



# Robustness of intra urban land-use regression models for ultrafine particles and black carbon based on mobile monitoring



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## ABSTRACT

Land-use regression (LUR) models for ultrafine particles (UFP) and Black Carbon (BC) in urban areas have been developed using short-term stationary monitoring or mobile platforms in order to capture the high variability of these pollutants. However, little is known about the comparability of predictions of mobile and short-term stationary models and especially the validity of these models for assessing residential exposures and the robustness of model predictions developed in different campaigns.

We used an electric car to collect mobile measurements ( $n = 5236$  unique road segments) and short-term stationary measurements ( $3 \times 30$  min,  $n = 240$ ) of UFP and BC in three Dutch cities (Amsterdam, Utrecht, Maastricht) in 2014–2015. Predictions of LUR models based on mobile measurements were compared to (i) measured concentrations at the short-term stationary sites, (ii) LUR model predictions based on short-term stationary measurements at 1500 random addresses in the three cities, (iii) externally obtained home outdoor measurements ( $3 \times 24$  h samples;  $n = 42$ ) and (iv) predictions of a LUR model developed based upon a 2013 mobile campaign in two cities (Amsterdam, Rotterdam).

Despite the poor model  $R^2$  of 15%, the ability of mobile UFP models to predict measurements with longer averaging time increased substantially from 36% for short-term stationary measurements to 57% for home outdoor measurements. In contrast, the mobile BC model only predicted 14% of the variation in the short-term stationary sites and also 14% of the home outdoor sites. Models based upon mobile and short-term stationary monitoring provided fairly high correlated predictions of UFP concentrations at 1500 randomly selected addresses in the three Dutch cities ( $R^2 = 0.64$ ). We found higher UFP predictions (of about 30%) based on mobile models opposed to short-term model predictions and home outdoor measurements with no clear geospatial patterns. The mobile model for UFP was stable over different settings as the model predicted concentration levels highly correlated to predictions made by a previously developed LUR model with another spatial extent and in a different year at the 1500 random addresses ( $R^2 = 0.80$ ). In conclusion, mobile monitoring provided robust LUR models for UFP, valid to use in epidemiological studies.

## 1. Introduction

Traffic is considered a major source of intra-urban air pollution (Morawska et al., 2008; Ghassoun et al., 2015). Multiple studies have linked traffic proximity and traffic related air pollution to increased risks of adverse health effects (Brook et al., 2010; Hoek et al., 2010). With about 75% of the population living in urban environments in Europe (Environmental, 2016), it is important to characterise intra-

urban air pollution with high spatial-resolution, especially for primary pollutants that exhibit large spatial variability within city limits such as ultrafine particles (UFP) and black carbon (BC) (Morawska et al., 2008; Van den Bossche et al., 2015; Peters et al., 2014). UFP and BC measurements are therefore increasingly performed with densely distributed networks or mobile platforms. Mobile monitoring provides the possibility to sample more spatially diverse environments in less time, with a limited number of monitoring devices. This is cost-effective and

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especially within city limits, it can capture the high variability of UFP and BC in a complex urban terrain (Zwack et al., 2011a, 2011b).

Several land use regression (LUR) models for UFP and BC have been developed using mobile measurements in North America (Hankey and Marshall, 2015; Farrell et al., 2016; Weichenthal et al., 2016a, 2014; Patton et al., 2014; Sabaliauskas et al., 2015; Larson et al., 2009) and Europe (Hasenfratz et al., 2015; Kerckhoffs et al., 2016), with promising results for effective exposure assessment. Mobile monitoring campaigns that developed LUR models used bikes (Hankey and Marshall, 2015; Farrell et al., 2016), cars (Weichenthal et al., 2016a; Patton et al., 2014; Larson et al., 2009; Kerckhoffs et al., 2016), public transport (Hasenfratz et al., 2015) or walking with backpacks (Weichenthal et al., 2014; Sabaliauskas et al., 2015) to collect their data. In a previous study, we developed UFP and BC models based on mobile measurements and found a high correlation ( $R^2 \sim 0.88$ ) of model predictions with LUR models based on short-term stationary measurements (30 min) from a combined (mobile and stationary) measurement campaign in two cities in The Netherlands (Kerckhoffs et al., 2016). The mobile model for UFP and BC did predict substantially (30–50%) higher concentrations than the short-term stationary model.

Although these results were encouraging for the application of LUR models based on mobile monitoring campaigns in epidemiological research some questions remain. First, we want to confirm our previous observation of high correlation of mobile versus short-term models in a new campaign involving additional cities in a different year. Second, in contrast to our previous study we added home outdoor measurements (3 times 24 h) allowing an unbiased comparison of the validity of both approaches. Third, we address the systematic difference in predicted concentration levels between mobile and short-term stationary models by exploring several methodologies to try to correct for this systematic difference. Fourth, we were interested if the derived LUR models are stable over space and time by comparing models derived from two independent measurements campaigns performed in 2013 and 2014/2015.

## 2. Methods

### 2.1. Study design

We used five different sets of data as can be seen in the Graphical abstract and Supporting information Table A.1. Four of them (on the left of the red dotted line) were collected and retrieved from the EXPOmICS campaign, conducted in 2014/2015. Mobile measurements from the MUSiC campaign in 2013 (right side) were used in additional analyses. The MUSiC measurements and models have been extensively described in previous publications (Kerckhoffs et al., 2016; Montagne et al., 2015; Klompaker et al., 2015). Data from the EXPOmICS campaign (Vineis et al., 2016) in the Netherlands consists of mobile, short-term stationary, and home outdoor 24 h air pollution measurements. The study design and models, based upon short-term stationary monitoring in six study areas including the Netherlands, have been reported before (Nunen et al., 2017).

