



A national fine spatial scale land-use regression model for ozone



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ABSTRACT

Uncertainty about health effects of long-term ozone exposure remains. Land use regression (LUR) models have been used successfully for modeling fine scale spatial variation of primary pollutants but very limited for ozone.

Our objective was to assess the feasibility of developing a national LUR model for ozone at a fine spatial scale. Ozone concentrations were measured with passive samplers at 90 locations across the Netherlands (19 regional background, 36 urban background, 35 traffic). All sites were measured simultaneously during four 2-weekly campaigns spread over the seasons. LUR models were developed for the summer average as the primary exposure and annual average using predictor variables obtained with Geographic Information Systems.

Summer average ozone concentrations varied between 32 and 61 $\mu\text{g}/\text{m}^3$. Ozone concentrations at traffic sites were on average 9 $\mu\text{g}/\text{m}^3$ lower compared to regional background sites. Ozone correlated highly negatively with nitrogen dioxide and moderately with fine particles. A LUR model including small-scale traffic, large-scale address density, urban green and a region indicator explained 71% of the spatial variation in summer average ozone concentrations.

Land use regression modeling is a promising method to assess ozone spatial variation, but the high correlation with NO_2 limits application in epidemiology.

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1. Introduction

High ground level ozone concentrations affect human health and vegetation (World Health Organization, 2006; World Health Organization, 2013; EPA, 2013). Ozone is a secondary pollutant formed in the atmosphere from precursors, primarily nitrogen oxides and volatile organic compounds (VOC) influenced by UV radiation (EPA, 2013). Motorized traffic and industry are main anthropogenic sources of nitrogen oxides and volatile organic compounds (World Health Organization, 2006; EPA, 2013). Because the scavenging effect of NO near sources dominates the slower formation processes, ozone concentrations are reduced in urban areas and especially near major roads (EPA, 2013). Ozone formation may result in higher ozone concentrations at distances of tens of km's downwind from source areas (EPA, 2013; European Environment Agency, 2009). Ground-level ozone concentrations may also be affected by stratospheric intrusion, particularly in spring time and because of the long atmospheric lifetime of ozone

above the boundary layer by continental transport (EPA, 2013).

Comparison with limited measurements in the late 19th century suggest that average ozone concentrations may have more than doubled or tripled in the 20th century (Vingarzan, 2004; Intergovernmental Panel on Climate Change IPCC, 2007). An assessment based upon monitoring data from the last few decades and a chemical transport model showed a slight decrease in ozone-concentrations in Europe related to reductions in total NO_x emissions (European Environment Agency, 2009). Trends were however not consistent across countries and studies in the UK have shown an average upward trend in ozone-concentrations at remote rural locations and a downward trend for rural and urban sites in the past two decades (Jenkin, 2008). In the US, overall ozone concentrations have been reduced somewhat in the past decade, with some heterogeneity (EPA, 2013). Ozone will likely remain an important environmental health issue, especially given projected increases in temperature related to climate change (Intergovernmental Panel on Climate Change (IPCC), 2007).

Effects of short-term exposure on respiratory morbidity and mortality have been well documented (World Health Organization, 2006; World Health Organization 2013; EPA 2013). Recent studies have suggested associations between long-term exposure

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to ozone and lung function growth, asthma incidence and asthma severity (World Health Organization, 2013; EPA 2013). In 2013, US EPA judged long-term ozone exposure as likely causally related to respiratory outcomes, but the evidence for other health outcomes was rated as at best suggestive (EPA, 2013). Compared to the large literature on long-term exposure effects of particulate matter and nitrogen dioxide, few studies have evaluated health effects related to long-term ozone exposure.

Epidemiological studies of long-term ozone exposure have generally characterized exposure by averaging concentrations from all monitors in the metropolitan area (Jerrett et al., 2009) or by interpolation of monitoring data (Lipsett et al., 2011; Jerrett et al., 2013). Epidemiological studies of particulate matter and nitrogen oxides have increasingly accounted for intra-urban variability in concentrations using dispersion models or land use regression (LUR) models (Jerrett et al., 2005; Hoek et al., 2008). Routine network measurements are typically not spatially dense enough to use directly as exposure estimates on address level of participants of an epidemiological study. Interpolation methods including kriging and inverse distance weighing are useful for characterizing the spatial variation of regional background concentration, but typically cannot assess the fine scale variation related to proximity to sources (Jerrett et al., 2005). Land use regression has been used successfully in mapping concentrations of pollutants such as NO_2 , $\text{PM}_{2.5}$, the soot content of $\text{PM}_{2.5}$ and VOC's

(Jerrett et al., 2005; Hoek et al., 2008). Fine scale LUR models have been reported for ozone in a single study in two cities in Sweden with moderate success (Malmqvist et al., 2014). LUR modelling for ozone has been conducted for the Europe Union, using a coarse grid of $1 \times 1 \text{ km}^2$ in which local variations on street level were not included (Beelen et al., 2009). Ozone concentration variance was mainly explained by altitude which is not relevant in the Netherlands. For air quality management applications, ozone concentrations have been modelled extensively using chemical transport models, typically at a coarser spatial scale, e.g. $4 \times 4 \text{ km}^2$ (EPA, 2013). Kriging and Bayesian maximum entropy methods using monitoring and modelling data have been applied as well (Nazelle et al., 2010; Liu and Rossini 1996).

