

# Satellite NO<sub>2</sub> data improve national land use regression models for ambient NO<sub>2</sub> in a small densely populated country

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## H I G H L I G H T S

- Satellite NO<sub>2</sub> data improved spatial models for large areas, such as United States.
- We tested NO<sub>2</sub> model improvement by satellite NO<sub>2</sub> in a small densely populated area.
- Satellite NO<sub>2</sub> correlated well with surface NO<sub>2</sub> concentrations at background sites.
- Satellite NO<sub>2</sub> improved land use regression models for spatial variation of NO<sub>2</sub>.

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## A B S T R A C T

Land use regression (LUR) modelling has increasingly been applied to model fine scale spatial variation of outdoor air pollutants including nitrogen dioxide (NO<sub>2</sub>). Satellite observations of tropospheric NO<sub>2</sub> improved LUR model in very large study areas, including Canada, United States and Australia. The aim of our study was to assess the value of satellite observations of NO<sub>2</sub> in modelling the spatial variation of annual average NO<sub>2</sub> concentrations in a small densely populated country.

We used surface level annual average NO<sub>2</sub> concentration and geographic information system data from 144 monitoring sites spread over the Netherlands: 26 regional background, 78 urban background and 40 traffic sites for developing land use regression models. For the 144 monitoring sites we obtained the annual average tropospheric NO<sub>2</sub> concentration for 2007 from the Ozone Monitoring Instrument (OMI) satellite sensor. These OMI data reflect a spatial scale of about 10 × 10 km. We calculated the correlation between satellite and surface level NO<sub>2</sub> concentrations for all sites and for background sites only. We next evaluated whether adding satellite observations improved land use regression models.

Annual average satellite observations of tropospheric NO<sub>2</sub> correlated well spatially with annual average urban plus regional background ( $R = 0.74$ ,  $n = 104$  sites) and especially regional background NO<sub>2</sub> concentrations ( $R = 0.88$ ,  $n = 26$ ). The correlation was moderate for all sites, including traffic locations ( $R = 0.51$ ,  $n = 144$ ). A LUR model including satellite NO<sub>2</sub> observations performed better (overall  $R^2 = 0.84$ ) than LUR models including geographical coordinates or indicator variables (overall  $R^2$  65–74%) in modeling concentrations at the 104 background sites across the Netherlands.

Satellite NO<sub>2</sub> observations agreed well with measured surface concentrations at background locations and improved land use regression models, even in a small densely populated country.

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## 1. Introduction

Modelling of the spatial variability of long-term average concentrations of outdoor air pollution remains challenging (Jerrett et al., 2005). In recent years, land use regression modelling has increasingly been applied to model the concentration of pollutants such as nitrogen dioxide (NO<sub>2</sub>), particles smaller than 2.5 or 10 micrometer (PM<sub>2.5</sub> or PM<sub>10</sub>) and black carbon, particularly in the framework of epidemiological studies (Hoek et al., 2008). Land use regression (LUR) models require monitoring data at representative locations and data on predictor variables often in a Geographic Information System (GIS). Linear regression is used to develop models to explain measured concentrations using predictor variables such as traffic and population density obtained by GIS. Most LUR models have been developed for metropolitan areas (Hoek et al., 2008). Recently, studies have developed LUR models for larger study areas, such as entire countries or the European Union (Vienneau et al., 2010; Beelen et al., 2009; Bechle et al., 2013; Hystad et al., 2011; Vienneau et al., 2013; Knibbs et al., 2014). One of the challenges of land use regression models for larger study areas is the modelling of the (regional) background variation, which is not well modelled even with the largest buffers of traffic, population and land use (5–10 km) included in LUR models. LUR models do not explicitly include large-scale modelling of atmospheric transport, chemical formation and removal mechanisms. Studies have characterized large scale variation with simple indicator variables for region of the country, geographic coordinates and interpolation of regional background measurements (Eeftens et al., 2012, 2011).

Satellite observations have been shown to be useful to characterize surface concentrations of NO<sub>2</sub> (Lamsal et al., 2008; Martin, 2008; Boersma et al., 2009), although the exact relationship between surface and tropospheric column NO<sub>2</sub> needs further work (Wallace and Kanaroglou, 2009; Dieudonné et al., 2013; Flynn et al., 2014). A high fraction of the total column NO<sub>2</sub> is generally present in the lower troposphere, related to high emissions of local sources and the limited atmospheric lifetime of NO<sub>2</sub> (Martin, 2008). While there is good evidence that satellite NO<sub>2</sub> correlates temporally with surface NO<sub>2</sub>, there is much less information on how well satellite NO<sub>2</sub> reflects the spatial variation of surface NO<sub>2</sub> (Bechle et al., 2013).