We gathered mobile measurements between short-term stationary measurements (30 min) when driving from one site to the next; 240 short-term stationary sites and 5236 unique road segments were sampled in the winter, spring and summer in 2014/2015. Measurements were about equally divided over 84 days and started after 9:15 A.M. and stopped before 4:00 P.M. About 8 short-term sites were sampled each day over 8–10 routes per city and per season. This way, we captured the within-day, day-to-day and seasonal variability of UFP and BC concentration levels (Padró-Martínez et al., 2012). Rush hour traffic was avoided for better comparability between road segments. Short-term stationary sites were selected with a wide range of traffic characteristics and land use in and around the cities of Amsterdam, Utrecht and Maastricht, The Netherlands. We selected traffic sites (> 10,000 vehicles per day (Weijers et al., 2004)), urban background sites,

industrial areas, sites near urban green, regional background sites and sites near rivers or canals (Nunen et al., 2017). In further comparisons between traffic sites and urban background sites, all sites that are not traffic sites are considered urban background sites.

Short-term stationary and on-road measurements were made using an electric vehicle (REVA, Mahindra Reva Electric Vehicles Pvt. Ltd., Bangalore, India). A condensation particle counter (TSI, CPC 3007) and a micro Aethalometer (AethLabs, CA, USA) were used to monitor UFP and BC concentrations respectively. The CPC had a measurement every second, whereas the Aethalometer averaged measurements over one minute. The geographical location of the electric car was recorded using a Global Positioning Unit (GPS, Garmin eTrex Vista) and linked to the instruments in the car based on date and time.

To compare the predictions of UFP and BC exposure from mobile and short-term LUR models in the general population we used 1500 randomly selected addresses equally divided between Amsterdam, Utrecht and Maastricht. Furthermore, three temporally adjusted 24-h measurements of UFP and PM<sub>2.5</sub> absorbance (as a proxy for BC) were performed at home (outdoor) addresses at 42 locations in Utrecht and Amsterdam, according to protocols described by van Nunen et al. (2017) and Eeftens et al. (2012). UFP measurements were monitored using MiniDiSCs (Testo AG, Lenzkirch, Germany) which sampled every second. Previous studies have shown good agreement between CPCs and MiniDiSCs with limited differences in absolute values (Asbach et al., 2012; Meier et al., 2013). PM<sub>2.5</sub> absorbance samples were measured using Harvard Impactors and were found to be highly correlated with Black carbon (Eeftens et al., 2012). These external addresses are referred to as “home outdoor sites” and used to compare LUR estimates at the home location from the mobile and short-term stationary LUR models (external validation).

### 2.2. Data aggregation

Following our previous mobile monitoring measurement campaign (Kerckhoffs et al., 2016), we corrected for small spatial errors of the GPS by assigning all GPS points to the nearest road they were supposed to be on. Then we calculated average concentration levels of UFP per road segment, defined as a part of a road between two consecutive intersections (Farrell et al., 2016; Weichenthal et al., 2016a; Sabaliauskas et al., 2015). Road segments in tunnels or on bridges were deleted from the dataset, as they are not representative for concentrations at residential addresses. Road segments were on average 110 m long and accumulated 25 s of UFP data over the study period.

BC concentrations were sampled at a one-minute interval, but this is often too short to detect reliable changes in concentration levels (Kerckhoffs et al., 2016; Hagler et al., 2011). To reduce the noise of the instrument Hagler et al. (2011) proposed a method to only assign minute averages when the attenuation value of the filter in the instrument increased sufficiently. In our campaign this meant that about one measurement was obtained every two or three minutes. So, minute values with a too small change in attenuation (> 75% of the values) were averaged over time until the criteria was met. These values were then assigned to every road segment the car was on in that period (on average 7 road segments, ~ 140 s). When the BC measurement changed during a road segment, an average was calculated.

### 2.3. Data processing

UFP values of 500 particles/cm<sup>3</sup> or less were removed from the data set, as these reflect malfunctioning of the instrument. If the UFP data increased or decreased in one second by a factor 10 or more, the data was removed as well. Both criteria were used in previous studies (Kerckhoffs et al., 2016; Montagne et al., 2015; Klompaker et al., 2015) and resulted in less than 1% removal of UFP data. We defined observations during mobile monitoring influenced by local exhaust plumes if UFP concentration was three standard deviations above the

previous measurement second, based on the concentrations distribution for that day. Observations remained flagged until they dropped beneath the day average plus one standard deviation. This is based on methods used by [Drewnick et al. \(2012\)](#) and [Ranasinghe et al. \(2016\)](#). For the main analyses we used all measurements, including road segments with local exhaust plumes. For a sensitivity analysis, we excluded them.