To generate fine scale average ozone concentrations for epidemiological studies of long-term ozone exposure, we monitored ambient ozone concentrations at 90 locations across the Netherlands using passive samplers. The purpose of this paper is to assess the feasibility to develop land use regression models, combining the measured ozone concentrations and predictors obtained with geographic information systems (GIS). An additional aim was to explore the correlation between measured NO_2 and O_3 , which is important for epidemiological studies that attempt to assess the independent effects of ozone and primary traffic-related pollutants for which NO_2 can be considered a surrogate. Therefore we additionally measured NO_2 and NO_x concentrations at the same

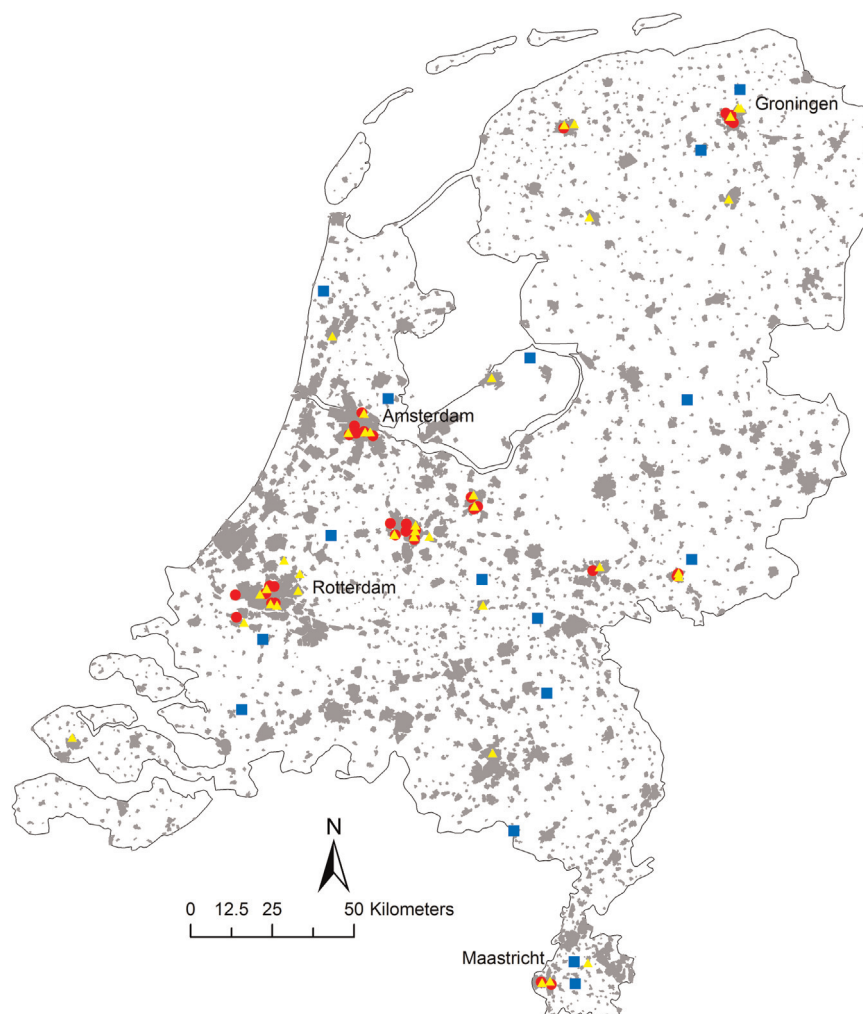


Fig. 1. Monitoring locations for ozone passive sampling.

Note: Blue squares are regional background sites, yellow triangles urban background and red circles traffic sites. Grey areas are towns. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

sites with passive samplers.

2. Methods

2.1. Sampling site selection

We selected 90 sites covering the entire country (Fig. 1, area of 37,354 km²). 68 of these sites have been used in the European Study of Cohorts for Air Pollution Effects (ESCAPE) study in 2009 for measurements of NO₂ and particulate matter concentrations (Cyrys et al., 2012; Eeftens et al., 2012a, 2012b). ESCAPE sites were selected to allow comparison of the spatial pattern of newly measured ozone concentrations with recent (2009) measurements of nitrogen oxides and particles. Twelve additional sites were selected from a project that measured nitrogen oxides at 144 sites in 2007 (Eeftens et al., 2011) and 10 new sites were added to capture the entire Netherlands, as the ESCAPE study did not cover some border regions in the country. The 90 sites were divided over three different location types, being regional background (19 sites), urban background (36 sites) and traffic locations (35 sites). Ozone concentrations in the Netherlands are routinely monitored within the framework of the National Air Quality Monitoring Network at 38 locations across the country of which 22 are classified as regional background, 10 as urban background and 6 as a traffic site. Monitoring is performed with UV photometry continuous analysers. Compared to the National Monitoring Network we selected more sites in cities and selected our regional background sites in small towns rather than outside the towns. This choice was motivated by the goal to assess population exposure. Our traffic locations are situated in major cities (Amsterdam, Rotterdam, Utrecht, cities with more than 250,000 inhabitants) and smaller cities represented in the epidemiological cohorts to which the model will be applied. Our traffic sites were located in urban streets with more than 10,000 vehicles per day (Cyrys et al., 2012). Measurements were made at the façade of homes.

2.2. Sampling and analysis of ozone

Two week average measurements were performed simultaneously at all sites in the four seasons. The specific study weeks were selected to represent the yearly average and to capture the peak and minimum of ozone concentrations during a year, based on monthly average concentrations in the National Air Quality Monitoring Network in the Netherlands from 1997 to 2009 (Supporting information Fig. S1) (Rijksinstituut voor Volksgezondheid en Milieu (RIVM), 2011). Sampling periods were selected outside the major holidays as traffic patterns may differ. The measurement periods were February 28–March 15; April 24–May 10; September 4–20 and November 28–December 14 2012. Each site was measured for 14 days within these periods. All samplers were placed on drain pipes or balconies, with a minimum of 20 cm distance from the building, at a height of approximately 3 m from the ground to prevent vandalism. Concentrations of ozone were measured with Ogawa passive samplers (Ogawa & Co. USA Inc.), following the Ogawa protocol (Ogawa, 2001). Ogawa samplers consist of a cylindrical polymer body (2 cm diameter and 3 cm length). At each end of the cylinder a nitrite based filter is placed between two stainless steel screens and sealed with a cap with holes where diffusion can take place. Ogawa samplers for ozone have been used before to map ozone concentrations (Liu and Rossini, 1996; Koutrakis et al., 1993; Geyh et al., 2000; Lozano et al., 2009; Gibson et al., 2009) and shown to agree well with continuous ozone monitors. The principle of the method is that ozone oxidizes nitrite to nitrate. To minimize the effect of wind, rain and direct solar radiation, the passive samplers were