Recent studies within North America, Australia and Europe showed that satellite observations of NO<sub>2</sub> improved national and supra-national land use regression models by providing particularly the background concentration (Vienneau et al., 2013; Novotny et al., 2011; Hystad et al., 2011; Knibbs et al., 2014). The published studies were conducted in very large study areas and it is currently not known whether satellite NO<sub>2</sub> contributes to modelling in smaller densely populated areas such as the Netherlands (surface area 41,000 km<sup>2</sup>).

The first aim of this study was to evaluate the agreement between annual average satellite observations of NO<sub>2</sub> and surface level concentration measurements in a purpose designed network of 144 sites across the Netherlands. The second aim was to evaluate whether satellite data improved land use regression models based upon these measurements and traffic and land use predictor variables. The monitoring network was specifically designed for developing LUR models, compared to exploiting data from routine monitoring networks in all previous studies. Our network was much more spatially dense than previous studies, e.g. 27 sites included in the Californian study with a similar study area of 27,000 km<sup>2</sup> (Bechle et al., 2013) and 68 sites covering all Australia (Knibbs et al., 2014). Our network further included more traffic sites than previous studies.

## 2. Methods

### 2.1. Study design

We used measured surface level NO<sub>2</sub> concentrations and geographic information system data on predictor variables from the TRACHEA study (Traffic Related Air pollution & Children's respiratory HEalth and Allergies) (Eeftens et al., 2011). Measurements were conducted in 2007 at 144 locations spread over the Netherlands (Fig. 1), of which 26 were regional background, 78 urban background and 40 traffic sites. For the 144 monitoring sites we obtained the annual average tropospheric NO<sub>2</sub> concentration for 2007 from the Ozone Monitoring Instrument (OMI) satellite sensor. These OMI data reflect a spatial scale of about 10 × 10 km<sup>2</sup>. We calculated the spatial correlation between annual average satellite and surface level NO<sub>2</sub> concentrations for all sites and because of the spatial scale of the satellite observations for (urban plus regional) background sites only. We next evaluated whether adding satellite observations improved land use regression models compared to characterizing regional background variation with regional indicator variables, geographical coordinates, interpolation of regional background measurements or no regional variable.

### 2.2. Surface level NO<sub>2</sub> concentration and GIS data

The surface level NO<sub>2</sub> concentration data have been described in detail before (Eeftens et al., 2011). Briefly, integrated 1-week average concentrations of nitrogen oxides (NO<sub>2</sub> and NO) were measured simultaneously at all 144 sites for four periods in the four seasons of 2007. The exact sampling dates were 17–24 January 2007, 18–25 April 2007, 13–20 June 2007 and 26 September – 3 October 2007. NO<sub>x</sub> and NO<sub>2</sub> concentrations were measured using Ogawa passive badges. Because of the use of passive samplers, we did not have information about surface NO<sub>2</sub> concentrations at the time of day of satellite overpass. We used the average concentration of the four measurement periods in further calculations.

All measurement locations were successfully geocoded using the ACN (Address Coordinates Netherlands) database from the year 2000. For all coordinates a set of 24 predictor variables was derived: the number of inhabitants and the home addresses within 300 m, 1,000 m and 5,000 m buffers of the site and 6 different land-use variables were calculated in the same buffers: land use for low-density residential purposes, industry, ports, urban green, forests and agriculture. We selected the buffer sizes to reflect known distances of pollutant dispersion, taking into account the resolution of the available maps. Both home address density and population density maps were available for 1999. Land-use maps on a 100 m × 100 m raster were available from Corine (COordination and INformation on the Environment programme, European Commission) for the year 2000. We finally added the more detailed traffic data from the ESCAPE study (Eeftens, 2012).

### 2.3. Satellite data

The Ozone Monitoring Instrument (OMI) is an ultra-violet–visible spectrometer on board of the NASA EOS Aura satellite, which was launched in July 2004 (Levelt et al., 2006). The OMI data record already consists of more than ten years of data. OMI provides daily global coverage, meaning that it measures the complete Earth within a day. The typical overpass time of the Aura satellite is 13:30 local time. The spatial resolution of OMI is 13 × 24 km<sup>2</sup> (along x across track) at nadir. The spectral range covered by OMI is 270–500 nm and is resolved with a spectral resolution of approximately 0.5 nm.

