 3. Comparison of all models

Year	ear Model With Satellite						Without Satellite								
		Model ^a		Evalu	iation ^b				Model ^a		Evalu	lation ^b			
		Variables ^c	Adj-R ²	R ²	ME	MAE	MB	MAB	Variables ^c	Adj-R ²	R ²	ME	MAE	MB	MAB
NO ₂ M	lodels														
2005	Corine	SNO2-05, Minrd-1800, Nat-600, Majrd-100, Tbu-300, Minrd-1800-10000	0.58	0.56	-1.8	8.1	13	37	Tbu-2000, Minrd-400-10000, Nat_600, Majrd-100, Minrd-400	0.55	0.54	-1.7	8.8	16	42
	Global	SNO2-05, Minrd-400-1800, Majrd-100, Tree-300, Minrd-400, Isurf-800	0.56	0.57	-1.6	8.0	13	37	Minrd-500-2500, Majrd-100, Tree-700, Minrd-2500-10000, Minrd-500	0.51	0.51	-1.6	9.1	18	44
2006	Corine	SNO2-06, Minrd-1500, Majrd-100, Nat-500, Minrd-1500-10000, Tbu-300	0.55	0.50	-1.5	8.3	11	35	Tbu-1500, Minrd-100-10000, Nat-500, Majrd-100, Minrd-100, Pop-1000	0.53	0.47	-1.6	8.5	11	36
	Global	SNO2-06, Minrd-200-1500, Majrd-100, Isurf-500, Minrd-200, Tree-700	0.54	0.50	-1.0	8.3	13	36	Majrd-100, Minrd-200, Minrd-200-10000, Tree-700, Isurf-500, Tsea	0.49	0.46	-1.1	8.9	14	39
2007	Corine	SNO2-07, Minrd-1500, Majrd-100, Nat-600, Tbu-300, Minrd-1500-10000,	0.55	0.50	-1.3	8.5	18	41	Tbu-1200, Minrd-200-10000, Nat-600, Majrd-100, Minrd-200	0.51	0.48	-1.6	8.8	19	43
	Global	SNO2-07, Minrd-300-1500, Majrd-100, Tree-500, Minrd-300, Isurf-700	0.54	0.54	-1.4	8.1	20	43	Minrd-400-2500, Majrd-100, Tree-800, Minrd-2500-10000, Minrd-400, Talt, Tsea	0.48	0.46	-1.8	9.1	23	49
2005- 2007	Corine	SNO2-05-07, Minrd-1500, Nat-600, Majrd-100, Minrd-1500-10000, Tbu-300	0.60	0.46	-1.8	8.4	8	34	Tbu-2000, Minrd-200-10000, Nat-600, Majrd-100, Minrd-200	0.56	0.48	-1.8	8.3	10	35
	Global	SNO2-05-07, Minrd-400-1500, Majrd-100, Tree-700, Isurf-500, Minrd-400	0.58	0.50	-1.5	8.0	11	34	Minrd-500-2500, Majrd-100, Tree-800, Minrd-2500-10000, Minrd-500	0.52	0.41	-1.2	8.6	14	38
PM ₁₀ N	Aodels														
2005	Corine	SPM, Ycoord, Hdr-1200, Nat-500, Ind-100, Tsea	0.35	0.37	-0.7	5.7	5	22	Ycoord, Tbu-10000, Nat-1000	0.22	0.25	-1.1	6.1	4	23
	Global	SPM, Ycoord, Isurf-1000, Tree-500, Tsea, Majrd	0.35	0.44	-0.8	5.4	4	21	Tree-800, Ycoord, Isurf-1000-10000, Isurf-1000	0.22	0.25	-1.1	6.0	4	23
2006	Corine	SPM, Ycoord, Tbu-600, Pop-1800, Tsea, Majrd-100, Nat_600	0.37	0.36	-1.2	6.0	3	22	Pop-1800, Ycoord, Nat-1000, Talt, Majrd-100, Ldr-10000	0.25	0.25	-1.9	6.4	1	23
	Global	SPM, Ycoord, Isurf-800, Tree-100, Majrd-100	0.36	0.35	-1.5	6.0	2	22	Tree-1000, Ycoord, Isurf-1000, Majrd-100, Talt	0.24	0.20	-1.8	6.6	2	24
2007	Corine	SPM, Ycoord, Minrd-2500, Talt, Tbu-100	0.49	0.45	-0.6	4.8	3	18	Ycoord, Nat-2000, Minrd-10000, Tbu-100, Talt, Majrd	0.42	0.36	-0.5	5.2	4	19
	Global	SPM, Ycoord, Minrd-200-2500, Talt, Minrd-200, Majrd, Tree-100	0.50	0.47	-0.5	4.6	4	17	Tree-1200, Ycoord, Minrd-200-6000, Talt, Minrd-200, Majrd-100	0.40	0.37	-0.8	5.1	3	19
2005- 2007	Corine	SPM, Ycoord, Nat-1200, Tsea, Pop-1800, Tbu-400, Majrd-100	0.48	0.48	-0.2	4.4	3	17	Nat-2000, Ycoord, Tbu-300-10000, Talt, Tbu-300	0.36	0.31	-0.2	4.4	3	17
	Global	SPM, Ycoord, Isurf-200, Tree-600, Tsea, Majrd-100	0.48	0.48	-0.3	4.4	3	17	Tree-1000, Ycoord, Minrd-10000, Talt, Majrd-100	0.36	0.34	-0.8	4.9	3	18