#### 2.4. Temporal variation

A reference site with the same equipment as the electric vehicle and the home outdoor measurement sites was set up near Utrecht (about 2 km outside the city border of Utrecht, 40 km to Amsterdam and 140 km to Maastricht), The Netherlands, to correct for temporal variation. We used the difference method for correcting the spatial data, following previous work in the stationary campaign ([Nunen et al., 2017](#)) and the previous mobile monitoring campaign ([Kerckhoffs et al., 2016](#)). First, the overall mean concentration of the entire campaign at the reference site was calculated. Next, for each minute at the reference site an average of 30 min around time  $x$  was calculated which was subtracted from the overall mean concentration at the reference site. The difference is then used to adjust the original concentration measured at the sampling locations. We co-located instruments when the instruments were transferred between cities to check comparability and found a median ratio (averaged over 1 min) for the CPCs of 1.09 (SD = 0.16) and 0.98 (SD = 0.63) for the Aethalometers.

#### 2.5. Model development

In accordance with our previous and most other mobile monitoring studies ([Farrell et al., 2016](#); [Weichenthal et al., 2016a](#); [Sabaliauskas et al., 2015](#); [Kerckhoffs et al., 2016](#)), we identified the middle of each road segment and used this coordinate to acquire GIS predictors for LUR modelling (overview of GIS predictors see [Table A.2](#)). In summary, a range of traffic variables was defined, including traffic intensity and road length variables (in 50–1000 m buffers); ii) land use (e.g. port, industry, urban green, airports) and population / household density in buffers from 100 to 5000 m. Inverse distance to roads was used in the stationary model development, but not in the mobile monitoring model as this variable cannot be computed (distance is 0).

Variable selection was done using a supervised forward stepwise selection procedure ([Kerckhoffs et al., 2016](#); [Montagne et al., 2015](#)). The direction of the effect for the variables was determined a priori ([Table A.2](#)) and the variable with the highest adjusted  $R^2$  was entered first in the model. Model building stopped when new variables were not able to improve the adjusted  $R^2$ . The variables in the resulting models were checked for p-value (removed when p-value > 0.10), collinearity (variance inflation factor > 3 were removed), and influential observations (if Cook's  $D > 1$  the model was further examined). We accounted for autocorrelation in the mobile measurements using a first order autoregressive (AR-1) term in the ARIMA procedure ([Zwack et al., 2011b](#); [Farrell et al., 2016](#); [Weichenthal et al., 2014](#); [Buonocore et al., 2009](#); [Hsu et al., 2014](#)). If after adding an AR-1 term to the identified model, variables were no longer significant ( $p > 0.10$ ), they were removed from the model.

#### 2.6. Mobile LUR models versus short-term stationary LUR models

Mobile models of the 2014/2015 campaign were compared to short-term stationary models using different analyses, schematically shown and according to the numbers in the Graphical abstract. First, we predicted concentration levels at stationary measurement sites using the mobile LUR model and compared them to their respective short-term stationary measurements (1). Second, we compared mobile and short-term stationary models by predicting concentration on 500 random addresses in each city (2). Third, we compared stationary and mobile LUR model predictions to external average home outdoor

measurements based upon three times 24 h monitoring periods (3). In all data sets the GIS predictors were truncated to the range observed in the mobile monitoring campaign.

#### 2.7. Overestimation of mobile LUR models

We compared differences in predicted concentrations from mobile and stationary measurements for both the 2014/2015 and 2013 campaign to help understand the overestimation of mobile models from the 2013 campaign ([Kerckhoffs et al., 2016](#)). We explored four methodologies: i. using the distance between the road and the site where the prediction is made as an explanatory variable for the over-prediction; ii. LUR analyses with the delta (difference between predicted concentrations based on mobile model and observed short-term measurement) as a dependent variable with the available LUR GIS variables, iii. using a global correction based on the absolute and iv. relative differences between the predicted and measured concentration on the short-term stationary sites. Predictions based on the mobile model could then be subtracted by an absolute or relative value.

#### 2.8. Robustness of mobile LUR models

Stability of mobile LUR models was tested by comparing predictions of the mobile LUR models presented in this paper based on measurements in 2014/2015 with mobile LUR models based on measurements in 2013 ([Kerckhoffs et al., 2016](#)) (4). To rule out geographical differences between the campaigns analyses were restricted to Amsterdam, which was the only city represented in both campaigns. Other sensitivity analyses included the addition of a fixed city effect to the model, exclusion of the autocorrelation procedure, and exclusion of local emission peaks before model development.

### 3. Results

#### 3.1. Distribution of UFP and BC

The distribution of road segment averaged UFP and BC measurements is shown in [Fig. 1](#) and [Appendix Table A.3](#) and [Fig. A.1](#). Observed UFP and BC levels were on average higher on the road than at the short-term stationary sites, particularly the frequency of high UFP and BC concentrations is higher for mobile road segment averages than for short-term stationary averages. Stationary measurements are averaged over 30 min, while mobile measurements are averaged over a road segment (about 25 s), thus partly explaining the lower variability in stationary measurements. In [Fig. A.1](#), the distribution of UFP and BC measurements are stratified by city and site type (urban background (UB) and traffic). Measurements in Amsterdam were on average higher than measurements in the other two cities. Mobile UFP measurements were on average 1.44 times higher than short-term stationary UFP measurements. For BC, mobile measurements were on average 1.92 times higher ([Table A.3](#)).

#### 3.2. UFP: mobile LUR models versus short-term stationary LUR models

The developed LUR models based on UFP mobile and short-term stationary measurements are shown in [Table 1](#). Both the short-term stationary and mobile models include similar population density and traffic related variables. The short-term stationary model includes industry in a 500 m buffer whereas the mobile LUR model includes the area of ports and urban green area in the final model. As models were developed including an AR-1 term, we cannot report standard  $R^2$  values of our main mobile models. Instead, the reported  $R^2$  value is calculated by regressing the predicted concentrations based on the parameter estimates of the mobile AR-1 model without the AR-1 term. Due to the very short duration of measured concentrations and the large temporal variability, the  $R^2$  value of the mobile monitoring model is low (15%).