In this work we use results from the OMI DOMINO Collection 3



**Fig. 1.** Overview of all 144 sampling sites of the 2007 sampling network for nitrogen oxides. Light grey squares mark the positions of regional background sites, black circles represent urban background sites, and dark grey triangles indicate street sampling sites.

version 1.0.2 available from <http://www.temis.nl> (Boersma et al., 2007). This product contains the tropospheric  $\text{NO}_2$  column, which is dominated by  $\text{NO}_2$  in the boundary layer. In brief, OMI tropospheric  $\text{NO}_2$  columns are retrieved with a three step method: (1) performing a Differential Optical Absorption Spectroscopy (DOAS) fit at 405–465 nm to obtain a  $\text{NO}_2$  slant column (entire light path), (2) estimating the stratospheric contribution by data assimilation of slant columns in the TM4 chemistry transport model (Dirksen et al., 2011), and (3) calculating tropospheric  $\text{NO}_2$  from the slant column by applying the tropospheric air mass factor (AMF), the ratio of slant column and the desired vertical column.

Slant columns are derived by a fit of OMI reflectance spectra in the 405–465 nm window (Boersma et al., 2007). The DOAS fit with OMI Collection 3 data includes reference spectra for  $\text{NO}_2$ , ozone,  $\text{H}_2\text{O}$  and the Ring effect. For the reference solar irradiance spectrum a fixed spectrum based on all OMI solar calibration measurements of 2005 is used. Stratospheric  $\text{NO}_2$  is obtained by assimilating  $\text{NO}_2$  slant columns with TM4 (Transport Model version 4), which is driven by ECMWF (European Centre for Medium Range Weather Forecasts) meteorology. Tropospheric slant columns are calculated by subtracting the stratospheric slant column from the total slant column and are subsequently converted to vertical columns by applying the tropospheric AMF. The uncertainty in  $\text{NO}_2$  columns for individual retrievals is estimated at  $0.5\text{--}1.5 \cdot 10^{15}$  molecules/cm<sup>2</sup>

from the spectral fitting and an additional relative error of 10%–40% from errors in the calculation of the air mass factor (Boersma et al., 2004). The OMI dataset was successfully validated by Boersma et al. (2008) and Hains et al. (2010).

The individual overpass data were put onto a rectangular latitude–longitude grid with a resolution of  $0.02^\circ$  ( $1/50\text{th }^\circ$ , approximately  $2.2 \times 1.4 \text{ km}^2$ ), for the range  $47.5^\circ \text{ N}$  to  $57.5^\circ \text{ N}$ ,  $0^\circ \text{ E}$  to  $10^\circ \text{ E}$ . The fine grid of  $0.02^\circ$  was selected because the mean field, averaged over periods of a month or a year, shows finer scale features than the original spatial resolution of the instrument, which is due to the combination of stable pollution sources and robust spatial over-sampling. The over-sampling results from the small shifts in the center of the OMI ground pixels between successive days in OMI's 16-day repeat cycle. In the gridding process, near-nadir ground pixels, which have the highest spatial resolution, are given more weight than the larger off-nadir pixels. The weight given to a ground pixel in the gridding process is  $\cos \theta_0 \cos \theta e^{-2c_{\text{eff}}}$ , where  $\theta_0$  is the solar zenith angle,  $\theta$  is the viewing zenith angle and  $c_{\text{eff}}$  the effective cloud fraction. The solar zenith angle is included in this weighting factor because, depending on the latitude, two or even three OMI observations per day are possible over Europe. The weighting factor is largest for cloud free nadir observations with a small solar zenith angle. To reduce the effect of cloud contamination, only data for which at most 50% of the radiance originates

from the cloudy part of a scene. Likewise ground pixels with less cloudiness are given more weight than cloudy pixels and data for which more than 50% of the light was coming from clouds have been rejected. Monthly gridded data products were produced for all months of 2007, from which a mean tropospheric NO<sub>2</sub> field for 2007 was produced. Fig. 2 shows a map of the annual-average OMI NO<sub>2</sub> column data used in the current study.

#### 2.4. Land use regression modelling

Because of the spatial scale of the satellite data, we first evaluated whether satellite data improved the development of a background model, based upon the 104 regional and urban background locations. Developing background models first and next adding local impacts was possible because of the large number of monitoring sites. The two-stage approach has been proposed to better reflect the scales of air pollution sources (Beelen, 2009; Vienneau, 2010). The background model was developed by first forcing in satellite NO<sub>2</sub> and next offering the GIS predictor variables. We compared the performance of models developed with satellite data with four other approaches used to account for regional variation: interpolated regional background based upon the regional background sites ( $n = 26$ ), geographical coordinates (X and Y coordinate as two separate variables), a simple classification of regions of the country following previous studies (Eeftens et al., 2011) and finally no explicit regional variable. The last model was included for comparison with previous LUR studies that compared LUR model performance with and without satellite data. The interpolated regional background was estimated using inverse distance squared weighted interpolation of pollution levels measured at the 26 regional background sampling sites. For the regional background sites, the site itself was not used to estimate the regional component at that site.