Best models shaded grey a. Model building using natural logarithm of concentration (LN concentration) b. Model evaluation using concentration (μ g/m³): ME = mean error (μ g/m³); MAE = mean absolute error (μ g/m³); MB = mean bias (%); MAB = mean absolute bias (%) c. Variables listed by order of entry into models, with satellite forced into the model as the first variable



Figure 1. Map and profile plots of NO₂ concentration in 2005 using satellite data; scatterplot of modelled vs. measured NO₂ at evaluation sites



Figure 2. Map and profile plots of PM₁₀ concentration in 2007 using satellite data; scatterplot of modelled vs. measured PM₁₀ at evaluation sites

Supporting Information

Western European land use regression incorporating satellite- and ground-based measurements of NO_2 and PM_{10}

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		NO ₂	Sites		PM ₁₀ Sites					
Country	2005	2006	2007	2005- 2007	2005	2006	2007	2005- 2007		
AT	146	145	151	138	107	109	127	98		
BE	56	57	62	48	39	39	44	35		
DE	394	409	419	358	371	400	417	329		
DK	12	12	12	12	10	6	7	4		
ES	352	321	372	261	260	236	241	171		
FI	26	29	25	20	31	29	27	20		
FR	456	463	456	399	305	299	289	211		
GB	92	97	38	31	64	67	47	39		
GR	16	20	23	15	6	7	13	4		
HU	23	23	21	21	19	23	23	18		
IE	6	8	7	4	8	8	11	5		
IT	309	379	509	259	159	248	296	127		
LT	8	12	12	6	12	12	13	10		
LU	5	5	6	4	1	1	3	0		
NL	42	50	51	37	36	38	36	33		
PT	56	55	56	48	45	42	45	37		
SE	11	14	16	9	14	20	25	10		
Total	2010	2099	2236	1670	1487	1584	1664	1151		

Table S1. Number of monitoring sites by year

Countries: Austria (AT), Belgium (BE), Denmark (DK), Finland (FI), France (FR), Germany (DE), Greece (GR), Hungary (HU), Ireland (IE), Italy (IT) Lithuania (LT), Luxembourg (LU), the Netherlands (NL), Portugal (PT), Spain (ES), Sweden (SE), United Kingdom (GB)

Table S2. Evaluation statistics by regions for best models (based on concentration ($\mu g/m^3$))

		At Ev	valuatior	n Sites ^b		At All Sites				
Regions ^a	V Sa	Vith tellite	Withou	ıt Satellite	N	With Satellite		Withou	Ν	
5	\mathbf{R}^2	RMSE	R ²	RMSE		\mathbf{R}^2	RMSE	R ²	RMSE	
Overall	0.56	11.54	0.54	11.82	398	0.51	11.80	0.49	12.04	2010
DK-FI-SE-LT	0.73	8.63	0.75	8.47	11	0.62	8.28	0.58	9.51	57
BE-LU-NL	0.66	7.50	0.67	7.82	25	0.53	10.08	0.54	9.16	103
GB-IE	0.44	16.41	0.40	16.86	19	0.64	12.05	0.61	12.15	98
DE	0.60	8.55	0.61	8.68	69	0.58	9.79	0.65	8.99	394
FR	0.58	9.58	0.59	9.88	90	0.50	10.15	0.48	11.07	456
HU-AT	0.61	9.42	0.60	9.04	26	0.43	11.70	0.46	11.13	169
PT-ES	0.63	9.71	0.61	9.95	93	0.67	10.25	0.64	10.65	408
IT-GR	0.52	17.67	0.50	18.22	65	0.43	17.57	0.41	18.25	325