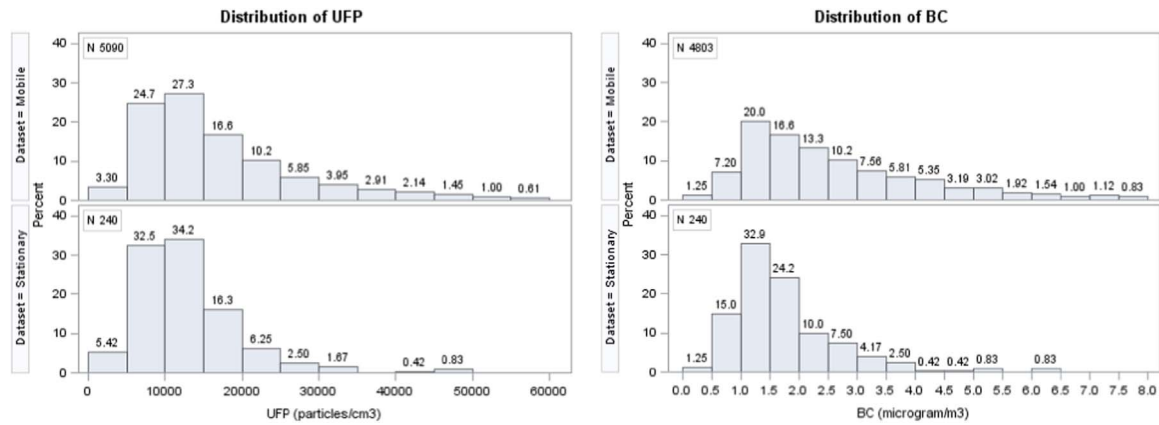


Fig. 1. Distribution of mobile and stationary UFP/BC measurements in 2014/2015. The number of mobile measurements does not match the total of road segments ( $n = 5236$ ), as the figure for UFP is cropped to a maximum 60,000 particles per  $\text{cm}^3$  and  $10 \mu\text{g}/\text{m}^3$  for BC (Max UFP = 209,140 particles per  $\text{cm}^3$ , max BC =  $38 \mu\text{g}/\text{m}^3$ ). Numbers above bars are their respective percentages of segments within that bin.

Table 1  
Mobile and short-term stationary UFP models.

Variable	UFP (particles/ $\text{cm}^3$ )	
	Short-Term	Mobile AR-1
Intercept	7784 (582)	8072 (968)
Population Density:		
Population density in a 5000 m buffer	4720 (977) <sup>a</sup>	
Residential land area in a 5000 m buffer		7763 (1155)
Traffic:		
Traffic intensity on the nearest road	2499 (860)	2244 (756)
Heavy traffic intensity on the nearest road		989 (536)
Traffic intensity in a 50 m buffer	3459 (782)	
Length of major roads in a 50 m buffer	2873 (998)	
Length of major roads in a 100 m buffer		4588 (524)
Land Use:		
Area of industry in a 500 m buffer	854 (450)	
Port area in a 5000 m buffer		3457 (995)
Urban green area in a 500 m buffer		–1001 (494)
$R^2$ of model	0.46	0.15 <sup>b</sup>
Number sites used for model development	240	5236

<sup>a</sup> Regression slopes and standard error (between brackets), multiplied by the difference between 10th and 90th percentile for all predictors.

<sup>b</sup>  $R^2$  of model without AR-1 term.

Models were also developed including a fixed effect for city. These models did not differ substantially from the original models (Table B.1). Other sensitivity analyses include models excluding the AR-1 term from model development and first excluding measurements flagged as local exhaust plumes before model development. All models are very similar and predicted concentrations based on these models on 1500 random addresses (500 per measurement city) are highly correlated ( $R^2 \sim 0.98$ ; Table B.1).

Although the LUR model for UFP explained only a small percentage of the variance in mobile measurements the model explained a much larger proportion of the variance of the short-term stationary measurements. The mobile LUR model for UFP explained 36% of the variance in the short-term stationary measurements (Fig. 2a), which is more than two times higher than the mobile model is able to explain its own measurements (15%).

Comparing predicted concentrations at random addresses ( $n = 1500$ ) revealed a strong correlation ( $R^2 = 0.64$ ) between mobile and short-term stationary model predictions (Fig. 2b). This correlation was reasonably similar for traffic and urban background sites ( $R^2$  of 0.71 vs. 0.60; results not shown).