The background model was then developed as described previously (Eeftens et al., 2011). All potential GIS predictor variables were first evaluated individually. The variable with the highest adjusted explained variance ( $R^2$ ) was included, if the sign of the parameter estimate had the a priori specified direction, e.g. positive for industry and ports and negative for urban green. An additional predictor variable was included if the adjusted  $R^2$  increased by at

least 1% compared to the previous model and the direction of the effect was as defined a priori. We further checked statistical

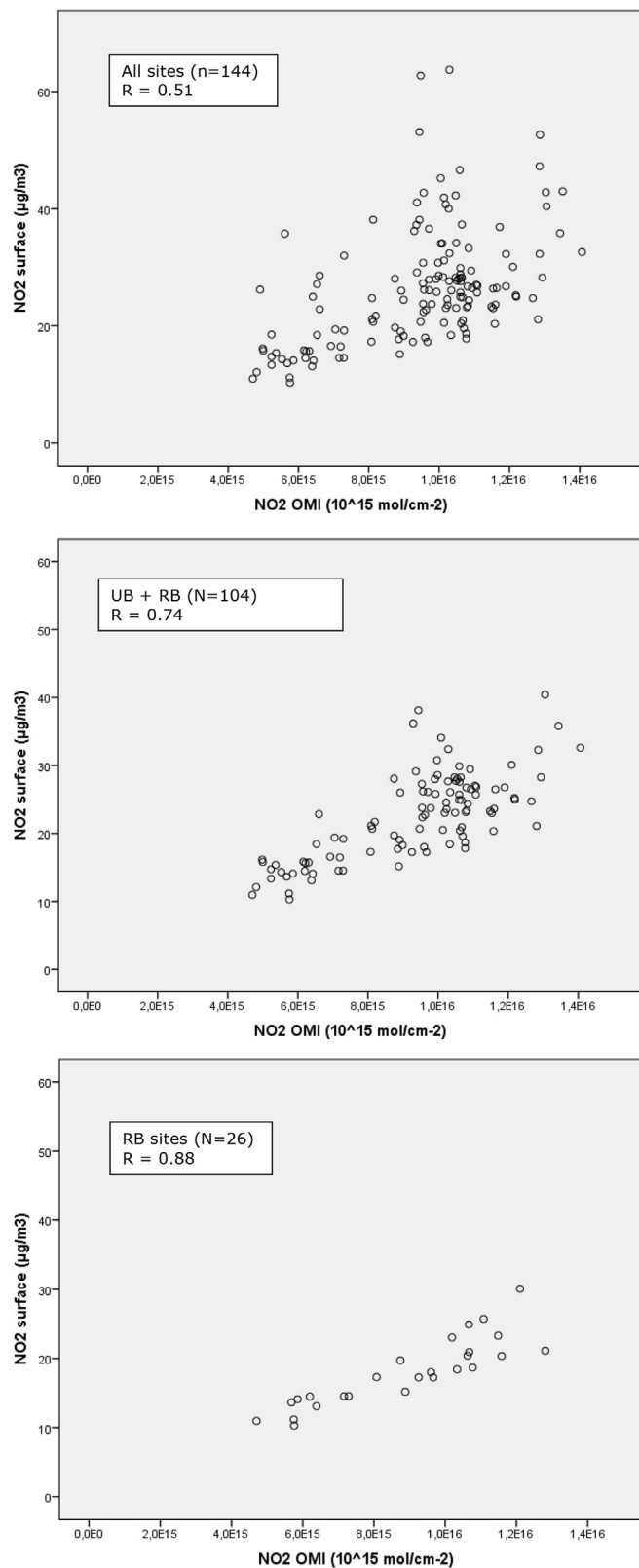


Fig. 3. Agreement between annual average NO<sub>2</sub> by surface monitoring and OMI tropospheric column for all, urban + regional background (UB + RB) and regional background (RB) sites.

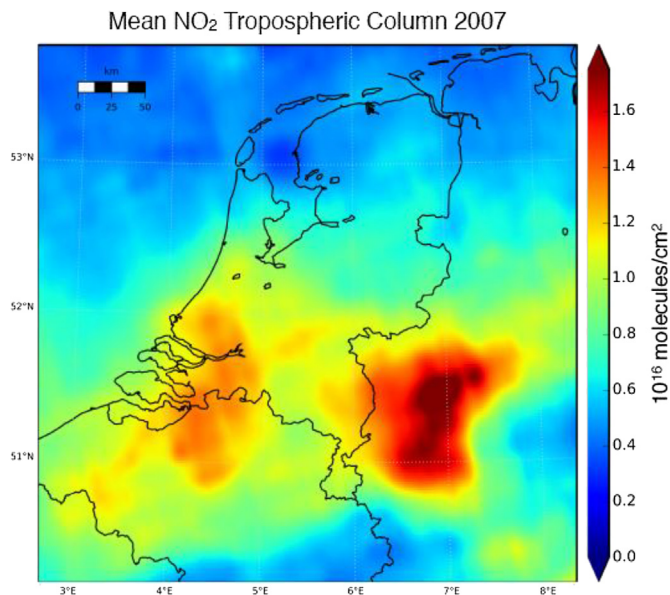


Fig. 2. Annual-average OMI NO<sub>2</sub> used in the current study.

