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Development of West-European $PM_{2.5}$ and NO_2 land use regression models incorporating satellite-derived and chemical transport modelling data

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Abbreviations: AB, absolute bias; AE, absolute error; AIRBASE, European air quality database; AOD, aerosol optical depth; COHOV, cross-over-hold-out-validation; COV, cross-over-validation; CTM, Chemical Transport Model; ESCAPE, European Study of Cohorts for Air Pollution Effects; FB, fractional bias; HOV, hold-out-validation; LUR, Land Use Regression; MB, mean bias; ME, mean error; SAT, Satellite-derived

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ABSTRACT

Satellite-derived (SAT) and chemical transport model (CTM) estimates of $PM_{2.5}$ and NO_2 are increasingly used in combination with Land Use Regression (LUR) models. We aimed to compare the contribution of SAT and CTM data to the performance of LUR $PM_{2.5}$ and NO_2 models for Europe.

Four sets of models, all including local traffic and land use variables, were compared (LUR without SAT or CTM, with SAT only, with CTM only, and with both SAT and CTM). LUR models were developed using two monitoring data sets: $PM_{2.5}$ and NO_2 ground level measurements from the European Study of Cohorts for Air Pollution Effects (ESCAPE) and from the European AIRBASE network.

LUR $PM_{2.5}$ models including SAT and SAT+CTM explained ~60% of spatial variation in measured $PM_{2.5}$ concentrations, substantially more than the LUR model without SAT and CTM (adjR^2: 0.33–0.38). For NO_2 CTM improved prediction modestly (adjR^2: 0.58) compared to models without SAT and CTM (adjR^2: 0.47–0.51). Both monitoring networks are capable of producing models explaining the spatial variance over a large study area.

SAT and CTM estimates of $PM_{2.5}$ and NO_2 significantly improved the performance of high spatial resolution LUR models at the European scale for use in large epidemiological studies.

1. Introduction

Recent studies have indicated that long-term air pollution concentrations can have adverse health effects even below current air quality standards (Pedersen et al., 2013; Beelen et al., 2014). To further quantify effects on health outcomes at these lower air pollution levels, it is necessary to undertake large epidemiological studies and/or pool data from multiple cohorts. Air pollution exposure estimates over large geographic areas at sufficient fine spatial resolution are thus needed.

Land use regression (LUR) models have been used extensively in the past decade to assess air pollution exposure at a fine spatial scale for epidemiological studies (Pedersen et al., 2013; Beelen et al., 2014). Most of the applications of LUR models in epidemiology involve relatively small study areas. For application to large areas, such as continents, modelling the background concentration is a challenge for LUR models as LUR cannot explicitly model secondary formation of components and losses due to deposition and other processes.

To improve prediction of background ground level concentrations, satellite-derived data is increasingly used over sub-continental to continental regions (Novotny et al., 2011; Vienneau et al., 2013; van Donkelaar et al., 2010; Kloog et al., 2012; Ma et al., 2014; Bechle et al., 2015; van Donkelaar et al., 2015a). Similarly, dispersion or chemical transport modelling has been used in conjunction with LUR in so-called hybrid models (Wilton et al., 2010; Arain et al., 2007; Akita et al., 2014). Because of the often relatively coarse spatial resolution of the data (~10 × 10 km), both satellite-derived (SAT) and chemical transport model (CTM) estimates represent area-averaged concentrations. Both also take into account the formation of secondary components with chemical transformations. Integrating satellite derived and/or chemical transport model estimates within LUR may help to achieve a better prediction of air pollution exposure in large-area or even continent-wide studies. Few studies have assessed whether the combination of satellite-based and chemical transport model estimates can help better explain the spatial variation of air pollution concentrations (Reid et al., 2015).

This study presents a comparative assessment of models aimed at characterising air pollution concentrations in Europe through harmonised methods at the local scale for future use in population health studies. In a LUR framework, we combine European-wide satellite-derived ground-level concentration estimates, outputs from a chemical transport model and land use predictors with two ground-based monitoring datasets; the European AIRBASE network (Reid et al., 2015) and the ESCAPE (European Study of Cohorts for Air Pollution Effects) measurement sites (EEA AirBase, 2015; Cyrys et al., 2012). We focus on the pollutants NO_2 and

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$PM_{2.5}$.

2. Methods

2.1. Study area

The study area consists of 19 European countries with available measurements in the AIRBASE dataset (see Fig. 1), the majority of which were part of the ESCAPE project (EEA AirBase, 2015; Cyrys et al., 2012). Measurement and geospatial predictor data were processed in GIS after being (re)projected into the European Terrestrial Reference System (ETRS Lambert Azimuthal Equal Area 52 10), also used for the European Environment Agency (EEA) reference grid.

2.2. Air pollution monitoring data

Air pollution monitoring data were used from the ESCAPE study and the AIRBASE database held by the European Environmental Agency (EEA AirBase, 2015).

We used the ESCAPE annual mean concentrations reflecting the period 2009–2010 from 1426 sites in 36 study areas for NO_2 and 436 sites in 20 study areas for $PM_{2.5}$ (Fig. 1). The ESCAPE measurement campaign was designed specifically to investigate relationships between long term exposure to air pollution and health in existing cohort studies using a land use regression approach. As such, the measurements were undertaken in the relevant study areas of the cohorts. Detailed information about the measurement campaign including site selection and measurement methods can be found elsewhere. (Cyrys et al., 2012; Eeftens et al., 2012a) In brief, over the period of one year (either 2009 or 2010), three 2-weekly measurement campaigns were held in all the study areas measuring $PM_{2.5}$ (20 sites per area) and/or NO_2 (40 sites per area) using Harvard Impactors and Ogawa badges respectively. A previous analysis showed that there was less than 1 $\mu g/m^3$ difference in overall AIRBASE NO_2, $PM_{2.5}$ and PM_{10} annual average concentrations between 2009 and 2010. (Cyrys et al., 2012; Eeftens et al., 2012a) We therefore assumed all ESCAPE measurements to be representative for 2010 in line with all the other data we obtained (AIRBASE $PM_{2.5}$ and NO_2, SAT and CTM). The 3 monitoring periods were held: one in the cool (winter), warm (summer) and intermediate season (autumn or spring) to capture seasonality. One reference monitor in each study period was run during the whole year and these measurements were used to calculate an adjusted annual mean of the measured pollutant concentrations at every site location.