Fig. 3 shows the correlation between predicted UFP concentrations for 42 home outdoor measurement sites and their respective average of 3 times 24 h-measurements, based on the mobile (Fig. 3a) and short-

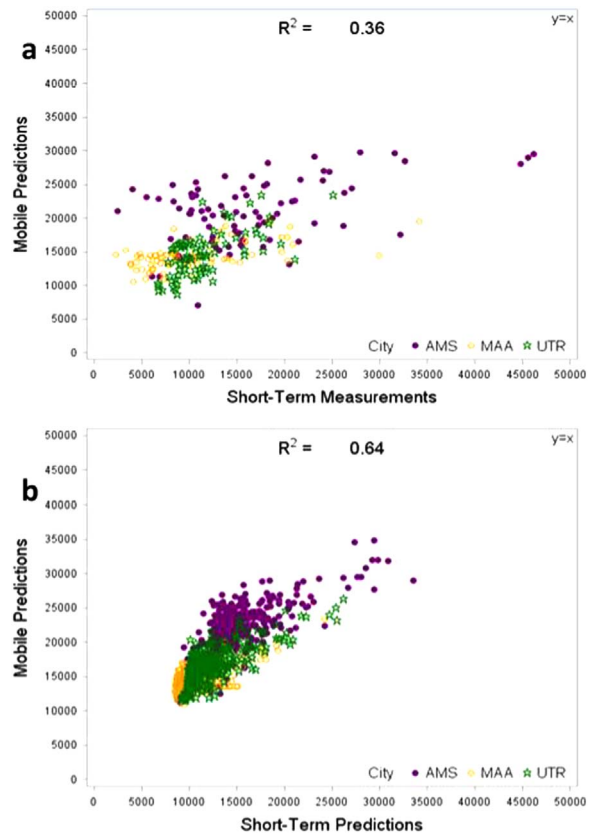


Fig. 2. (a) Predicted concentration levels (particles/ $\text{cm}^3$ ) at stationary sites based on mobile LUR model compared to stationary measurements. (b) Comparison of predicted concentration levels based on mobile and stationary LUR models at 1500 random addresses in Amsterdam (AMS), Utrecht (UTR) and Maastricht (MAA).

term stationary model (Fig. 3b). The mobile model for UFP predicts 57% of the variation in the home outdoor measurements, whereas the short-term stationary model predicts 46% of the variation. These results were consistent with new analyses of our previous campaign. The mobile model based on measurements from 2013 predicted 51% of the variance of home outdoor concentration levels in 2014/2015 (Fig. B.1).

### 3.3. BC: mobile LUR models versus short-term stationary LUR models

Like the UFP models, the mobile and short-term stationary LUR models for BC include population density and nearby traffic variables in



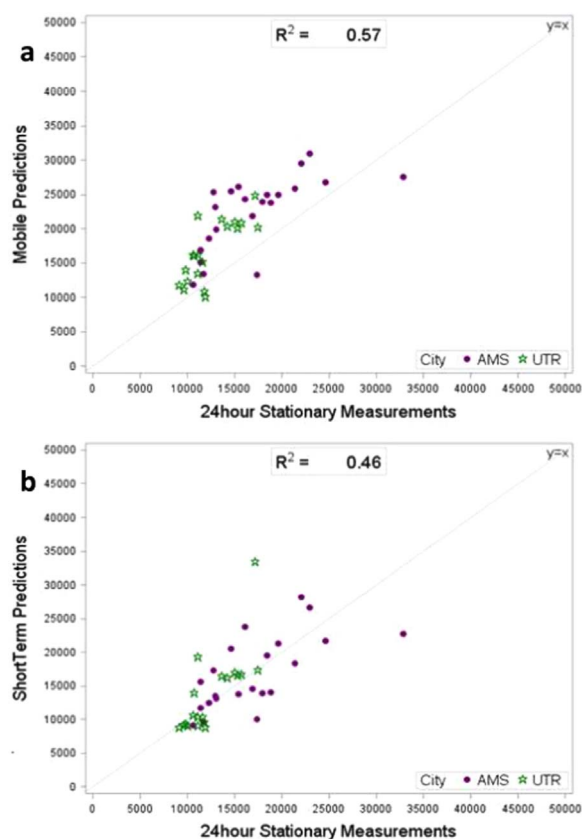


Fig. 3. Predicted concentration levels (particles/cm<sup>3</sup>) at home outdoor sites ( $n = 42$ ) based on mobile models (a) and short-term stationary models (b) compared to the average of  $3 \times 24$  h measurements at home addresses.

both models. For BC, urban green area is also included in the mobile model, similar as to the UFP mobile model. The LUR model and figures related to BC can be found in [Appendix C](#). The LUR model poorly explains the spatial variation in the mobile measurements ( $R^2 = 0.10$ ; [Table C.1](#)), comparable to the UFP model. Similarities with UFP stop when we try to use the model to predict concentration levels at the short-term stationary and home outdoor sites. The mobile model explained only 14% of the variance in the short-term stationary measurements and 14% of the variation in the home outdoor measurements ([Fig. C.1/C.2](#)). The stationary model explained 44% of the spatial variation in the stationary measurements ([Table C.1](#)) and 38% of the home outdoor measurements ([Fig. C.2](#)). Mobile BC model predictions at 1500 random households were only moderately correlated to the short-term stationary model predictions ( $R^2 = 0.37$ ; [Fig. C.1](#)).

Where the UFP mobile LUR was able to predict measurements with longer averaging periods ( $3 \times 24$  h) with greater accuracy, the mobile BC model could not. Predictions made by the mobile model based on 2013 BC measurements were also poorly correlated to home outdoor measurements in the current study ( $R^2 = 0.17$ ). Results are shown in [Fig. C.3](#), together with the results from the short-term stationary model predictions. Since mobile LUR models for BC (from 2013 and 2014/2015) did not predict the measurements with longer averaging periods well, we did not precede with further analyses of the BC LUR models in this paper. It appears, due to the long averaging time of the instrument, that our measurement device is unable to capture the fine spatial scale needed in urban settings.