A. Final NO₂ model, year 2005 (Corine + satellite vs. Corine only)

B. Final PM₁₀ model, year 2007 (Global + satellite vs. Global only)

		At Ev	valuatior	n Sites ^b		At All Sites					
Regions ^a	V Sa	Vith tellite	Withou	ıt Satellite	N	With Satellite		Withou	N		
	\mathbf{R}^2	RMSE	\mathbf{R}^2	RMSE		\mathbf{R}^2	RMSE	\mathbf{R}^2	RMSE		
Overall	0.47	6.74	0.37	7.40	325	0.49	6.26	0.35	7.07	1664	
DK-FI-SE-LT	0.30	7.29	0.31	5.73	15	0.38	6.64	0.44	5.49	72	
BE-LU-NL	0.38	3.38	0.48	3.64	16	0.32	4.28	0.34	4.32	83	
GB-IE	0.00	5.20	0.03	5.97	10	0.57	4.40	0.53	5.25	58	
DE	0.58	4.03	0.54	4.10	76	0.50	4.12	0.48	4.18	417	
FR	0.32	5.54	0.26	5.82	55	0.23	5.27	0.16	5.66	289	
HU-AT	0.06	5.16	0.10	5.04	27	0.26	4.94	0.35	4.97	150	
PT-ES	0.39	9.67	0.33	9.93	66	0.32	8.52	0.29	8.43	286	
IT-GR	0.54	7.86	0.19	10.35	60	0.45	8.02	0.07	11.00	309	

a. Regions listed north to south

b. Evaluation sites refers to the 20% sites not used in model building

Countries: Austria (AT), Belgium (BE), Denmark (DK), Finland (FI), France (FR), Germany (DE), Greece (GR), Hungary (HU), Ireland (IE), Italy (IT) Lithuania (LT), Luxembourg (LU), the Netherlands (NL), Portugal (PT), Spain (ES), Sweden (SE), United Kingdom (GB)

				Model Ev	aluation ^b	
Year	Model	Subset ^b	Model Building ^a	LN Concentration	Concentration (µg/m ³)	Decision
			Adj-R ²	\mathbf{R}^2	\mathbf{R}^2	
NO ₂				•	•	
2005	Corine with	1	0.58	0.58	0.56	final model
	satellite	2	0.58	0.58	0.39	
		3	0.59	0.53	0.45	
		4	0.58	0.60	0.54	
		5	0.58	0.60	0.52	
2006	Corine with	1	0.56	0.52	0.41	reject: spatial autocorrelation
	satellite	2	0.55	0.55	0.50	final model
		3	0.56	0.52	0.42	
		4	0.55	0.54	0.47	
		5	0.54	0.60	0.50	
2007	Corine with	1	0.55	0.59	0.50	final model
	satellite	2	0.56	0.56	0.41	
		3	0.57	0.50	0.43	
		4	0.56	0.54	0.43	
		5	0.55	0.58	0.52	
PM ₁₀	1			L	•	
2005	Global with	1	0.35	0.41	0.44	final model
	satellite	2	0.36	0.34	0.36	
		3	0.36	0.37	0.38	
		4	0.36	0.33	0.35	
		5	0.36	0.29	0.30	
2006	Corine with	1	0.35	0.40	0.38	reject: spatial autocorrelation
	satellite	2	0.36	0.32	0.34	final model
		3	0.38	0.30	0.30	
		4	0.34	0.44	0.43	
		5	0.38	0.29	0.32	
2007	Global with	1	0.50	0.50	0.47	final model
	satellite	2	0.50	0.53	0.48	
		3	0.50	0.53	0.52	
		4	0.50	0.53	0.52	
		5	0.53	0.41	0.41	

Table S3. Sensitivity analysis - rotating 20% evaluation subsets

a. Model building based on natural logarithm of concentration (LN concentration) using 80% of monitoring sites b. Model evaluation using 20% reserved monitoring sites