Annual mean concentrations for $PM_{2.5}$ (549 sites) and NO_2 (2400 sites) for 2010 were also derived from the AIRBASE v8 dataset (Fig. 1). (EEA AirBase, 2015) An annual average was only calculated when a site captured $\geq 75\%$ of the total hours (NO_2) and days ($PM_{2.5}$).

Table 1 describes summary statistics of the measured concentrations for the ESCAPE and AIRBASE datasets. Site types and their distribution differed between the AIRBASE and ESCAPE datasets. AIRBASE includes “background”, “industrial” and “traffic” sites, all of which are included here. ESCAPE site locations, classified as “regional background”, “urban background” and “traffic”, were selected to represent population exposures within the ESCAPE study areas, and thus traffic sites were overrepresented.

We randomly stratified both sets of monitoring data by region (defined below) and site type and created a derivation (80%) and a validation (20%) set. We performed the stratification six times and selected the stratification where all the strata where significant (P-value > 0.1). For NO_2 the study areas were divided into five regions (North, West, Central, South West, South East). For $PM_{2.5}$ four regions were chosen to have a sufficient number of sites in each (North, West, Central, South). Table S1 summarises the measured concentrations by region.

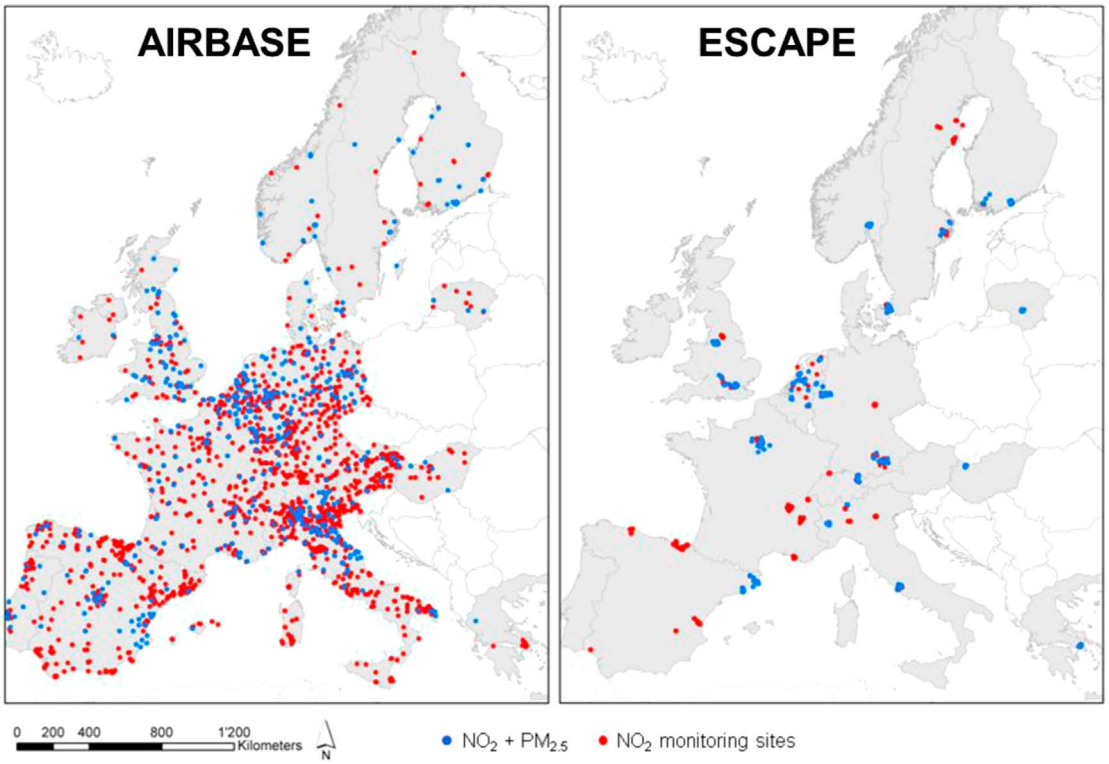

Fig. 1. Location of AIRBASE and ESCAPE monitoring sites for $PM_{2.5}$ and NO_2.

Table 1
Descriptive statistics for monitoring data.

	Site type	N	Mean[a]	Median[a]	SD[a]	P05[a]	P25[a]	P75[a]	P95[a]	Kurtosis	Skewness
$PM_{2.5}$											
ESCAPE	Rural BG[b]	54	13.7	13.9	4.7	5.3	9.4	16.8	21.8	−0.51	−0.05
	Street	207	17.2	17.1	5.9	8.9	12.3	20.5	29.8	0.41	0.68
	Urban BG[b]	175	15.0	14.8	5.3	7.5	11.0	18.5	24.7	0.04	0.50
	Total	436	15.9	15.8	5.7	7.9	11.7	19.2	25.9	0.46	0.61
AIRBASE	BG[b]	345	15.8	15.8	5.2	7.3	12.7	18.9	23.7	0.60	0.22
	Industrial	53	15.1	15.3	5.5	7.7	10.4	18.8	25.6	−0.79	0.41
	Traffic	151	16.3	16.6	4.9	8.5	13.0	19.7	23.7	−0.61	0.01
	Total	549	15.9	15.9	5.1	7.7	12.5	19.3	23.9	0.11	0.18
NO_2											
ESCAPE	Rural BG[b]	119	14.4	14.8	6.3	2.3	9.5	18.0	26.2	−0.10	0.20
	Street	739	38.1	33.4	18.0	16.2	25.3	48.6	72.8	1.15	1.08
	Urban BG[b]	578	23.2	22.3	10.5	7.8	14.8	29.9	42.8	−0.01	0.54
	Total	1436	30.1	26.8	16.9	9.5	18.0	37.7	64.7	2.15	1.28
AIRBASE	BG[b]	1288	21.5	21.0	9.8	5.7	14.8	28.0	37.2	0.19	0.35
	Industrial	372	19.4	18.2	10.1	4.5	11.5	26.7	38.1	0.14	0.61
	Traffic	740	40.2	38.7	14.6	20.3	30.4	47.7	65.8	1.92	0.92
	Total	2400	26.9	25.1	14.6	6.8	16.4	34.7	53.7	1.98	1.02

[a] in $\mu g/m^3$.
[b] BG = Background.