significance ( $p < 0.10$ ), influential observations (Cook's  $D < 1$ ) and collinearity (Variance Inflation factor  $< 3$ ).

Finally, we added small-scale traffic variables to the already identified background model for each regional variable to evaluate the contribution of satellite data to models for all sites including traffic sites. We added the four traffic variables from the NO<sub>2</sub> LUR model developed in the framework of the ESCAPE study for the Netherlands (Eeftens, 2012). The variables were traffic load in a 50 m buffer, inverse distance to the nearest major road, road length of major roads in a 25 m buffer and traffic load of heavy duty vehicles in a 500 m buffer.

### 3. Results

#### 3.1. Agreement between satellite and surface level NO<sub>2</sub>

The agreement of the annual average NO<sub>2</sub> concentration between satellite and surface monitoring is shown in Fig. 3. There was moderate agreement when all sites were included ( $R = 0.51$ ). When the traffic locations were excluded, the correlation improved considerably ( $R = 0.74$ ). Traffic locations are affected by emissions at a fine scale (10-s meters), which is not resolved by the satellite data. The agreement is very good for regional background sites ( $R = 0.88$ ). Because multiple sites were included in most cities and satellite data vary on a  $10 \times 10 \text{ km}^2$  scale and thus are not expected to reflect within-urban variability, we also calculated the correlation between average background ground level concentrations averaged per town and satellite data. The correlation was 0.72, similar to the overall correlation.

#### 3.2. Difference between street, urban and regional background sites

Though OMI NO<sub>2</sub> columns were higher on average for the urban background stations, the difference with regional background stations was not statistically significant (Table 1). Surface concentrations did increase significantly from regional background to urban background to street sites. An analysis restricted to the three largest cities (Amsterdam, Rotterdam, Utrecht) did show a significant difference for satellite NO<sub>2</sub> with regional background. The mean satellite NO<sub>2</sub> column was  $10.7 \times 10^{15} \text{ mol cm}^{-2}$  for the 20 urban background stations within the three largest cities. The difference with all regional sites was  $1.8 \times 10^{15} \text{ mol cm}^{-2}$  ( $p < 0.05$ ). The difference was  $1.3 \times 10^{15} \text{ mol cm}^{-2}$  ( $p < 0.05$ ) when sites located in the relatively clean north of the country were excluded (22 sites remaining). Because none of the major cities is located in the north, this is a fairer comparison. The urban-regional background difference of OMI observations was however smaller than for surface measurements: 17% for satellite versus 35% for surface measurements.

#### 3.3. Land use regression models

NO<sub>2</sub> from satellites and interpolated regional background NO<sub>2</sub> predicted background NO<sub>2</sub> concentrations much better than the

regional indicator variable and X and Y coordinates in univariate analyses (Table 2).

When LUR models were developed with the different regional variables offered, the highest explained variability of the full background model was found when satellite NO<sub>2</sub> was offered (Table 3). The explained variance (adjusted  $R^2$ ) and the root mean square error of the model including satellite data was substantially better than the models with regional indicators, X and Y coordinates and no regional variables. The OMI model was mildly better than the model with interpolated regional NO<sub>2</sub>. The correlation for all 144 sites of OMI and interpolated regional background NO<sub>2</sub> was 0.89. In all models the number of inhabitants in a 5000 m buffer and industry in a 5000 m buffer were present with consistent estimated contributions to surface NO<sub>2</sub>. The largest contributions in the models were from the regional component and the population predictor, the latter reflecting urban-rural differences.

The regression slope for OMI NO<sub>2</sub> was  $1.6 \mu\text{g/m}^3$  per  $10^{15}$  molecules/cm<sup>2</sup> in the model incorporating other predictor variables. The regression slope converts the NO<sub>2</sub> column into surface level concentrations in  $\mu\text{g/m}^3$ .

The model including OMI NO<sub>2</sub> predicted about 10% more variation than a model without regional variables and 4–7% more variation than models with indicators and coordinates when all sites including traffic sites were considered (Table 4). The model with interpolated regional background NO<sub>2</sub> had the same performance as the satellite model. Effect estimates for the four traffic variables were consistent across models.