	Model building ^a								
Variables	ßb	IQR	ß* IQR	VIF	Partial Adj-R ²				
2005 - Corine with satellite									
Constant	2.245								
Minor roads 1500m	4.37E-06	56158	0.25	2.6	0.37				
Satellite-derived surface NO ₂ 2005	6.46E-02	3.0	0.19	1.3	0.49				
Major roads 100m	6.02E-04	0.0	0.00	1.1	0.53				
Total built up land 300m	3.31E-03	55.2	0.18	2.2	0.56				
Minor roads 1500-10000m	1.19E-07	981014	0.12	1.9	0.57				
Semi-natural land 600m	-4.03E-03	4.4	-0.02	1.6	0.58				
2006 - Corine with satellite									
Constant	2.35								
Minor roads 2000m	2.49E-06	88596	0.22	2.7	0.34				
Satellite-derived surface NO ₂ 2006	4.30E-02	3.8	0.17	1.2	0.44				
Semi-natural land 500m	-4.85E-03	1.2	-0.01	1.6	0.48				
Major roads 100m	6.55E-04	0.0	0.00	1.1	0.53				
Total built up land 400m	3.44E-03	53.1	0.18	2.2	0.54				
Minor roads 2000-10000m	1.07E-07	939846	0.10	1.9	0.55				
2007 - Corine with satellite									
Constant	2.28								
Minor roads 1500m	4.35E-06	55676	0.24	2.5	0.33				
Satellite-derived surface NO ₂ 2007	6.51E-02	3.02	0.20	1.3	0.46				
Major roads 100m	6.21E-04	0.00	0.00	1.0	0.50				
Semi-natural land 600m	-4.35E-03	4.42	-0.02	1.6	0.53				
Total built up land 300m	3.20E-03	55.17	0.18	2.2	0.55				
Minor roads 1500-10000m	1.02E-07	917706.00	0.09	1.8	0.56				

Table S4. Sensitivity analysis - NO_2 models based on all monitoring sites

a. Model building based on natural logarithm of concentration (LN concentration) using 100% of monitoring sites b. All p-values < 0.000

Adj-R² including country dummy variables: 0.62 (year-2005), 0.61 (year-2006) and 0.62 (year-2007)

	Model bu	uilding ^a			
Variables	ß ^b	IQR	ß* IQR	VIF	Partial Adj-R ²
2005 - Global with satellite					
Constant	3.36				
Tree canopy 500m	-3.45E-03	7.5	-0.03	1.2	.12
Satellite-derived surface PM _{2.5} 2001-6	2.10E-02	7.1	0.15	1.1	.20
Y coordinate	-1.93E-07	775844	-0.15	1.1	.30
Impervious surface 800m	2.71E-03	40.5	0.11	1.2	.34
2006 - Corine with satellite					
Constant	3.47				
Satellite-derived surface PM _{2.5} 2001-6	2.23E-02	6.9	0.15	1.1	.13
Y coordinate	-1.82E-07	778267	-0.14	1.2	.25
Semi-natural land 1000m	-3.04E-03	10.4	-0.03	1.2	.31
High density residential 1500m	3.04E-03	13.1	0.04	1.2	.34
Major roads 100m	2.05E-04	0.0	0.00	1.1	.35
Distance to sea	-1.87E-01	0.4	-0.07	1.2	.36
2007 - Global with satellite					
Constant	3.65				
Y coordinate	-2.85E-07	780423	-0.22	1.2	.14
Satellite-derived surface PM _{2.5} 2001-6	2.00E-02	7.1	0.14	1.1	.30
Impervious surface 1000m	2.32E-03	36.4	0.08	1.4	.42
Altitude	-7.37E-01	0.2	-0.12	1.2	.47
Minor roads 200m	4.59E-05	1564	0.07	1.4	.49
Major roads	4.94E-04	0.0	0.00	1.1	.50

Table S5. Sensitivity analysis - PM_{10} models based on all monitoring sites

 Major roads
 4.94E-04
 0.0
 1.1
 .30

 a. Model building based on natural logarithm of concentration (LN concentration) using 100% of monitoring sites

 b. All p-values < 0.000</td>

Adj-R² including country dummy variables: 0.54 (year-2005), 0.53 (year-2006) and 0.54 (year-2007)