2.3. Satellite derived air pollution estimates

Satellite derived (SAT) estimates of $PM_{2.5}$ for Europe were extracted from the global datasets reported in van Donkelaar et al. (van Donkelaar et al., 2015b).These $PM_{2.5}$ estimates relate aerosol optical depth (AOD) retrievals from the NASA MODIS (Moderate Resolution Imaging Spectroradiometer), MISR (Multi-angle Imaging Spectroradiometer) and SeaWiFS instruments to near-surface concentrations using aerosol vertical profiles and scattering properties simulated by the GEOS-Chem chemical transport model. A dataset for the year 2010 (inferred from 2009 to 2011) was obtained at a $0.1^\circ \times 0.1^\circ$ ($\sim$10 km) resolution.

For NO_2, satellite derived estimates were obtained from the tropospheric NO_2 columns measured with the OMI (Ozone Monitoring Instrument) on board the Aura satellite. Similar to $PM_{2.5}$, the satellite column-integrated retrievals were related to ground-level concentrations using global GEOS-Chem model, producing an annual gridded NO_2 surface for the year 2010 at a 10 km resolution. (Novotny et al., 2011; Bechle et al., 2015; Bechle et al., 2013).

2.4. Chemical transport model estimates

Long range chemical transport model (CTM) estimates for $PM_{2.5}$ and NO_2 were derived from the MACC-II ENSEMBLE model, for the year 2010 at $0.1^\circ \times 0.1^\circ$ ($\sim$10 km) resolution, with estimates available across the whole study area (Inness et al., 2013). At each pixel, the ENSEMBLE model value was defined as the median value of the following seven individual regional CTMs at that particular point: CHIMERE, EMEP, EURAD, LOTOS-EUROS, MATCH, MOCAGE and SILAM. The obtained NETCDF files were imported in ArcGIS for further processing.

2.5. Other predictor variables (roads, land cover, altitude, north-south/east-west trend)

A spatial moving window summation function (focalsum in ArcGIS10) was used to calculate the local predictor variables (e.g. length of road and areas of different land covers) for selected distances around the sites (Table S2). More details about these data can be found in Vienneau et al. (Vienneau et al., 2013). Briefly, road data originated from the 1:10,000 EuroStreets digital road network (version 3.1, based on TeleAtlas MultiNet TM for year-2008). The road data was classified into 'all' and 'major' roads using the classification available in EuroStreets. These were then intersected with a 100 m base polygon and the sum of road length within each 100×100 m cell calculated and converted into a 100 m grid. For land cover, the 100 m European Corine Land Cover 2006 data set was obtained (ETC-LC Land Cover (CLC2006), 2014). This dataset covered the whole study area except Greece. For Greece, the dataset for year 2000 was used (ETC-LC Land Cover (CLC2000), 2013). From the available 44 land classes, six main groups were derived: residential (proxy for population density and domestic sources of air pollution), industry, ports, urban green space, total built up and natural land. For elevation we use the SRTM Digital Elevation Database version 4.1 with a resolution of one arc second (approximately 90 m) and a vertical error < 16 m (SRTM CGIAR-CSI 90m Digital Elevation Data, 2013). SRTM is available for most of the study area, up to 60°N latitude. For northern Scandinavia we used the 1 km resolution Topo30 data. The predictor variable grid reflecting north-south trend was constructed by taking the Y-coordinate (centroids) of the 100×100 m basegrid.

2.6. Statistical analysis

Geospatial analysis was performed in ESRI ArcGIS 10 and statistical analysis in SPSS version 23.

Models were developed for $PM_{2.5}$ and NO_2 based on the ESCAPE and AIRBASE measurement datasets separately. Because of the differences between the ESCAPE and AIRBASE monitoring data (see Section 4), we refrained from developing models combining the two datasets. Following the ESCAPE protocol (Eeftens et al., 2012b), supervised stepwise linear regression was used to predict concentrations at measurements sites on the basis of the available predictor variables. For each pollutant the following four LUR models at ESCAPE and AIRBASE sites were developed:

1. no SAT or CTM estimates (M1);
2. forcing in SAT estimates (M2);
3. forcing in CTM estimates (M3);
4. forcing in both SAT and CTM estimates (M4).

Models were developed using 80% of the sites and validated on the remaining 20%, the hold-out-validation (HOV) set. Predictor variables were only maintained in the model if they followed the expected direction of effect (e.g. positive for SAT, CTM, road length

and negative for green space and natural areas). We did not follow the rule previously used in the ESCAPE protocol, i.e. that variables had to add more than 1% to the adjusted R^2. Instead the addition of variables was repeated until no variable added to the adjusted R^2 of the previous model. This allowed significant predictors to enter the model which would have otherwise been ruled out. Compared to earlier ESCAPE analyses within study areas, we now analysed more sites and exploited much larger variation in measured concentrations. The latitude and longitude variables were only offered to the models if other variables were exhausted. Models were checked for co-linearity (VIF) and influential observations (Cook's D). Every model was also validated using the monitoring data not used in model building; i.e. ESCAPE models were validated at the complete set of AIRBASE monitoring sites and vice versa (cross-over-validation, COV). We additionally performed cross-over validation at just the HOV dataset of the other dataset (cross-over-hold-out-validation, COHOV) to compare the performance of the ESCAPE and AIRBASE models.