protected by shelters designed by Ogawa & Co. After 14 days of sampling, the badges were placed in resealable bags and brought back to the laboratory, where the filter is extracted with ultra-pure (Milli-Q) water. The nitrate and nitrite concentration in the filter extract was analysed by ion chromatography (IC) with a Dionex DX100 equipped with a Dionex AS124A column. Extraction and analysis were conducted according to standard operating procedures (Brunekreef et al., 2005). The nitrate ion concentration is used to calculate the total amount of ozone collected. Nitrite was measured to check that sufficient nitrite was present on the filter. The sampling rate for the Ogawa passive sampler given in the Ogawa protocol was used as a constant of 21.8 mL min⁻¹ (Ogawa, 2001). The same sampling rate has been used different articles using the Ogawa passive sampler (Geyh et al., 2000; Gibson et al., 2009; Gerboles et al., 2006; Salem et al., 2009). The collection rate of passive samplers was shown not to be affected by temperature and humidity (Geyh et al., 2000; Gibson et al., 2009).

Measurements and analysis of NO₂ and NO_x were performed simultaneously at the same sites as the ozone measurements with passive Ogawa samplers using tri-ethanolamine coated filters and spectrophotometric analyses following the standard operating protocols of the ESCAPE project (Cyrys et al., 2012).

2.3. Quality assurance and control

In each sampling period 9 field duplicates and 9 field blanks were collected for quality control. Field blanks were used to calculate the limit of detection. The mean field blank was subtracted from the measurements. Field duplicates were used to calculate the precision of the measurements (Eeftens et al., 2012a, 2012b).

In each campaign, we compared measurements of the Ogawa badge with measured concentrations in the same 14-day period at six locations (two regional background, two urban background and two traffic sites) from the national air quality monitoring network. Ozone is measured with Thermo Scientific model 49i UV photometry continuous analysers (Thermo Scientific, West Palm Beach, USA). Sampling periods with more than 25% of the hourly network values missing were deleted from the comparison.

We further used the continuous network data to compare the full summer (April–September) and full year averages from the national monitoring network with the average of the two/four sampling periods used in this study. This comparison was made to assess how well the absolute ozone concentrations found in our campaigns, which were temporally limited, agreed with averages calculated for the complete period.

2.4. GIS predictors

Geographical Information Systems (GIS) were used to generate a total number of 91 variables to characterise the street, traffic load, land use and population density in different buffers around the coordinates of a sampling point. Definitions of the ESCAPE study were used (Eeftens et al., 2012a, 2012b). Land cover data were taken from the CORINE (Coordination of information on the environment) land cover dataset of the year 2000 (Eeftens et al., 2012a, 2012b). Supporting information Table S1 lists the variables collected. Each sampling point was carefully imported in Google Earth to set coordinates, which are used to collect all GIS information. All coordinates were also checked with a GPS. GIS data was collected with ArcGis 9.3 as in the ESCAPE study (Eeftens et al., 2012a, 2012b). Geographical location was included to further explain ozone concentrations. Indicator variables for region of the country were defined using regional patterns in network data on ozone concentrations (Rijksinstituut voor Volksgezondheid en Milieu (RIVM), 2011). The interpolated regional background concentration was also evaluated using inverse distance squared

weighed concentrations of the 19 regional background sites (Eeftens et al., 2012a, 2012b).

2.5. Site characterisation

In addition to geographical information, we also collected data of the site in terms of traffic counts, height of buildings and street characteristics using the ESCAPE protocol (Table S1). Traffic counts were performed for all 35 street locations during a period of 15 min. Distinctions were made between light vehicles, heavy vehicles, mopeds and buses. All observations were made between 10.00 am and 3.00 pm to avoid peak traffic flow. Measurements were then multiplied by 48 to get traffic intensities during day hours and next by 1.29 to calculate traffic intensities per 24 h (Van Roosbroeck et al., 2008). Most street characteristics, including height of buildings, were assessed using Google Earth. Traffic counts for the 55 background locations were obtained from the ESCAPE and Traffic Related Air pollution and Children's respiratory Health and Allergies (TRACHEA) study (Eeftens et al., 2011).

2.6. Land use regression model building and evaluation

Land use regression models were built using the ESCAPE method (Eeftens et al., 2012a, 2012b). Briefly, the method tries to maximize the percentage of explained variability (R^2) using a supervised forward stepwise procedure. For all potential predictor variables we defined a priori a direction of effect (e.g. negative for traffic intensity), see Supporting information Table S1. We did not allow positive slopes for traffic intensity because the evaluated buffer sizes ranged between 25 m and 5 km, distances at which ozone formation is likely not important. The model starts with the variable with the highest explained variance in a univariate analysis with a regression slope with the a priori defined direction. Then all other variables were added to this model separately and assessed for the increase in the adjusted explained variance. The predictor variable with the highest increase in adjusted R^2 was included in the model if the increase in adjusted R^2 is greater than 1%; the coefficient has the pre-specified direction; and the direction of effect for the predictors already in the model does not change. This procedure was continued until none of the variables could fit the criteria mentioned above. In a final step, variables with a p -value > 0.1 were removed from the model. Standard diagnostic tests for regression were applied, which included Cook's D (influence statistic required to be < 1), variance inflation factor (< 3) and residual plots to evaluate heteroscedasticity and distribution.

To represent regional variation, we separately offered regional indicator variables (north, west, east, south); combinations of X and Y coordinates and interpolated regional background concentrations. We evaluated Moran's I to test for spatial autocorrelation in the residuals of the regression models.

The final model was evaluated with 10-fold cross-validation (Wang et al., 2012). We randomly selected 90% of the sites to develop a model and then used the remaining 10% of the sites for model evaluation. This procedure was repeated ten times, such that each site was in a test set once.