### 3.4. Exploration of overestimation of mobile UFP LUR models

In all analyses we observed higher predicted concentration levels based on mobile UFP models than predictions made by short-term

Table 2

Differences between the 2013 and 2014/2015 mobile measurement campaigns.

	2014–2015 Campaign	2013 Campaign
Cities	Amsterdam, Utrecht, and Maastricht	Amsterdam and Rotterdam
Seasons	Winter, Spring and Summer	Winter and Spring
UFP over-prediction <sup>a</sup>	33% (5000 particles/cm <sup>3</sup> )	29% (4200 particles/cm <sup>3</sup> )
– Traffic	25% (3600 particles/cm <sup>3</sup> )	31% (6000 particles/cm <sup>3</sup> )
– Urban background	35% (5200 particles/cm <sup>3</sup> )	29% (4000 particles/cm <sup>3</sup> )

<sup>a</sup> Difference between predicted concentration levels based on mobile and short-term stationary LUR models, tested on 500 random addresses in Amsterdam.

stationary models, consistent with our previous work ([Kerckhoffs et al., 2016](#)). Predictions made on randomly selected addresses were on average about 5000 particles/cm<sup>3</sup> and 30% higher than models based on short-term stationary measurements ([Table 2](#)). No significant differences in overestimation were found between traffic and urban background sites. Predicted UFP concentrations based on mobile models also overestimated 24 h home outdoor measurements. The 2014/2015 mobile model overestimated the home outdoor measurements by 27% (about 4100 particles/cm<sup>3</sup> on average), whereas the short-term stationary models did not over-predict concentrations.

We explored four methodologies to correct for the difference between mobile and short-term stationary predictions. Distance to the road was not related to the difference between mobile predictions and measured UFP for the short-term stationary sites and the home outdoor sites ([Fig. B.2](#)). We also developed several LUR models with the delta as dependent variable, but could not derive a reasonable and interpretable LUR Model. The other two methods considered are to compensate the overestimation of mobile LUR models by reducing the mobile predicted levels overall by 30% or 5000 particles/cm<sup>3</sup>. These methods were also compared to the short-term stationary predictions on random addresses. In these analyses, the relative reduction of 30% to the mobile model predicted concentration seems to have a better agreement with the short-term stationary model predictions ([Fig. B.3](#)).

### 3.5. Robustness of mobile LUR models

As we conducted measurement campaigns in 2013 and 2014/15 we were interested to see if the model predictions were similar when using measurements from different geographical and temporal settings for model development. Mobile models from the 2013 campaign ([Table B.3](#)) are based on measurements in Rotterdam and Amsterdam, both industrialised and busy cities with the presence of a harbour. The mobile models from the 2014/2015 are based on the cities of Amsterdam, Utrecht and Maastricht. The cities of Utrecht and Maastricht do not have a port area and are smaller cities with less traffic than Amsterdam and Rotterdam. UFP models from both time periods were used to predict concentration levels at 1500 random addresses in Amsterdam, Maastricht and Utrecht. These predictions were highly correlated as shown in [Fig. 4](#) ( $R^2 = 0.80$ ). Predictions made by the two short-term stationary models were also highly correlated as shown in [Fig. B.4](#) ( $R^2 = 0.60$ ), but less than the mobile models.

The mobile UFP model from 2013 had a lower intercept and included natural area in a 5000 m buffer, resulting in the observed deviance in absolute concentration predictions at the lower end of the concentration range. Most of these sites are located in Maastricht, a less urban area compared to Rotterdam and Amsterdam.

To exclude the influence of geographical differences, mobile LUR models were also created for the city of Amsterdam only. These LUR models are shown in [Tables B.2 and B.3](#). Correlation between the 2013 and 2014/2015 mobile models is less than models with all cities

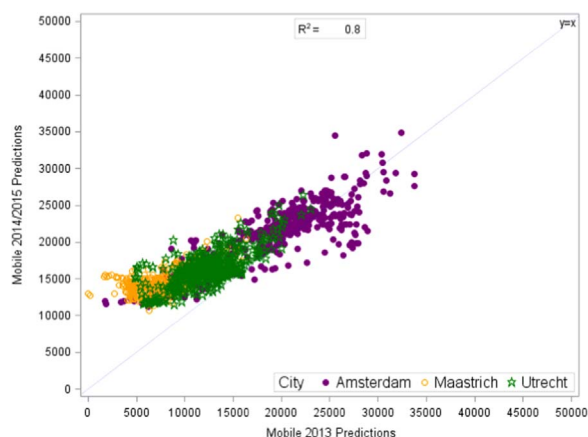


Fig. 4. Mobile predictions (particles/cm<sup>3</sup>) based on 2013 measurement campaign versus mobile predictions based on measurement presented in this paper (2014/2015), on 1500 random addresses in Amsterdam, Utrecht and Maastricht.

included ( $R^2 = 0.51$ ; Fig. B.5). Random variability due to developing models on a smaller number of sites may have contributed to the lower correlation between the two mobile models.

#### 4. Discussion

Our novel analyses demonstrate many scenarios in which LUR model predictions for UFP are robust from data collection design and sampling temporal range. Models based upon mobile and short-term stationary monitoring provided highly correlated predictions of UFP concentrations at 1500 randomly selected addresses in three Dutch cities ( $R^2 = 0.64$ ). Mobile and short-term models explained 57% and 46% of the variability in measured average home outdoor UFP concentrations at 42 external sites in Amsterdam and Utrecht. We found a high correlation ( $R^2 = 0.80$ ) between predicted UFP levels based on the mobile LUR model and a previously developed mobile LUR model (with another spatial extent and in a different year) at 1500 random addresses in Amsterdam, Maastricht and Utrecht. Predicted UFP concentrations made by the mobile models were on average 30% higher than predicted by the stationary models. Distance to the road and land-use/traffic predictors did not explain the overprediction.