For the validations (HOV, COV and COHOV) we calculated the following performance statistics: R^2, RMSE, the constant and slope of the regression line, and additionally for HOV: mean error (ME), absolute error (AE), mean bias (MB), absolute bias (AB) and fractional bias (FB). For insight into suitability of models to estimate exposures for cohorts in smaller regions, we also evaluated model performance at the regional level using R^2, RMSE, constant and slope. The performance of models was also evaluated by site type (R^2, RMSE) to investigate possible bias by monitoring location characteristics.

To compare the separate contribution of predictor variables in the models, the regression slope (ß) of each variable was multiplied by the difference between the 95th and 5th percentile of each predictor value (predicted P95-P5 in $\mu g/m^3$).

For the purpose of this assessment we defined squared correlations (R^2) as weak (0 – 0.2), moderate (> 0.2–0.4), moderately strong (> 0.4–0.6) and strong (> 0.6).

3. Results

3.1. Correlation measured concentration, SAT and CTM

$PM_{2.5}$ SAT estimates have a moderately strong association with measured $PM_{2.5}$ concentrations at ESCAPE and AIRBASE sites (Fig. S1, $R^2 = 0.44$ and 0.49 respectively). $PM_{2.5}$ CTM estimates show a similar correlation at AIRBASE sites ($R^2 = 0.40$) but less so at ESCAPE sites (0.22). $PM_{2.5}$ SAT and $PM_{2.5}$ CTM are weakly correlated at ESCAPE sites and moderately strong at AIRBASE sites. The weak association seems partially caused by a number of sites in Paris were CTM estimates are nearly 2–3 times higher than SAT estimates and higher than the ground measurements.

NO_2 CTM estimates and especially NO_2 SAT estimates are less successful in explaining the variation in measured NO_2 concentrations. SAT and CTM estimates are moderately strongly correlated, although CTM estimates are more than 3 times higher than SAT at both AIRBASE and ESCAPE sites.

3.2. $PM_{2.5}$ models

The M1 models without satellite and chemical transport model estimates performed moderately, explaining 38% (33%) of the variation in measured $PM_{2.5}$ concentrations during the model derivation at the ESCAPE (AIRBASE) sites (Table 2). The models are dominated by non-traffic variables (e.g. urban green, natural, y-coordinate). The M2 models, including the SAT estimate, performed better, explaining 58% (ESCAPE) and 61% (AIRBASE) of the observed variation. In both ESCAPE and AIRBASE M2 models, the SAT variable has the largest contribution (partial R^2: 0.45 and 0.48, P95-P5: 10.99 and 10.18 $\mu g/m^3$) (Table S3). Despite improving the partial R^2 only slightly, local traffic indicators with small buffer sizes enter the models, explaining some of the residual local variation in $PM_{2.5}$ concentrations. Including the CTM estimate (M3) also produced better models, especially at AIRBASE sites where we observe a higher derivation R^2 (adj $R^2 = 0.52$) than model M1. The ESCAPE M3 model performs less well (derivation adj $R^2 = 0.40$). The CTM variable had the largest influence in the AIRBASE M3 model (P95-P5 = 10.49 $\mu g/m^3$). This was not the case in the ESCAPE model, where the y-coordinate took the largest share (P95-P5 = −7.38 $\mu g/m^3$). The best predictive models for $PM_{2.5}$ at ESCAPE and AIRBASE sites included both SAT and CTM estimates (M4). The M4 ESCAPE and AIRBASE models explained about 2% more variation than the models with SAT only. Local predictor variables retained in the ESCAPE model included two local road variables (major roads, all roads 100 m), residential area, altitude and y-coordinate. The AIRBASE model was very similar also including a local road variable (major roads 100 m).

Overall, model HOV R^2 was 5–8% lower than the derivation R^2 suggesting robust models. All validation statistics (HOV, COV and COHOV) showed the same pattern as the model R^2: ESCAPE and AIRBASE M2 and M4 models were consistently better than the respective M1 and M3 models (Table 2 and S4). Importantly, the M4 models explained 52–59% of the variation in the other dataset (COV validation), which is almost equal to the respective HOV R^2, further supporting the robustness of the models.

The AIRBASE M4 $PM_{2.5}$ model was applied to the whole study area at a 100 m resolution (Fig. 2). The map shows generally lower estimated $PM_{2.5}$ concentrations in the north than in the south. Large conurbations like Paris (France) and London (UK) can be detected, as can large industrial regions like the Po valley (Italy) and the Ruhr area (Germany). On closer inspection (see inset) the influence of the larger scale predictor variables from the satellite model becomes apparent. Also apparent are the cells with 'no data' as a result of the grid cells without a satellite estimate. The transect graph illustrates that the $PM_{2.5}$-SAT and $PM_{2.5}$-CTM variables contribute most to the predicted $PM_{2.5}$ concentration, with variables like major roads and residential area adding to the local variation (Fig. 2). The scatterplots show the moderately strong correlation for both HOV and COV validations. Regression lines are close to the 1:1 line (Table S4).

3.3. NO_2 models

The M1 models without satellite and chemical transport model estimates performed moderately strong, explaining 47% and 52% of the variation in measured NO_2 concentrations at the ESCAPE and AIRBASE sites respectively (Table 2). The ESCAPE and AIRBASE models with CTM explained 7–10% more variation than the models without SAT or CTM, more than the models with SAT (2–4% additional explained variance). All 3 ESCAPE models (M1, M2 and M3) included similar predictor variables like length of major road and all roads and residential area within different buffers and y-coordinate, with M1 and M2 also including ports and industrial/commercial area. CTM was by far the strongest variable in ESCAPE M3 (P95-P15 = 24.83 $\mu g/m^3$) (Table S3). In the AIRBASE M3 model CTM and length of all roads within 2 km were strongly influential (P95-P5 = 17.25 and 11.95 $\mu g/m^3$ respectively). Forcing SAT and CTM together in M4 caused the coefficient of the satellite predictor to become negative, invalidating both the ESCAPE and AIRBASE models.