Models were built with the summer average as the dependent variable as the primary analysis. The summer average was selected because key epidemiological studies were based upon the summer average concentration (Jerrett et al., 2009). The summer average was calculated from the May and September campaigns. We further assessed the annual average. Because all measurements were performed simultaneously, no adjustment for temporal variation was needed as in the ESCAPE study.

Statistical analyses were performed with the statistical programs SAS version 9.2 for Windows.

3. Results

3.1. Quality assurance and control

Laboratory blanks and field blanks were close to zero in the entire study period. Mean blank was $-0.0003 \mu\text{g}/\text{m}^3$ and the limit of detection calculated as three times the standard deviation of the blank $0.0004 \mu\text{g}/\text{m}^3$. IC analysis showed that enough nitrite was left on the filter after exposure to ozone.

A total of 35 duplicates was taken. The coefficient of variation (standard deviation divided by the mean) was 3%, indicating good precision for the first three campaigns. The nine duplicates taken in the December campaign gave a coefficient variation of 28%. Concentrations were lower in the December campaign, but expressed in absolute differences between samples and duplicates, the precision was also much poorer compared to the first three campaigns. Comparison between sample and duplicates for the NO_x measurements gave a coefficient variation of 4%, with no differences between campaigns.

Comparison of ozone measurements with the Ogawa badge and the continuous monitors are shown in Fig. S2 for the first three campaigns. The 14-day average Ogawa badge concentrations showed a 1:1 relationship with the concentrations measured with continuous monitoring of the corresponding 14-day period in the network ($R^2 = 0.97$), indicating good agreement. For the December campaign, agreement was poor: the R^2 was 0.27 and the regression model was $\text{ozone_monitor} = 20.15 + 0.28 \cdot \text{ozone_ogawa}$, very different from the equation for the first three campaigns.

Because of the poor quality of the ozone data in the fourth (December) campaign, we decided to exclude the data collected during this campaign from further modeling. We do not have a solid explanation for the poor data quality, but speculate that the lower temperature may adversely have affected sampling. In the December campaign minimum temperatures were below 0°C for half of the days, with the lowest temperature being -8.3°C . Such low temperatures did not occur in the other three campaigns. No further experiments were performed to test this hypothesis. The low ozone concentrations during the December campaign is an unlikely explanation as network ozone measurements showed that the ozone concentrations during the December and March campaign differed less than $1 \mu\text{g}/\text{m}^3$. The annual average was next calculated as the average of the May, September and twice the March campaign. The March campaign was counted twice to have the warm and cold season represented equally in the calculation of the annual average. Ozone concentrations are higher in the warm than in the cold season.

3.2. Distribution of concentrations

In the first three campaigns valid samples were collected for 260 out of 270 samples. Data from two sites were not used in further calculations due to building renovation and smoking near the sampling site, leaving 88 sites.

The correlations among the first three campaigns were between 0.68 and 0.76 (Table S2), documenting that the ranking of sites is fairly similar across campaigns. The correlation between ozone concentrations in the December campaign with the March, May, and September campaigns was low (0.21 to 0.37).

Mean concentrations were $26.5 \mu\text{g}/\text{m}^3$ in March, $51.3 \mu\text{g}/\text{m}^3$ in May and $39.2 \mu\text{g}/\text{m}^3$ in September (Table 1, Fig. S3). The summer and annual average varied substantially across sites, with almost a factor two between minimum and maximum concentration (Fig. 2, Table 1). The Pearson correlation between the summer average and annual average concentration was 0.96. Small differences were found between urban and regional background locations, but traffic locations showed significantly lower ozone concentrations

Table 1
Distribution of 14-day average ozone concentrations for all campaigns ($\mu\text{g}/\text{m}^3$).

Campaign	Mean	Standard deviation	Median	Minimum	Maximum
March	26.5	5.3	25.7	17.8	40.4
April–May	51.3	5.6	52.0	38.5	61.9
September	39.2	8.0	38.3	23.0	64.8
December ^a	14.1	5.7	13.5	5.0	30.8
Summer average ^b	45.2	6.2	45.2	31.7	60.6
Annual average ^b	35.8	5.5	35.6	25.0	47.8

^a Not used in further modeling because of unreliable measurements.

^b Summer average is the average of the April–May and September campaigns. Annual average is the average of the April–May campaign, September campaign and the March campaign counted twice.