In contrast, mobile model predictions for BC correlated only moderately with those of short-term stationary BC models. Mobile BC models did not explain home outdoor BC concentrations at the 42 external sites well ( $R^2 = 14\%$ ).

##### 4.1. Mobile versus short-term stationary monitoring models for UFP

Our mobile UFP LUR model explains 36% of the spatial variability of the short-term stationary measurements, which is more than two-fold the explained variance of the mobile measurements where the model is based on (15%). Similar results were found in the 2013 campaign (Kerckhoffs et al., 2016), where the mobile LUR model was able to explain 26% of the short-term stationary measurements, two times higher than the explained variability of the mobile measurements the mobile model was based on (13%). In this study we were additionally able to compare mobile and short-term models to external measurements with longer averaging periods ( $3 \times 24$  h) and found that UFP mobile models predicted an even larger fraction of the variability of these longer term measurements ( $R^2 = 0.57$ ). This analysis further supports the assertion that despite the low  $R^2$  of mobile UFP LUR models they provide robust exposure estimates at residential addresses.

The low model  $R^2$  has been attributed to the high temporal variability in measured concentrations of very short duration per site (Kerckhoffs et al., 2016; Montagne et al., 2015). Temporal predictors are purposely left out model development as we set out to develop a

spatial model. We have now documented in two combined short-term and mobile monitoring studies that the explained variance of measurements increases when the model is compared with measurements with longer duration (Kerckhoffs et al., 2016; Montagne et al., 2015; Nunen et al., 2017). This is due to the significant decrease in total variance from temporal averaging. LUR models based on longer term UFP monitoring campaigns (Abernethy et al., 2013; Hoek et al., 2011; Eeftens et al., 2016; Wolf et al., 2017; Cattani et al., 2017) explained spatial variability of their own measured UFP concentrations a lot better than our study, with  $R^2$  values ranging from 0.48 to 0.89. In the current study, an increase in the averaging time of measurements led to an increase of the ability of mobile models to predict these measurements; from 15% to mobile measurements (median 25 s), 36% to short-term stationary measurements ( $3 \times 30$  min) and 57% to home outdoor measurements ( $3 \times 24$  h). Consistently, studies that have repeated mobile monitoring at the same road segment more often than in our studies have reported fairly high model and validation  $R^2$  values (Hankey and Marshall, 2015; Weichenthal et al., 2016a; Sabaliauskas et al., 2015).

For the 2014/2015 campaign, the model predictions of the mobile and short-term model at external addresses ( $n = 1500$ ) were fairly highly correlated ( $R^2 = 0.64$ ), replicating, albeit somewhat lower, our previous observation based on the 2013 monitoring campaign ( $R^2 = 0.92$ ). The lower correlation in our current work could be due to the larger and more diverse study area. The mobile model was slightly better than the short-term stationary model in predicting concentration levels on the home outdoor sites (57% versus 46%). For the 2013 campaign, mobile and short-term stationary models explained 51% and 55% of the concentration variability at the home outdoor sites (Fig. B.1). We conclude that mobile and short-term stationary monitoring lead to very similar predictions of spatial exposure contrasts, with no consistent difference in validity.

##### 4.2. Mobile versus short-term stationary monitoring models for BC

The moderate agreement between mobile and short-term stationary model predictions for black carbon in the current study ( $R^2 = 0.37$ ) is inconsistent with our previous evaluation, based on a mobile monitoring campaign in 2013 ( $R^2 = 0.88$ ) (Kerckhoffs et al., 2016). When we compared the mobile model predictions with the home outdoor measurements from 2014/2015, we poorly explained the variability in monitored concentrations (14%). The predicted levels on these sites based on the mobile model from 2013 was also poorly correlated with the measurements ( $R^2 = 0.17$ ). The short-term stationary models in both campaigns explained more variation of the home outdoor sites ( $R^2 = 0.38$  and  $0.28$ ; Figs. C.2 and C.3).

The BC measurement device used in the 2013 and 2014/15 campaign had a temporal resolution of one minute, which was later adjusted to two or three minutes because of noise of the instrument. This is too long to detect the high spatial variation of BC, especially within city limits. The derived mobile LUR model has a relatively large estimate for residential land area in a 5000 m buffer, probably representing the difference between cities. Variation within cities could not be sufficiently assessed by our BC instrument using mobile monitoring by car driving. In contrast, short-term stationary monitoring can be performed with a Micro-Aethalometer as each measurement consist of 30 1-min averages. The Micro-Aethalometer may be useful in mobile monitoring in much higher pollution environments and in mobile monitoring campaigns using slow moving platforms such as bicycles and backpacks (whilst walking). Lonati et al. (2017) used bicycles to measure BC in a city in Northern Italy and found that the 1-min time resolution of the Micro-Aethalometer always exceeded the suggested attenuation threshold. Hankey and Marshall (2015) also needed at least 1 min averages to smooth the noise of the instrument and reported moderate model  $R^2$  for cycling-based mobile monitoring for BC (35–49%), though lower than for particle number (58–61%).