In cross-over validation, the ESCAPE model M3 explained 55% (COV) of the measured variation at the AIRBASE sites, whereas the AIRBASE M3 model explained 50% at the ESCAPE sites (Table 2). As observed for $PM_{2.5}$, the COV R^2 was similar to the HOV R^2

Table 2
Comparison of basic performance statistics (R^2, SEE) for all models including derivation, HOV, COV and COHOV.

			DERIVATION[c]			HOV[d]		COV[e]		COHOV[f]	
	Model[a]	Predictor variables[b]	R^2	Adj R^2	SEE[g]	R^2	SEE[g]	R^2	SEE[g]	R^2	SEE[g]
$PM_{2.5}$											
ESCAPE	M1	All roads (0.7 km), Urban green (1.8 km), Natural (10 km), Residential, Major roads (0.1 km), Y coordinate	0.394	0.383	4.43	0.318	4.77	0.205	4.57	0.220	4.78
	M2	PM2.5 SAT, All roads (5 km), Residential, Altitude, Major roads, Y coordinate	0.591	0.584	3.65	0.510	4.03	0.575	3.35	0.553	3.61
	M3	PM2.5 CTM, All roads (0.1 km), Natural (0.8 km), Residential, Major roads, Y coordinate	0.410	0.400	4.37	0.369	4.59	0.282	4.34	0.293	4.55
	M4	PM2.5 SAT, PM2.5 CTM, All roads (0.7 km), Residential, Major roads, Altitude, Y coordinate	0.606	0.598	3.59	0.544	3.89	0.592	3.30	0.485	4.01
AIRBASE	M1	Natural (10 km), Natural (0.4 km), Urban green (10 km), Altitude, Major roads (0.1 km), Residential, Urban green (0.6 km), Ind/comm (10 km), Y coordinate	0.339	0.325	4.15	0.268	4.63	0.388	4.43	0.412	4.43
	M2	PM2.5 SAT, Altitude, Natural (0.2 km), All roads (0.1 km), Residential (0.2 km), Major roads, Y coordinate	0.613	0.606	3.18	0.562	3.58	0.558	3.77	0.510	4.03
	M3	PM2.5 CTM, Altitude, Residential (0.2 km), Major roads (0.1 km), Natural (0.1 km), Urban green (1.8 km)	0.527	0.520	3.50	0.445	4.03	0.230	4.97	0.295	4.85
	M4	PM2.5 SAT, PM2.5 CTM, Altitude, Residential (0.2 km), Major roads (0.1 km), Natural (0.1 km), Y coordinate	0.636	0.630	3.08	0.583	3.49	0.523	3.91	0.534	3.93
NO_2											
ESCAPE	M1	All roads (5 km), All roads (0.2 km), Residential (1.8 km), Major roads, Ind/comm(10 km), Ports (0.4 km), Y coordinate	0.474	0.471	12.10	0.377	14.22	0.463	10.69	0.490	10.63
	M2	NO2 SAT, All roads (5 km), All roads (0.2 km), Urban green (1.8 km), Residential (1.5 km), Major roads, Ind/comm(10 km), Ports (0.4 km), Y coordinate	0.514	0.510	11.64	0.400	13.95	0.505	10.26	0.516	10.36
	M3	NO2 CTM, Major roads, Residential (1.5 km), All roads (0.2 km), All roads (2 km), Urban green, Y coordinate	0.573	0.571	10.90	0.442	13.45	0.553	9.75	0.563	9.84
	M4	n.a.									
AIRBASE	M1	All roads (2 km), Major roads (0.1 km), Total build up (10 km), Natural (1.5 km), Residential (0.5 km), Ports (0.2 km), Altitude, All roads, Y coordinate	0.516	0.513	10.13	0.536	10.14	0.366	13.46	0.295	15.13
	M2	NO2 SAT, Major roads (0.1 km), All roads (10 km), Residential (1.8 km), Ports (0.2 km), Residential (0.3 km), All roads (10 km), Y coordinate	0.537	0.535	9.90	0.549	10.00	0.416	12.92	0.350	14.53
	M3	NO2 CTM, Major roads (0.1 km), All roads (2 km), All roads, Ports (0.2 km), Residential (0.3 km), Natural (0.5 km)	0.582	0.581	9.40	0.599	9.43	0.502	11.93	0.425	13.66
	M4	n.a.									

[a] M1 = no SAT or CTM; M2 = including SAT; M3 = including CTM; M4 = including both SAT and CTM.
[b] Predictor variables without a distance attached are the origin cell without a focalsum (e.g. for CORINE variables this is the value within the 100 × 100 m cell).
[c] Derivation: model development on 80% of monitoring sites.
[d] HOV: hold-out-validation on 20% of monitoring sites.
[e] COV: cross-over-validation on full dataset (COV for ESCAPE model on AIRBASE full monitoring sites and vice versa).
[f] COHOV: cross-over-hold-out-validation on 20% of the dataset (COHOV for ESCAPE model on the HOV dataset from AIRBASE monitoring sites and vice versa).
[g] Standard Error Estimate ($\mu g/m^3$).