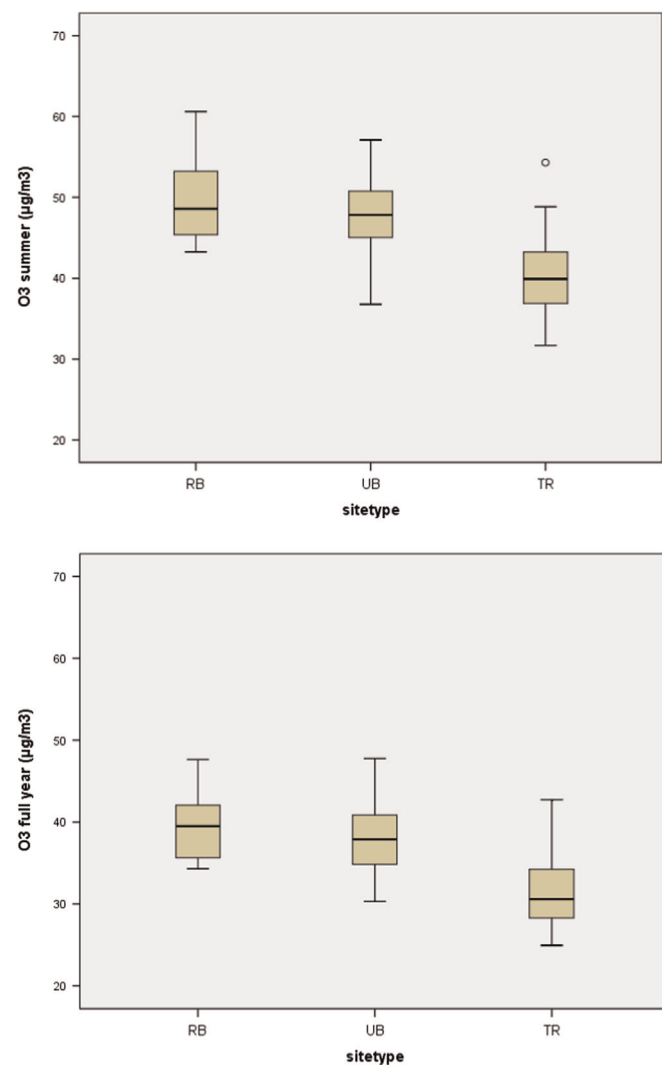


Fig. 2. Distribution of summer and full year average ozone concentration ($\mu\text{g}/\text{m}^3$) by site type.
Note: RB=regional background, UB=urban background, TR=traffic. Box indicates 25th and 75th percentile, horizontal line in box is median. Individual observations shown as outliers if more than 1.5 (o) or 3 (*) times the interquartile range away from the box.

(Fig. 2). Overall average differences between regional and urban background locations were within $2 \mu\text{g}/\text{m}^3$ for all three seasons. The summer average was $49.5 \mu\text{g}/\text{m}^3$ for regional background locations and $47.9 \mu\text{g}/\text{m}^3$ for urban background sites. When we

compared the 11 urban background sites in the three major cities (Utrecht, Amsterdam and Rotterdam) with the 7 regional background sites in the west and middle of the country, the difference in ozone concentration was similarly small ($-1.6 \mu\text{g}/\text{m}^3$). Urban background summer average concentrations in the three major cities showed limited variability: $39\text{--}50 \mu\text{g}/\text{m}^3$. Traffic sites had a summer average of $40.4 \mu\text{g}/\text{m}^3$.

The average difference between the summer average calculated from the May and September two-weekly average concentrations measured within the monitoring network and the averages for the total summer period was $2.5 \mu\text{g}/\text{m}^3$, suggesting that the temporally sparse monitoring campaigns resulted in only small underestimation of the absolute summer concentration averages (Table S3). The annual average estimate based on three sampling periods differed only $2.8 \mu\text{g}/\text{m}^3$ from the true annual average (Table S4).

3.3. Correlation of measured ozone and NO_2 concentrations

NO_2 and ozone concentrations for the summer and yearly average were strongly negatively correlated (Pearson R of -0.85 and -0.87). Fig. 3 illustrates the relationship for summer average concentrations. Correlations between NO_2/NO_x and ozone were lower when the traffic locations were excluded: $R = -0.66$ and -0.62 for NO_2 and NO_x respectively. The March, May and September campaigns also showed a high negative correlation between ozone and NO_2 concentrations of -0.83 , -0.85 and -0.71 . The correlation between O_3 and NO_2 in the December campaign was only -0.34 . Correlations between O_3 and NO_2 based upon continuous network differed by season (Table S5).

Annual average ozone concentrations correlated similarly high with annual average NO_2 concentrations measured three years earlier in the ESCAPE study ($R = -0.83$, $n = 65$ sites). Ozone was significantly negatively correlated with $\text{PM}_{2.5}$, PM_{10} and $\text{PM}_{2.5}$ absorbance concentrations of 2009 ($R = -0.67$, -0.70 and -0.82 respectively, $n = 33$ sites). The NO_2 average of the current 2012 campaign correlated very high with the NO_2 average of the 2009 ESCAPE campaign ($R = 0.97$).

3.4. Land use regression models

The final LUR model for the summer average ozone concentration is given in Table 2. Variables included in the summer average model describe traffic, urban background as well as

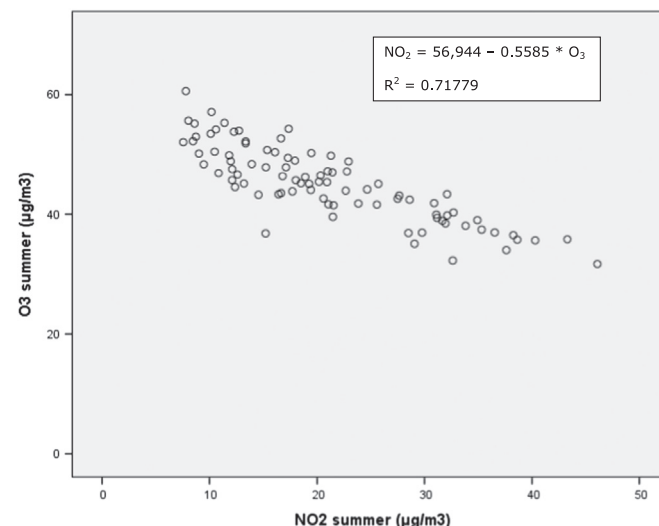


Fig. 3. Relationship between measured summer average NO_2 ($\mu\text{g}/\text{m}^3$) and ozone concentrations ($\mu\text{g}/\text{m}^3$).

regional background impacts. Traffic intensity multiplied by total road length in a 50 m buffer entered the model first, with an explained variance of 39%. A second small-scale traffic variable was the length of major roads in a 50 m buffer. Low density residential land in a buffer of 5 km, explains variability between large urban and rural areas. Regional differences are described in the model through the indicator variable North. The last variable that entered the model is green space in a buffer of 500 m. When we multiply the regression coefficients with the difference between the 90th and 10th percentile of the distribution of each predictor, the two small-scale traffic variables together and the regional indicator variable predict the largest variability in ozone concentration. The highest Cook's *D* coefficient in this model was 0.077, indicating no influential observations. Variance inflation factors were below 2, indicating no co-linearity of the predictor variables. The model R^2 was 0.71 with an adjusted model R^2 of 0.69. The root mean squared error (RMSE) was $3.5 \mu\text{g}/\text{m}^3$. Tenfold cross-validation of the ozone models resulted in a cross-validation R^2 of 0.50. There was no spatial autocorrelation in the residuals of the LUR models (Moran's *I* near zero and non-significant, $p > 0.40$).