### 4. Discussion

Annual average OMI observations of tropospheric NO<sub>2</sub> columns correlated well with annual average (urban plus regional) background and especially regional background surface NO<sub>2</sub> concentrations. LUR models with OMI NO<sub>2</sub> observations had slightly higher explained variance than models with interpolated regional background concentrations and much higher explained variance than models with regional indicators or geographic coordinates.

#### 4.1. Satellite column versus surface NO<sub>2</sub>

The good spatial agreement between OMI NO<sub>2</sub> observations and measured surface NO<sub>2</sub> concentrations at a large number of background locations across the Netherlands agrees closely with findings from a recent study from southern California (Bechle et al., 2013). In the California South coast air basin area, the correlation between annual average OMI satellite column and surface NO<sub>2</sub> for 25 sites spread over the area was 0.93 (Bechle et al., 2013). The density of the Californian network was about three times lower

**Table 1**

Mean (standard deviation) of 2007 annual average concentration of measured surface level NO<sub>2</sub> and tropospheric NO<sub>2</sub> by site type.

	NO <sub>2</sub> surface ( $\mu\text{g/m}^3$ ) <sup>a</sup>	NO <sub>2</sub> _OMI ( $10^{15} \text{ mol cm}^{-2}$ )	N
Regional Background	18 (5)	8.9 (2.4)	26
Urban Background	24 (6)	9.5 (2.2)	78
Street	37 (10)	9.7 (2.1)	40

<sup>a</sup> difference between three site types statistically significant ( $p < 0.05$ ) for surface measurements. No significant differences for satellite NO<sub>2</sub> ( $p = 0.35$ ).

**Table 2**

Percentage explained variability (adjusted  $R^2$ ) of annual average surface NO<sub>2</sub> concentration by four regional predictor variables.

	Regional background (N = 26)	Regional + urban background (N = 104)
Indicator variables <sup>a</sup>	25.7	20.5
X and Y coordinate <sup>b</sup>	38.7	39.0
Interpolation <sup>c</sup>	58.9	58.1
Tropospheric NO <sub>2</sub> satellite	75.1	54.5

Adjusted  $R^2$  corrects the model  $R^2$  for differences in degrees of freedom between models.

<sup>a</sup> Three variables indicating whether the sampling site was in the North, West, Middle or South of the country.

<sup>b</sup> As two separate variables.

<sup>c</sup> Inverse distance squared used as weight.

**Table 3**

Land use regression models for background locations (N = 104), using different methods to represent regional variation.

Model	1		2		3		4		5	
	Satellite		Interpolation		X and Y		Region indicators		No indicator	
Predictor	$\beta^a$	se	$\beta$	se	$\beta$	se	$\beta$	se	$\beta$	se
Intercept	3.1	1.1	2.5	1.3	41.2	2.9	13.1	1.0	16.6	0.7
Industry, 5000 m buffer	2.7	0.8	3.3	0.9	2.6	1.0	2.6	1.3	3.5	1.3
Population, 5000 m buffer	6.4	0.6	4.0	0.9	7.3	0.9	7.4	1.3	8.0	1.1
Port, 5000 m <sup>a</sup>			1.2	0.4			1.0	0.6		
Port, 1000 m									1.0	0.5
NO <sub>2</sub> _OMI	9.8	0.7								
NO <sub>2</sub> _interpolation			9.1	0.8						
Xcoord					−4.3	0.9				
Ycoord					−6.4	0.9				
Region middle							6.2	1.1		
Region south							5.3	1.5		
Region west							3.9	1.2		
Adjusted R <sup>2</sup> of model (%)	83.7		81.5		73.8		65.4		55.9	
RMSE ( $\mu\text{g}/\text{m}^3$ )	2.6		2.7		3.2		3.7		4.2	

<sup>a</sup> Regression slope ( $\beta$ ) and standard error (se) in  $\mu\text{g}/\text{m}^3$  were multiplied by the difference between the 10th and 90th percentile of each predictor. These differences were 8.83% (Industry), 245813 (Population), 3.92% (Port),  $6.14 \times 1015$  (NO<sub>2</sub>\_OMI), 11.0  $\mu\text{g}/\text{m}^3$  (interpolated regional background NO<sub>2</sub>), 119.7 km (X-coordinate), 163.7 km (Y-coordinate). Slopes for the region indicators represent the difference with region North.