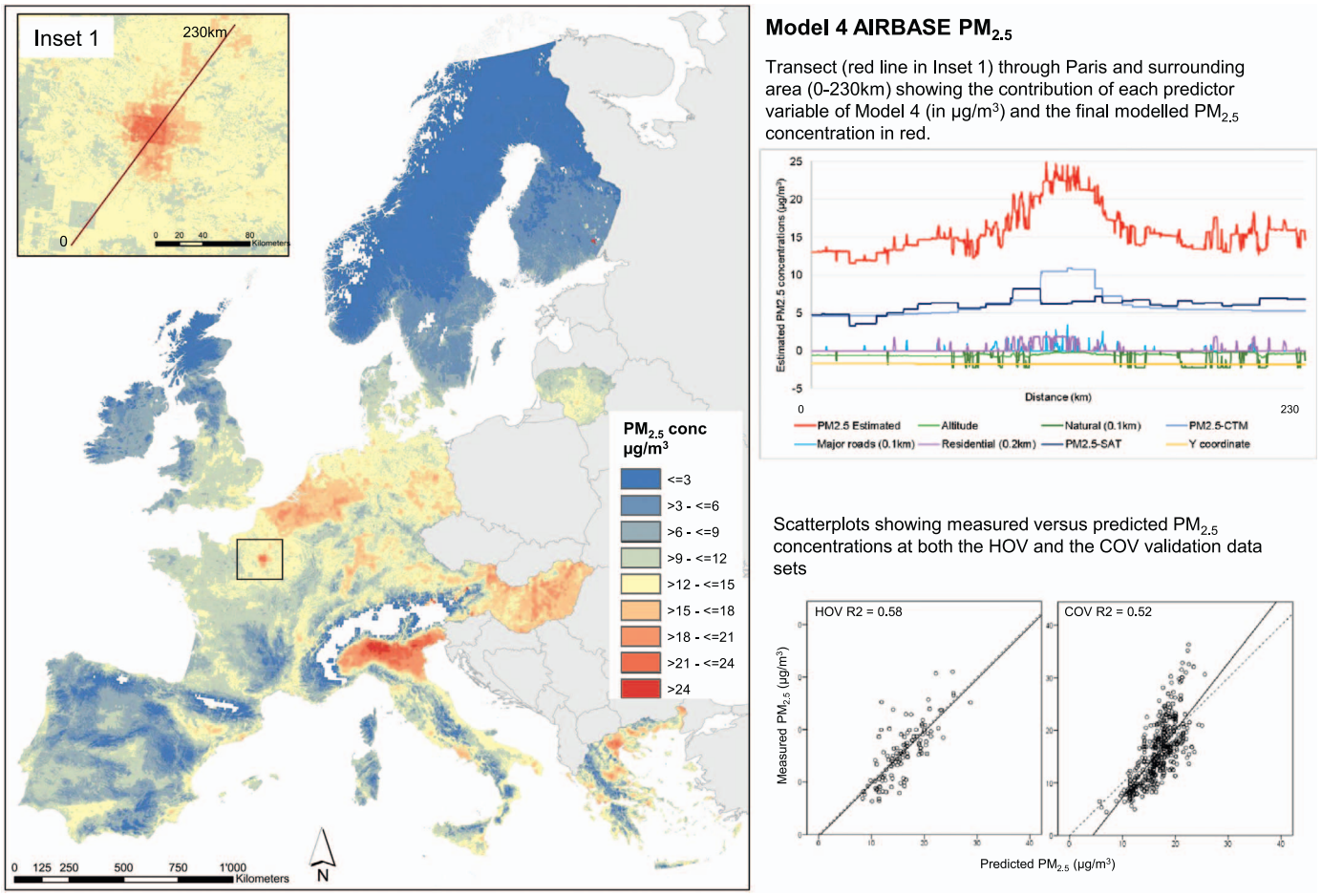

Fig. 2. Map and profile plot of $PM_{2.5}$ concentration predicted by Model 4 (with SAT and CTM) using AIRBASE sites in 2010; scatterplots of modelled vs. measured $PM_{2.5}$ at evaluation sites.

determined within each respective dataset.

Fig. S2 shows the NO_2 M3 AIRBASE model (highest derivation R^2 and HOV R^2) applied to the study area at 100 m resolution. Individual cities show up with higher estimated NO_2 concentrations. Additionally, the influence of the road predictor variables clearly shows up in the inset. The transect shows how the NO_2 CTM variable builds the regional variation and the roads and residential predictors add the local (i.e. spikey) variation. Scatterplots of the HOV and COV validation show moderately strong correlations and regression lines almost 1:1.

3.4. Model performance at subcontinental level and by site type

$PM_{2.5}$ M4 and NO_2 M3 (ESCAPE and AIRBASE), performed moderately to strong in all regions for the COV validation (Table S5, R^2 0.30–0.75). Equally, the performance of these models were similar for the different site types for both the HOV and COV validation (Table S6, R^2 0.32–0.72), except for a lower explained variance of both NO_2 models for traffic sites. Here the lack of traffic intensity data and street configuration data may have played a role.

3.5. Sensitivity analysis

As a sensitivity analysis we developed models based on combined AIRBASE and ESCAPE derivation sites (80%), adding an indicator for network as the last predictor. Similar to the main models, this was validated with a 20% HOV. We performed this test for $PM_{2.5}$ M4 (including both SAT and CTM) and NO_2 M3 (including CTM), finding very similar results to the main models (see Table S7). The indicator variable for network was significant in both models, providing empirical evidence against combining the monitoring datasets.

4. Discussion

We assessed the contribution of satellite or chemical transport modelling data to Europe-wide long-term $PM_{2.5}$ and NO_2 land use regression models using two monitoring databases. Satellite and chemical transport model estimates added to LUR models increased the explained spatial variation of measured air pollution concentrations separately ($PM_{2.5}$ and NO_2) and together ($PM_{2.5}$).

$PM_{2.5}$ models including both satellite and chemical transport model estimates performed best at both monitoring datasets: 20–30% additional variance explained compared to models with only land use and traffic variables. The satellite component had the largest contribution to the overall predicted $PM_{2.5}$ concentrations. For NO_2 the best models included the chemical transport model estimates (6–10% additional explained variance). Validation of models within the same monitoring dataset and across monitoring datasets suggested robust models.