The model developed for the annual average concentrations included exactly the same predictor variables as for the summer average (Table 2), consistent with the high correlation between the summer and annual average concentration. The model R^2 was 0.77, slightly higher than for the summer average.

Models developed for the 53 urban and regional background sites only, explained a moderately high fraction of the measured variability (Table 3). Three variables were included in the models: indicator variables for region of the country (north, west) and urban green in a 500 m buffer. The indicator variables north and the urban green variable were included in the full models as well. These models can be used in epidemiological studies focusing on subjects living away from major roads, to lower the correlation between ozone and NO_2 .

3.5. Additional analyses

When we replaced the indicator variable for region of the country with interpolated regional background concentrations from our regional background sites or functions of *X* and *Y* coordinates, the explained variance of the model decreased with 5%. Traffic variables obtained by site observations did not predict better than variables obtained by GIS. The short-term traffic counts and the indicator for canyon-type streets did not add to the explained variance of the final regression model and also did not predict better when offered as alternative traffic variables.

When we excluded all observations in the region North – leaving 69 sites in the more urbanized part of the country – the model R^2 was 60% and the coefficients of all four predictor variables remained highly significant and similar to those of the final

model, supporting the robustness of the model.

We applied the existing ESCAPE model for NO_2 in the Netherlands (Beelen et al., 2013) to further investigate the need to develop a specific ozone model. The NO_2 model explained 46% of the variability in annual average ozone concentration, substantially less than the currently developed ozone model. In the ESCAPE model for NO_2 traffic load in a 50 m buffer was included as in the ozone model, but four other traffic variables as well. Furthermore, the regional background pattern was different for both pollutants. Finally urban green was part of the ozone model, but not the NO_2 model.

Fig. 4 shows maps of the new ozone model developed and the NO_2 model from ESCAPE. Ozone concentrations were low near the main roadnetwork. NO_2 shows the opposite pattern. The NO_2 maps shows more fine scale variation, consistent with the presence of more small-scale predictors in the model.

4. Discussion

Substantial spatial variation of average ozone concentrations across the Netherlands was found. Ozone concentrations at urban background and traffic locations were 2 and $9 \mu\text{g}/\text{m}^3$ lower compared to regional background sites. A land use regression model was developed that explained 71% of the measured spatial variation in summer average ozone concentration. Predictor variables reflected especially scavenging of ozone by primary nitrogen oxide emissions: traffic intensity and major road length within 50 m reflecting local traffic; address density within a 5 km and urban green in a 500 m buffer reflecting urban scale influences; indicator for region of the country reflecting variation of regional background. High negative correlations were found between measured average ozone, nitrogen oxide and fine particle concentrations, documenting that disentangling health effects of ozone and pollutants reflecting primary traffic emission is challenging. Excluding traffic sites substantially reduced the correlation between measured concentrations of O_3 and NO_2 .

4.1. Concentration patterns

The good precision ($\text{CV} < 5\%$) and very good agreement ($R^2 = 0.97$) with continuous UV photometry monitors in the national air quality monitoring network in three of the four campaigns agrees with previous studies. A previous comparison between Ogawa passive samplers and continuous monitors showed a near 1:1 slope and R^2 values of 0.82, 0.95 and 0.95 at three comparison sites (Gibson et al., 2009). An earlier study found a coefficient variation of 9.8% comparing nitrite-impregnated filters for ozone and standard continuous monitors (Koutrakis et al., 1993).

We found lower concentrations in urban areas and especially at

Table 2
Land-use regression model for summer and annual average ozone concentration ($\mu\text{g}/\text{m}^3$).

Variable	Summer		Full year	
	Regression coefficient (SE)	Cumulative R^2	Regression coefficient (SE)	Cumulative R^2
Intercept	48.32 (0.83)		38.30 (0.64)	
Traffic intensity 50 m buffer.	−3.63 (1.18)	0.39	−2.76 (0.91)	0.36
North	6.18 (0.94)	0.59	6.87 (0.72)	0.67
Low Density residential land 5 km buffer	−4.25 (1.15)	0.64	−4.15 (0.88)	0.73
Major road length 50 m buffer	−3.84 (1.29)	0.68	−2.73 (0.99)	0.76
Urban green space 500 m buffer	1.45 (0.53)	0.71	1.10 (0.41)	0.77

SE=standard error. Regression slopes and standard errors multiplied by the difference between 90th and 10th percentile for each predictor, intercept directly from model. Difference between 90th and 10th percentile of predictors were 2,239,961.58; 1; 32,758,395.85; 184.53 and 98,014.87 respectively. The R^2 of the model is 0.71 for the summer and 0.77 for the annual average.

Table 3

Land-use regression model for summer and annual average ozone concentration ($\mu\text{g}/\text{m}^3$): background locations ($n=53$).

Variable	Summer		Full year	
	Regression coefficient (SE)	Cumulative R^2	Regression coefficient (SE)	Cumulative R^2
Intercept	47.33 (0.85)		37.36 (0.64)	
North	5.84 (1.25)	0.42	6.64 (0.94)	0.61
West	−2.12 (1.13)	0.47	−1.98 (0.85)	0.64
Urban green space 500 m buffer	1.12 (0.57)	0.50	0.70 (0.43)	0.67

SE=standard error. Regression slopes and standard errors multiplied by the difference between 90th and 10th percentile for each predictor, intercept directly from model.

traffic sites compared to regional background sites. This pattern is consistent with scavenging of O_3 by NO , emitted by combustion sources including motorized traffic. The difference between regional and urban background is larger in the Dutch national monitoring network, where the difference between urban and regional background locations is approximately $5 \mu\text{g}/\text{m}^3$ (Rijksinstituut voor Volksgezondheid en Milieu (RIVM), 2011). The larger difference in the network can be explained by the fact that in the network regional sampling locations are located outside any town, whereas our measurements were mainly conducted in small villages, because our goal was to assess population exposure. Our urban background locations further included smaller cities. However also the urban background in the three major cities was only $1.6 \mu\text{g}/\text{m}^3$ lower than at nearby regional sites. The variability of the urban background was relatively small in the major cities, consistent with observations for US cities such as Atlanta (EPA, 2013) and a recent study in Malmö, Sweden (Malmqvist et al., 2014). In other large US cities such as Los Angeles, substantial variability of urban background concentrations occurred (EPA, 2013).