than our study. Spatial correlations for calculated surface satellite NO<sub>2</sub> using a local or global model for vertical distribution of NO<sub>2</sub> resulted in slightly lower correlations with measured surface NO<sub>2</sub> ( $R = 0.89$  and  $0.91$ ). The higher correlation in the California study could partly be due to their use of surface concentrations averaged for the satellite overpass period to compare with satellite NO<sub>2</sub>, which we could not calculate because of the use of passive samplers. For our application of long-term population exposure assessment, the comparison with the average concentration is however relevant. The contrast in the California study was substantially higher (~factor 20 between lowest and highest satellite NO<sub>2</sub>) than in our study (factor 3). The larger contrast is likely due to much larger differences in land use, population density and altitude in California compared to do the densely populated and largely flat Netherlands. The on average longer lifetime of NO<sub>2</sub> in the Netherlands compared to the photochemically more active California region, especially at 13:30 h when OMI measures, may

further have contributed to a larger contrast. Finally, higher wind speed over the Netherlands may result in a more dispersed signal. A study at about 2000 West-European sites from routine monitoring networks reported correlations of 0.33–0.37 for annual averages from OMI and surface measurements (Vienneau et al., 2013). The sites included background and traffic sites. The lower correlation compared to our overall correlation of 0.51 could be due to differences in monitoring between countries in the Vienneau paper and differences in interferences by other nitrogen components between (e.g. south and north European) countries. Molybdenum-based chemiluminescence monitors are likely affected by other nitrogen components than NO<sub>2</sub> (Lamsal et al., 2008; Boersma et al., 2009; Bechle et al., 2013).

Despite the good spatial correlation, satellite column NO<sub>2</sub> measured only half of the urban-regional background contrast for the three major cities observed by the surface network. This can be understood from (a) the mixing and transport of NO<sub>2</sub> through the

**Table 4**

Land use regression models for background and traffic locations (N = 144), using different methods to represent regional variation.

Model	1		2		3		4		5	
	Satellite		Interpolation		X and Y		Region indicators		No indicator	
Predictor	$\beta^a$	se	$\beta^a$	se	$\beta^a$	se	$\beta^a$	se	$\beta^a$	se
Intercept	3.0	1.5	1.9	1.7	37.6	3.6	12.7	1.1	15.6	0.8
Industry, 5000 m buffer	5.2	1.0	5.4	1.0	5.3	1.1	5.2	1.4	6.0	1.3
Population, 5000 m buffer	4.0	0.8	1.8	1.1	5.0	1.0	5.1	1.3	5.5	1.0
Port, 5000 m			0.8	0.5			0.7	0.6		
Port, 1000 m									1.0	0.6
NO <sub>2</sub> _OMI	9.3	1.0								
NO <sub>2</sub> _interpolation			9.0	1.0						
Xcoord					−4.4	1.1				
Ycoord					−5.4	1.1				
Region middle							5.6	1.2		
Region south							3.3	1.3		
Region west							3.6	1.6		
Traffic load 50 m buffer	4.9	1.1	4.8	1.1	4.4	1.2	4.8	1.3	4.5	1.4
Inverse distance to major road	3.5	0.6	3.7	0.7	3.7	0.7	3.3	0.8	3.4	0.8
Major road length 25 m buffer	2.0	1.0	2.4	1.0	1.8	1.1	1.7	1.2	1.9	1.2
Traffic load HDV 500 m buffer	2.1	0.9	2.3	0.9	1.7	1.0	2.7	1.1	2.8	1.1
Adjusted R <sup>2</sup> of model (%)	83.6		83.6		79.7		76.9		74.1	
RMSE ( $\mu\text{g}/\text{m}^3$ )	4.0		4.0		4.4		4.7		5.0	

HDV = heavy duty vehicles; RMSE = root mean squared error.

<sup>a</sup> Regression slope ( $\beta$ ) and standard error (se) in  $\mu\text{g}/\text{m}^3$  were multiplied by the difference between the 10th and 90th percentile of each predictor (Table 3). Slopes for the region indicators represent the difference with region North.

boundary layer, which causes a spatially more diffuse column signal, (b) the relatively large satellite pixel size compared to the size of Dutch cities and (c) retrieval difficulties related to multiple scattering, aerosol interference, and advection of NO<sub>2</sub>.

A further major issue in application of satellite column data is how to convert a column into a surface NO<sub>2</sub> concentration. To that end, we need to understand the NO<sub>2</sub> profile shape at the size of a satellite pixel with good accuracy. Current recipes to establish this ratio either (1) fully rely on chemistry-transport models (CTMs) to simulate the profile shapes (e.g. Novotny et al., 2011; Bechle et al., 2013), or (2) use empirical relationships based on satellite columns and collocated surface NO<sub>2</sub> measurements, discarding the shape of the profile altogether (e.g. Lee et al., 2009; Wallace and Kanaroglou, 2009). Although these approaches have had some success in predicting seasonal and annual mean surface NO<sub>2</sub> levels mainly over North America, they provide limited insight in how the shape of the NO<sub>2</sub> profile actually ties the satellite column to a surface concentration, and there are serious concerns on the effective spatial resolution of model-based surface-to-column ratios, and on assumptions regarding the mixing of NO<sub>2</sub> throughout the boundary layer (Wallace and Kanaroglou, 2009; Dieudonné et al., 2013; Flynn et al., 2014).