4.1. ESCAPE versus AIRBASE

There was little difference between the overall performance of the ESCAPE and AIRBASE models for both pollutants. Also the HOV, COV and COHOV validations showed similar results. This is

remarkable, given the differences between the ESCAPE and AIRBASE monitoring datasets. Fig. 1 shows the striking difference in spatial coverage of the monitoring networks. The ESCAPE monitoring sites are located in 20 study areas for $PM_{2.5}$ and 36 areas for NO_2, with only the Netherlands having a near national coverage. The AIRBASE monitoring sites, on the contrary, are more evenly spread across the countries. Another difference between the two datasets is the number of sites, which for both pollutants is larger for AIRBASE than ESCAPE (2400 versus 1436 [NO_2]; 549 versus 436 [$PM_{2.5}$]). A crucial difference is the location of the monitoring sites in relation to residential exposure. The choice of the location of monitoring sites in the ESCAPE study was purposely designed such that it would best reflect the spatial variation of air pollution exposures among the cohort study populations within each ESCAPE study area, without the explicit aim to cover Europe as a whole. The monitoring locations in the regulatory AIRBASE network are chosen on a different principle, namely to check for breaches of the air quality guidelines at background sites, near busy roads or in industrial zones. Another important difference is the sampling method used in the two networks. In the ESCAPE study, a harmonised study protocol was implemented based on the Harvard impactor (gravimetry) to monitor $PM_{2.5}$ and the Ogawa badge (diffusion) for NO_2. Because the AIRBASE network is a collection of sites from national networks, the measurement techniques and equipment can differ although reference methods do exist. The most common AIRBASE measurement technique for NO_2 was chemiluminescence, with a minority of AIRBASE sites using Griess-Saltzman reaction, chromatography or diffusion from a variety of monitoring equipment. $PM_{2.5}$ measurements in AIRBASE were carried out mainly by beta ray attenuation or gravimetry, with some sites using TEOM and light scattering devices. Because of the above mentioned differences we refrained from developing models combining the two datasets.

Despite these differences, the two different monitoring datasets performed very similarly in terms of explained variation in monitored concentrations predictor variables and regression coefficients. Also the performance of the best models tested on the opposite monitoring sites were adequate (COV=50–59%, HOCOV=43–53%), giving weight to the robustness of the models developed here. This finding is of high relevance for future air pollution and health research in Europe. AIRBASE data are available in the long term, and our results indicate that future epidemiological studies may rely on these routine data for regulated air pollutants to model exposure distributions within and across European cities.

In the majority of models (12 out of 14), y coordinate was included as an influential predictor variable, reflecting the North (lower) to South (higher) trend in the measured concentrations at both ESCAPE and AIRBASE monitoring sites. This is likely caused by a combination of higher source/population density, emission factors and a generally warmer and drier climate in the South and the influence of Sahara dust in the Mediterranean area.

Application of the ESCAPE model to epidemiological studies in study areas not covered by the original study areas, may be associated with additional misclassification of exposure. The good performance of the ESCAPE model to predict concentrations at AIRBASE sites, covering a large number of regions not covered by ESCAPE monitoring, suggest this may not be a major issue.

4.2. SAT versus CTM

$PM_{2.5}$ SAT estimates fitted much better to measured data, both at the ESCAPE and AIRBASE monitoring sites, than the CTM model and consistently improved LUR models more than CTM. Similar comparison results, using satellite-derived $PM_{2.5}$ estimates, were found by van Donkelaar et al. (van Donkelaar et al., 2015b) indicating a R^2 of 0.53 between the $PM_{2.5}$ SAT surfaces and European Monitoring and Evaluation Programme (EMEP) measurements from 2001 to 2010. Our correlation is lower because EMEP sites are predominantly located at regional background.

In contrast, for NO_2, CTM correlated better with the measurements and contributed more to the LUR model performance. The difference between $PM_{2.5}$ and NO_2 might be explained by better performance of CTM for the gaseous component NO_2 than for fine particles. $PM_{2.5}$ is a complex pollutant, consisting of primary and secondary particles, and therefore with probably less well-characterized emission input data. This is supported by the overestimation of $PM_{2.5}$ by CTM in the Paris region, contributing to a weak correlation between CTM and $PM_{2.5}$ measurements in the ESCAPE data. The spatial scale of SAT and CTM was similar at about 10*10 km, so neither method predicted well the small-scale spatial variation related to local sources.

While for $PM_{2.5}$, the inclusion of CTM (M4) improved the LUR model with SAT (M1) slightly, for NO_2, SAT (M2) did not provide any additional prediction to models with CTM (M3). The correlation between SAT and CTM estimates is stronger for NO_2 than for $PM_{2.5}$, especially at the ESCAPE sites.

The NO_2 model estimates underestimated the measured concentrations at the ESCAPE and AIRBASE modelling sites with between 10 (CTM) and 20 (SAT) $\mu g/m^3$ (Fig. S1). Underestimation of the 2010 NO_2 MACC data is also reported by Giordano et al. (Giordano et al., 2015), showing a consistent underestimation of the MACC model compared to surface NO_2 concentrations at 285 rural stations across Europe. They speculated that this could be due to underestimated emissions in the inventories used by the MACC re-analysis, as well as an underestimation of the chemical lifetime of NOx, too high modelled dry deposition and an underestimation of the natural emissions from soils and/or lightning. Another explanation they offer is the possible systematic positive bias of the NO_2 measurements, i.e. for some instruments only 70–83% of the actual measured NO_2 is attributable to real NO_2 (Giordano et al., 2015). By offering both the spatially coarse CTM estimates and variables which simulate emission sources at a more detailed resolution, the ESCAPE and AIRBASE LUR models can add to the overall R^2 of the CTM model, as shown here, by 20–30% respectively. This is also illustrated in the transect (Fig. S2) where the CTM provides an area-averaged surface that is dominated by the regional background, on top of which the local variables add the necessary more local variation.

The underestimation of NO_2 SAT ($\sim$20 $\mu g/m^3$) was also reported by Vienneau et al. (Vienneau et al., 2013) for the years 2005–2007, when comparing with NO_2 measurements at over 2000 AIRBASE sites in Europe. A similar finding was reported by Bechle et al. (Bechle et al., 2013) for the South Coast Air Basin of California (USA) where the difference between NO_2 SAT and measured NO_2 in 2005 was around 10 ppb ($\sim$19 $\mu g/m^3$) and by Kharol et al. (Kharol et al., 2015) who report absolute concentrations from in situ measurements in North America where twice those from satellite-derived estimates. This is not surprising as the coarse scale satellite-derived NO_2 estimates are area-averaged concentrations, while NO_2 monitors represent the concentration at a specific point, often at roadside or close to city centres. Similarly as with the NO_2 CTM data, the difference was made up by predictor variables representing local sources.