4.2. Previous land use regression studies

A European scale land use regression model for ozone had model R^2 values of 0.62 at the rural scale and 0.06 at the urban scale (Beelen et al., 2009). Model performance cannot be compared directly with our study as the previous model was developed for background locations across the European Union at $1 \times 1 \text{ km}^2$ grids. Rural variations were mainly explained by altitude, which are not relevant explaining variations in ozone levels in the Netherlands alone. LUR models for Malmö and Umea, Sweden explained 40% and 67% of the variability of average ozone concentrations using similar predictor variables and model development procedures as in the current study (Malmqvist et al., 2014). The modest explained variance in Malmö was likely due to low variability of concentrations. The structure of the Umea model was very similar to our model (Malmqvist et al., 2014). A spatio-temporal model for daily daytime average ozone concentrations from routine monitors in Quebec (Canada), improved ozone predictions compared to kriging of monitored data (Adam-Poupert et al., 2014). The LUR model explained 47% of the variability in measured daily ozone concentrations, including temporal (temperature, precipitation, day of year, year) and spatial predictors (road density, latitude).

The explained variance of the model of 0.71 was within the range of model R^2 reported for well-studied pollutants such as NO_2 and $\text{PM}_{2.5}$ (Hoek et al., 2008). Compared to the recently published

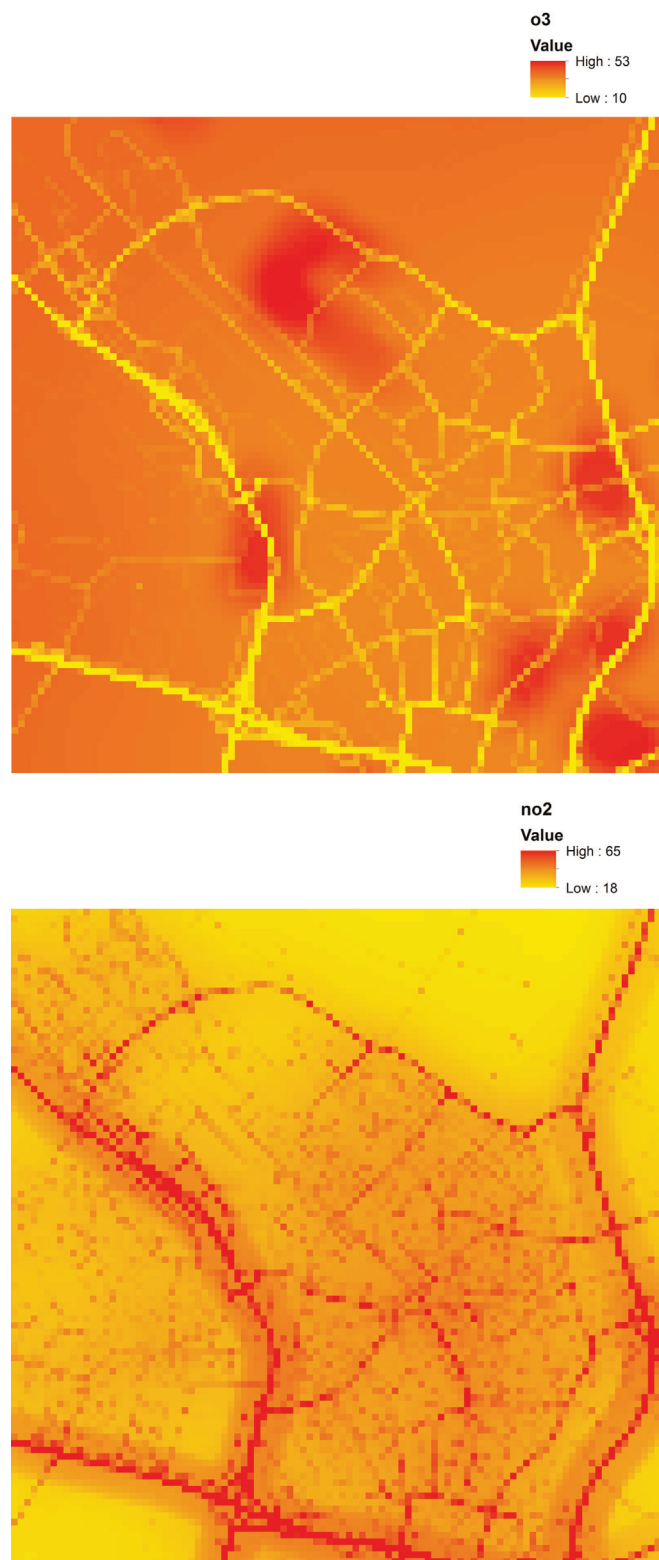


Fig. 4. Maps of the model predictions for annual average ozone and NO_2 concentration for the city of Utrecht, based on $50 \times 50 \text{ m}^2$ grids in a $10 \times 10 \text{ km}^2$ domain.

models from the ESCAPE study in the Netherlands, our model R^2 was lower than reported for NO_2 ($R^2 = 0.86$) and $\text{PM}_{2.5}$ absorbance ($R^2 = 0.92$) but somewhat higher than for $\text{PM}_{2.5}$ ($R^2 = 0.67$), coarse PM ($R^2 = 0.51$) and sulphur content of $\text{PM}_{2.5}$ ($R^2 = 0.32$) (Eeftens et al., 2012a, 2012b; Beelen et al., 2013; De Hoogh et al., 2013). We compared extensively with the ESCAPE study because the same predictor variables and model identification method were used

and the sampling sites overlapped substantially with the ESCAPE study. It is plausible that model R^2 is lower for components which are more secondary compared to nitrogen oxides and soot which are directly emitted with motorized traffic as an important source. Most predictor variables available in a GIS represent source terms (e.g. traffic intensity, population density) or simple distance to a source. Formation of a secondary pollutant is not well represented in a land use regression model, except with distance. Inclusion of a large-scale chemical transport model within the LUR framework could improve model fit, possibly using Bayesian approaches as recently applied for NO_2 (Akita et al., 2014).