Table S6. Final NO_2 models by year

	Model building ^a								
Variables	ß ^b	IQR	ß* IQR	ga VIF 2.6 1.3 1.6 1.1 2.1 2.0 2.2.5 1.3 1.1 2.0 2.2.5 1.3 1.1 1.6 2.2.5 1.3 2.1 2.0 2.2.5 1.3 2.2.5 1.3 2.2.6 1.3	Partial Adj-R ²				
2005 - Corine with satellite									
Constant	2.31								
Minor roads 1800m	3.22E-06	74727	0.24	2.6	0.38				
Satellite-derived surface NO ₂ 2005	6.13E-02	3.0	0.18	1.3	0.48				
Semi-natural land 600m	-4.84E-03	4.4	-0.02	1.6	0.52				
Major roads 100m	5.91E-04	0.0	0.00	1.1	0.56				
Total built up land 300m	3.15E-03	55.2	0.17	2.1	0.57				
Minor roads 1800-10000m	1.04E-07	978059	0.10	2.0	0.58				
2006 - Corine with satellite									
Constant	2.35								
Minor roads 1500m	3.96E-06	54683	0.22	2.5	0.33				
Satellite-derived surface NO ₂ 2006	4.30E-02	4.0	0.17	1.3	0.43				
Major roads 100m	6.49E-04	0.0	0.00	1.1	0.48				
Semi-natural land 500m	-5.19E-03	1.2	-0.01	1.6	0.52				
Minor roads 1500-10000m	1.22E-07	965644	0.12	1.7	0.54				
Total built up land 300m	3.10E-03	51.7	0.16	2.2	0.55				
2007 - Corine with satellite									
Constant	2.3								
Minor roads 1500m	4.21E-06	55549	0.23	2.6	0.32				
Satellite-derived surface NO ₂ 2007	6.37E-02	3.1	0.20	1.3	0.45				
Major roads 100m	6.33E-04	0.0	0.00	1.0	0.49				
Semi-natural land 600m	-4.30E-03	5.3	-0.02	1.6	0.53				
Total built up land 300m	3.13E-03	58.6	0.18	2.2	0.54				
Minor roads 1500-10000m	1.01E-07	912789	0.09	1.8	0.55				
2005-2007 - Corine with satellite									
Constant	2.3								
Minor roads 1500m	4.09E-06	54754	0.22	2.5	0.37				
Satellite-derived surface NO ₂ 2005-2007 ^c	5.29E-02	3.2	0.17	1.3	0.48				
Semi-natural land 600m	-4.79E-03	4.4	-0.02	1.6	0.53				
Major roads 100m	5.93E-04	0.0	0.00	1.1	0.57				
Minor roads 1500-10000m	1.22E-07	961813	0.12	1.8	0.58				
Total built up land 300m	3.16E-03	48.3	0.15	2.3	0.60				

a. Model building based on natural logarithm of concentration (LN concentration) using 80% of monitoring sites b. All p-values < 0.000