#### 4.2. Land use regression models

Incorporation of satellite NO<sub>2</sub> data improved the prediction of background NO<sub>2</sub> concentrations with 10–18% compared to models including regional indicator variables or X and Y coordinates. Land use regression models for all sites including traffic locations were improved with 4–10%. In a study of European NO<sub>2</sub> surface concentrations derived from routine monitoring, European LUR models were improved by on average 5% (Vienneau et al., 2013). The best final model R<sup>2</sup> was 60% and included in addition to OMI NO<sub>2</sub> road length, natural land and total-built up area. A study in the US reported an increase in adjusted model R<sup>2</sup> of 9–11% of a national LUR model based on 423 monitors of the EPA network (Novotny et al., 2011). The final model R<sup>2</sup> was 77–78%. The model further included road length, impervious land use, vegetation, elevation and distance to coast. In a national Canadian model OMI NO<sub>2</sub> explained an additional 4% of surface NO<sub>2</sub> concentrations from the routine monitoring network (Hystad et al., 2011). Sites were primarily located away from local sources. The model R<sup>2</sup> was 73% and further contained industrial land use and road length in buffers of 2 and 10 km and summer rainfall (Hystad et al., 2011). OMI NO<sub>2</sub> explained 5–10% of the variability of the annual average NO<sub>2</sub> concentration across Australia using a network of 68 predominantly background monitoring stations across the entire continent (Knibbs et al., 2014). The model further contained large scale traffic, industry and built-up area predictors. The model R<sup>2</sup> was 81% when column OMI NO<sub>2</sub> was included as a predictor and 79% when surface estimated OMI NO<sub>2</sub> (using ratios estimated with a chemical transport model) was included, suggesting there was no benefit in transforming the column measurements to surface concentrations for this application. In our study we did not convert the column NO<sub>2</sub> to estimated surface NO<sub>2</sub> using a chemical transport model. Given the geographical scale of the conversion factors and the uncertainty of the factors, we anticipated little benefit at the scale of the Netherlands. We further did not use satellite NO<sub>2</sub> as a direct estimate of exposure in epidemiological studies, so we did not need satellite data represented at the surface concentration scale. The regression slope of our model showed an average conversion of 1.6 µg/m<sup>3</sup> per 10<sup>15</sup> molecules/cm<sup>2</sup> for the Netherlands. This agrees well with the default value of 1.9 µg/m<sup>3</sup> per 10<sup>15</sup> molecules/cm<sup>2</sup> used in the South-Californian study (Bechle et al., 2013) and 2.1 µg/m<sup>3</sup> per 10<sup>15</sup> molecules/cm<sup>2</sup> from the LUR model in the Australian

study (Knibbs et al., 2014).

Our study thus supports the use of satellite NO<sub>2</sub> as an additional predictor in development of NO<sub>2</sub> models, even for a small densely populated country with a dense ground network such as the Netherlands. More local scale predictor variables such as traffic need to be included because of the current coarse spatial scale of the satellite data. The combination of satellite and local (traffic) predictors allows development of national models that predict fine scale spatial variation, an important requirement for NO<sub>2</sub>. A recent study in 36 study areas across Europe showed that 60% of the variance of annual average NO<sub>2</sub> was within study area (Cyrys et al., 2013).

Models with satellite NO<sub>2</sub> data performed only slightly better than models that included the interpolated regional background NO<sub>2</sub>. Obtaining regional background NO<sub>2</sub> however requires a significant sampling effort, in the current study 26 sites.

Our study was based on average concentrations. A recent study documented the potential to use satellite NO<sub>2</sub> data in addition with land use and surface monitoring to predict spatially resolved daily NO<sub>2</sub> concentrations (Lee et al., 2014).

## 5. Conclusion

Annual average OMI observations of tropospheric NO<sub>2</sub> columns correlated well with annual average urban and especially regional background surface NO<sub>2</sub> concentrations. LUR models with satellite observations had slightly higher explained variance than models with interpolated regional background concentrations and much higher explained variance than models with regional indicators or geographic coordinates. Our study supports the application of OMI NO<sub>2</sub> data to assess population exposure even in a small densely populated country.

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