The final model included three variables with a negative slope, reflecting scavenging by primary NO emissions by local traffic (traffic load and major road length both in a 50 m buffer) and the collection of urban sources (address density in a 5 km buffer). At the scale of urban areas in the Netherlands, the reaction of ozone with NO is more important than the formation of ozone from precursor emissions because of the faster reaction rates. Urban green in a 500 m buffer and region North increased levels of ozone in the model. Urban green includes green areas larger than 25 ha within urban areas, including public parks, cemeteries with vegetation, private green areas and sport fields. The urban green effect could be explained by absence of primary NO sources near the site or less likely at the 500 m scale by biogenic VOCs increasing ozone formation. During the growing season, especially when temperatures are higher, plants emit highly reactive hydrocarbons (San-derson et al., 2003) which in the presence of ozone precursor such as NO_x increase levels of ozone (EPA, 2013). The regional variable North was constructed based upon previous knowledge that levels of ozone differ between North and South of The Netherlands. Higher concentrations in the much less populated North may reflect the combination of less scavenging by NO and formation of ozone from precursors emitted by the large population centers in the west and south west of the country. The predominant wind direction in the Netherlands overall and during the summer two-weekly measurements was south-west.

The hold-out validation (HV) R^2 of 0.50 was 0.19 lower than the model adjusted R^2 , comparable to previous observations for LUR models of NO_2 (Wang et al., 2012). In the previous study based on passive NO_2 measurements across the Netherlands and the same model development method, adjusted R^2 versus HV R^2 were 0.85 versus 0.67 for 48 sites and 0.82 versus 0.73 for 96 sites. The moderate HV R^2 suggests significant misclassification of ozone exposures may occur.

4.3. Correlation between ozone, NO_2 and PM: implications for epidemiology

Spatially measured ozone and NO_2 concentrations showed a high negative correlation. Consistently, the ozone LUR model contained similar but not identical variables as the ESCAPE NO_2 model for the Netherlands. Beelen et al. (2013) Traffic load in a 50 m buffer was common to both models. Both models also contained urbanization in a large buffer, further small-scale traffic variables and regional background variables. The variables included in the ozone model predicted measured NO_2 well ($R^2=0.83$), only slightly less than the identified NO_2 model. The high correlation is explained by the structure of the ozone model which largely reflects scavenging by NO and less the ozone formation processes. The ESCAPE NO_2 model did however explain measured ozone variation substantially less well than the newly developed ozone model, supporting there is benefit in developing ozone models. The ESCAPE model for NO_2 contained more traffic variables than the ozone model; the regional pattern was different and the NO_2 model did not contain the urban green variable.

We focused on correlation of average concentrations. Correlations likely differ seasonally and for different times of the day.

We further found high negative spatial correlations of ozone with $\text{PM}_{2.5}$ absorbance measured at 33 Dutch sites within the ESCAPE study three years earlier, reflecting again the reverse influences at traffic locations. Correlations of ozone with $\text{PM}_{2.5}$ were negative as well but more moderate ($R=-0.67$). A recent cohort study in California also reported a negative correlation of ozone with NO_2 ($R=-0.71$), but found a positive correlation between ozone and $\text{PM}_{2.5}$ ($R=0.56$). Jerrett et al. (2013) The difference between our and the California study is likely due to differences in topography and distribution of sources. The Netherlands has very small altitude differences and much of the limited variability in $\text{PM}_{2.5}$ is related to traffic.

The negative spatial correlation between average ozone and other pollutants implies that studies of NO_2 and PM in the Netherlands ignoring ozone may be biased to the null if ozone affects health. The correlation with NO_2 and $\text{PM}_{2.5}$ absorbance is likely too high to assess the independent health effect of ozone and NO_2 and $\text{PM}_{2.5}$ absorbance in an epidemiological study. To disentangle ozone and NO_2 effects, the analysis could be limited to subjects living at regional and urban background conditions. The correlation between ozone and NO_2 was lower than 0.7 and substantial variability remained across sites (37–61 $\mu\text{g}/\text{m}^3$ summer average). In the Netherlands about 5–10% of the population lives in busy streets. For those subjects, the model developed upon background locations only (Table 3) could be used.

4.4. Strength and limitations

This study intended to capture the spatial variation of ozone in the Netherlands. It used a large number of measurement sites located especially in urban areas to develop land use regression models for population exposure assessment. The model could explain a large fraction of the spatial variance of ozone, especially in the context of ozone being a secondary pollutant.

Temporal coverage of the year was limited as in all purpose-designed field campaigns involving a large number of sites. Although the 14-day average concentrations measured with the passive sampler agreed very well with the continuous monitor, we could not calculate day-time average or 8-h maximum ozone concentrations. Hence, we could not develop the spatiotemporal models recently developed in Quebec (Adam-Poupert et al., 2014) Measurements in the December campaign were of low quality limiting our ability to assess annual averages. We did not incorporate meteorological data, as differences in long-term temperature are small across the Netherlands. The LUR model was developed using the ESCAPE methodology. Different methods have been applied to identify models, but few comparisons have been made of the performance of these methods (Basagaña et al., 2012). A methodological study in Girona found only small differences in performance of the ESCAPE methodology and the Deletion Substitution Algorithm (Basagaña et al., 2012).

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at <http://dx.doi.org/10.1016/j.envres.2015.04.014>.

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