c. Average of annual satellite-derived surface NO_2 for the three year period

	Model building ^a								
Variables	ß ^b	IQR	ß* IQR	VIF	Partial Adj-R ²				
2005 - Global with satellite									
Constant	3.42								
Y coordinate	-1.87E-07	756842	-0.14	1.1	0.13				
Satellite-derived surface PM _{2.5} 2001-6	2.26E-02	6.9	0.16	1.1	0.24				
Impervious surface 1000m	2.46E-03	39.1	0.10	1.2	0.31				
Tree canopy 500m	-2.86E-03	7.7	-0.02	1.3	0.33				
Distance to sea	-2.20E-01	0.4	-0.08	1.1	0.34				
Major roads	4.47E-04	0.0	0.00	1.1	0.35				
2006 - Corine with satellite									
Constant	3.471								
Satellite-derived surface PM _{2.5} 2001-6	2.19E-02	6.9	0.15	1.1	0.13				
Y coordinate	-2.00E-07	780076	-0.16	1.1	0.26				
Total built up land 600m	8.86E-04	52.2	0.05	1.9	0.31				
Population 1800m	1.04E-06	38155	0.04	1.3	0.33				
Distance to sea	-2.42E-01	0.4	-0.09	1.1	0.35				
Major roads 100m	1.97E-04	0.0	0.00	1.1	0.36				
Semi-natural land 1000m	-1.77E-03	1.8	0.00	1.6	0.37				
2007 - Global with satellite									
Constant	3.67								
Satellite-derived surface PM _{2.5} 2001-6	1.93E-02	7.1	0.14	1.1	0.16				
Y coordinate	-2.83E-07	778777	-0.22	1.2	0.31				
Minor roads 200-2500m	6.03E-07	119820	0.07	1.7	0.42				
Altitude	-6.93E-01	0.2	-0.11	1.2	0.47				
Minor roads 200m	3.52E-05	1538.0	0.05	1.6	0.48				
Major roads	5.07E-04	0.0	0.00	1.1	0.49				
Tree canopy 100m	-1.86E-03	8.2	-0.02	1.3	0.50				
2005-2007 - Corine with satellite									
Constant	3.61								
Y coordinate	-2.40E-07	683381	-0.16	1.1	0.17				
Satellite-derived surface PM _{2.5} 2001-6	2.24E-02	7.1	0.16	1.1	0.31				
Semi-natural land 1200m	-2.30E-03	12.9	-0.03	1.7	0.40				
Distance to sea	-3.45E-01	0.3	-0.11	1.1	0.43				
Population 1800m	7.48E-07	37142	0.03	1.3	0.46				
Total built up 400m	1.18E-03	51.0	0.06	1.7	0.47				
Major roads 100m	1.42E-04	0.0	0.00	1.1	0.48				

Table S7. Final PM_{10} models by year

a. Model building based on natural logarithm of concentration (LN concentration) using 80% of monitoring sites

b. All p-values < 0.000

	Model	Model building ^a			Model Evaluation ^b									
Year		Adj-R ²	SEE	Ν	LN concentration		Concentration (µg/m ³)							N
					R ²	SEE	\mathbf{R}^2	RMSE	ME	MAE	MB	MAB	Regression line ^c	IN
NO ₂														
2005	Corine with satellite	0.58	0.42	1612	0.58	0.44	0.56	11.54	-1.8	8.1	13	37	y=0.98x+2.47	398
2006	Corine with satellite	0.55	0.43	1674	0.55	0.42	0.50	11.35	-1.5	8.3	11	35	y=0.90x+4.15	425
2007	Corine with satellite	0.55	0.42	1786	0.59	0.42	0.50	11.80	-1.3	8.5	18	41	y=0.91x+3.76	450
2005-2007	Corine with satellite	0.60	0.39	1330	0.56	0.42	0.46	11.66	-1.8	8.4	8	34	y=0.84x+6.07	340
PM ₁₀														
2005	Global with satellite	0.35	0.27	1184	0.41	0.26	0.44	7.07	-0.8	5.4	4	21	y=1.13x-2.53	303
2006	Corine with satellite	0.37	0.26	1263	0.35	0.27	0.36	7.56	-1.2	6.0	3	22	y=1.04x+0.22	321
2007	Global with satellite	0.50	0.23	1339	0.50	0.23	0.47	6.74	-0.5	4.6	4	17	y=1.07x-1.36	325
2005-2007	Corine with satellite	0.48	0.22	895	0.46	0.21	0.48	5.99	-0.2	4.4	3	17	y=1.00x+0.39	256

Table S8. Summary of model building and evaluation statistics

a. Model building based on natural logarithm of concentration (LN concentration) using 80% of monitoring sites

b. Model evaluation using 20% reserved monitoring sites

c. See Figures S5 and S5 for scatterplots

ME = mean error ($\mu g/m^3$); MAE = mean absolute error ($\mu g/m^3$); MB = mean bias (%); MAB = mean absolute bias (%)



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 PM_{2.5}, at all monitoring sites



Figure S3. Map and profile plots of NO₂ concentration in 2005 without satellite data; scatterplot of modelled vs. measured NO₂ at evaluation sites



Figure S4. Map and profile plots of PM₁₀ concentration in 2007 without satellite data; scatterplot of modelled vs. measured PM₁₀ at evaluation sites



Figure S5. Modelled vs. measured NO₂ concentration (µg/m3) at evaluation sites for final models shown in Tables S3 and S5



Figure S6. Modelled vs. measured PM₁₀ concentration (µg/m3) at evaluation sites for final models shown in Tables S4 and S5