SAT and CTM improved $PM_{2.5}$ models based on land use/traffic variables more than the NO_2 models. This is consistent with the larger impact of variation of the (regional) background on concentrations of $PM_{2.5}$ compared NO_2. For the ESCAPE monitoring data, we previously reported that 81% of total variance of $PM_{2.5}$ was between study area variance, whereas for NO_2 40% was between and 60% within study area variance (Cyrys et al., 2012; Eeftens et al., 2012a). The differences are a result of the magnitude

of local sources, atmospheric formation and loss processes and the resulting atmospheric lifetime of components.

4.3. Comparison with other continental models

It has been previously shown that the inclusion of satellite data improved model performance of European-wide LUR models for NO_2 and PM_{10} respectively by 0.05 and 0.11, explaining 46–56% of the variation in annual means for NO_2 and 36–48% of PM_{10} (Vienneau et al., 2013). At the time of this previous study, it was not possible to develop $PM_{2.5}$ models due to the sparseness of $PM_{2.5}$ monitoring data (195 sites operating in Western Europe during 2007). Recently, however, there has been a sharp increase in the number of $PM_{2.5}$ monitoring sites; 549 sites with $>75\%$ annual data capture operating in 2010 (AIRBASE). At the same time, the ESCAPE study undertook a monitoring campaign measuring $PM_{2.5}$ at 436 sites across Europe. Compared to the NO_2 models described in Vienneau et al. (Vienneau et al., 2013), using AIRBASE and similar GIS predictor variables as used here, the performance of our models developed here are similar; no direct comparison between the PM models was possible, however our best $PM_{2.5}$ models outperform the previous PM_{10} models by more than 10% (36–48% v 60–63%). No other high resolution air pollution models are available for Europe as a whole. Authors have developed models in North America with similar performance results. Hystad et al. (Hystad et al., 2012) developed $PM_{2.5}$ and NO_2 models using satellite-based estimates explaining respectively 52% and 38% of measured spatial variation in Canada for the time period 1975–1994. They compensated for the lack of historical $PM_{2.5}$ measurements, which only started in 1984, by developing predictive models based on co-located $PM_{2.5}$ and Total Suspended Particles (started in 1970) measurements from 1984 to 2000 to predict $PM_{2.5}$ concentrations back to 1974. Van Donkelaar et al. (van Donkelaar et al., 2015a) developed a model that combined geographic weighted regression with satellite-based estimates to explain 82% of the measured spatial variation in $PM_{2.5}$ for North America for 2004–2008. A similar study in the USA by Novotny et al. (Novotny et al., 2011) also incorporating satellite-based estimates developed a national NO_2 model explaining 78% of the concentrations measured in 2006. Models for North America may perform better than ours because measured concentrations are generally lower and show less variability and are therefore less sensitive to missing predictor variables representing local sources.

Wang et al. (Wang et al., 2014) also used the ESCAPE monitoring dataset to build $PM_{2.5}$ and NO_2 models at European scale. It is important to note the difference in modelling approaches between Wang et al. (Wang et al., 2014) and this study. Wang et al. (Wang et al., 2014) used all available ESCAPE predictor variables including local sourced variables like traffic intensity. Despite traffic intensity being available also in areas outside the ESCAPE study areas, within the scope of this study it was not possible to source these separately, and for this reason we could not use these local predictor variables. Despite this, the NO_2 models in both studies are comparable in terms of explained variation (56% in Wang, 58% here). For $PM_{2.5}$, Wang et al. (Wang et al., 2014) obtained a higher explained variance, $R^2=0.80$, compared to $R^2=0.59$–0.63 here. The better $PM_{2.5}$ model performance by Wang et al. (Wang et al., 2014) was driven by the inclusion of measured regional background concentration, something which was not possible at the scale of this study.

4.4. Limitations

No traffic data is available in the whole European study area, so we have used road length for major and all roads as a proxy for traffic sources. The resolution of the SAT and CTM estimates available to us at the time of the study is coarse ($\sim 10 \times 10$ km). Efforts are taking place to refine the resolution to 3 or 1 km grids, which, in future, will help increase the performance of the predictive models. The $PM_{2.5}$ SAT data has cells with missing values, especially in high altitude areas (Alps, Pyrenees, Norway) and over water bodies (Sweden), which is apparent in Figs. 2 and S2. Although these areas typically have a low population density, this is something to keep in mind when using the data in subsequent health analysis.

In summary we found that satellite-derived estimates contribute considerably to the explained variance in $PM_{2.5}$ measured at both ESCAPE and AIRBASE sites. For NO_2, the estimates produced by a chemical transport model improve model predictions compared to a models based only on local and satellite-derived data. Either of the large scale predictors (SAT, CTM) can help produce better continental scale LUR models.

The comparison between measurement data from the routine and purpose-designed monitoring network show that both networks are useful for producing models explaining the spatial variance over a large study area. Especially encouraging is that models based on routine monitoring data are sufficient to predict air pollution exposures at the ESCAPE monitoring sites, with the prospect of future model development for regulated and routinely measured air pollutants. Dedicated measurement campaigns will be required to assess very small-scale differences in exposure and health effects of unregulated air quality indicators.

The resulting $PM_{2.5}$ and NO_2 surfaces at 100×100 m resolution will be made available for the large scale epidemiological studies. To our knowledge, this is the first detailed surface of annual mean $PM_{2.5}$ produced for large parts of Europe. Together with the NO_2 surface the data will be made freely available to the research community to facilitate further epidemiological research.

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This study does not involve human subjects.

Appendix A. Supplementary material

Supplementary data associated with this article can be found in the online version at http://dx.doi.org/%2010.1016/j.envres.2016.07.005.

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