#### 4.3. Over-prediction of mobile UFP models

The mobile UFP LUR models generated higher predicted concentrations than short-term stationary models for the same locations. In our previous study, we could not distinguish between overprediction by the mobile model and under prediction of the short-term model or a combination of both. In our current study this is corroborated in the comparison of the mobile and short-term at home outdoor sites for which we had independent measurements available. The mobile but not the short-term stationary model over predicted home outdoor concentrations. This mostly related to mobile measurements being taken on-road where concentration levels are likely to be higher than at roadside residential addresses. Multiple studies have observed sharp UFP and BC gradients in near-road urban environments with gradients similar to what was observed in our previous study (Morawska et al., 2008; Van den Bossche et al., 2015; Peters et al., 2014; Weijers et al., 2004; Hagler et al., 2009; Kaur et al., 2005; Fujita et al., 2014; Baldwin et al., 2015; Hitchins et al., 2000). However, no studies have measured actual difference between measuring on-road and near the side of the road. Ragettli et al. (2014) compared measurements of UFP on the sidewalk and at the façade of buildings and found a difference in concentration levels of about 20%. Kaur et al. (2005) found a difference between measuring at the edge of the curb side near the road and measuring at the side of the building. They observed pedestrian exposure whilst walking curb side of about 86,000 particles/cm<sup>3</sup>, while an average of about 73,000 particles/cm<sup>3</sup> was measured walking along the building side of the pavement (difference about 13,000 particles/cm<sup>3</sup> which amounts to 15%). These relative differences are in the range of the finding in this paper with concentration differences between on-road and sidewalk of about 30%. This was also found in the 2013 campaign, suggesting that this number is not significantly affected by geographical differences within The Netherlands.

In our dataset we also found no correlation between the mobile model overprediction and the distance of the short-term measurement sites to the road. On top of that, LUR analyses of the delta (difference between predicted and observed at the short-term measurement site) generated no interpretable results. One of the reasons for this is probably the lack of accuracy of GIS and GPS of the measurements when it comes to differences in the range of 5–20 m. Short-term stationary sites were mostly located within 2–10 m from the edge of the road. Within these distances the mobile models are not able to scale down concentration levels to residential addresses (Kerckhoffs et al., 2016). Furthermore, mobile monitoring campaigns usually do not have short-term stationary measurements to make adjustments based on distance or LUR analyses. For the use in epidemiology, we suggest to either perform no corrections at all, as relative ranking are preserved, or use of an empirical determined factor to scale down mobile LUR model predictions, based on study-area specific data.

A rationale for no adjustment is that other factors can influence the over- or under-prediction of mobile LUR models. All our measurements are sampled between 9:15 A.M. and 4:00 P.M., excluding rush hour. This could lead to some underestimation of our LUR models. The exclusion of night-time period could in contrast lead to an over-prediction of 24 h average concentrations. Other studies only sampled during rush hour (Hankey and Marshall, 2015; Farrell et al., 2016; Weichenthal et al., 2014) or only sampled in the summer season (Hankey and Marshall, 2015; Farrell et al., 2016; Sabaliauskas et al., 2015), which respectively would cause some overestimation and underestimation (Morawska et al., 2008; Rizza et al., 2017) of concentration levels. As such the observed difference here between mobile and short-term stationary LUR models may well be within the error of other limitations in these campaigns.

#### 4.4. Robustness of mobile LUR models

We compared LUR models developed from two different monitoring

campaigns (including different cities) and found highly correlated predicted concentration levels at 1500 random addresses, providing further support for the robustness of LUR models based upon mobile monitoring data. The comparability of the two models is consistent with previous observations of stable spatial contrast of air pollution over short periods (here 1–2 years), and a previous analysis of the 2013 campaign suggesting no difference between the combined city model and city-specific models (Montagne et al., 2015). In comparable Dutch cities, similar predictor variables (mainly small-scale traffic), explain a major fraction of UFP spatial variability.

In general, models from the 2013 and this campaign included similar predictors, which was also found by Hatzopoulou et al. (2017) reviewing LUR models of several Canadian cities. Both Dutch models include a large scale population density buffer, the length of major roads in a small buffer, the area of natural land, the presence of a nearby port and traffic intensity variables. The area of airports was included in the model from 2013, but not in the 2014/2015 LUR model. It could be that the area of airports was not included our LUR model because of limited measurements near airports (only Amsterdam in the 2014/2015 campaign).

#### 4.5. Advantages and limitations of mobile monitoring

Mobile monitoring is a cost-effective method to generate LUR models, as a wide range of conditions can be captured in a limited amount of time and with a limited amount of instruments (Ranasinghe et al., 2016; Rizza et al., 2017). A high spatial density of measurements can be obtained, sampling more sites which are more representative for people's exposures such as near-intersections and close proximity to traffic lights. Conversely, mobile monitoring decreases sampling time significantly opposed to stationary measurement campaigns leading to substantial uncertainties in concentration fields (Ranasinghe et al., 2016). This is reflected in our study in very low R<sup>2</sup> values for mobile models explaining the spatial variability in mobile measurements. For LUR model development, however, the short sampling time per road segment is likely counterbalanced by the increased spatial variability (Hatzopoulou et al., 2017; Peters et al., 2013), which explains the consistent selection of explanatory variables and good external dataset prediction.

Several mobile monitoring studies suggest to use a minimum temporal resolution (Weichenthal et al., 2016a, 2016b; Apte et al., 2017) or minimum number of visits (Farrell et al., 2016) to adequately assign average concentrations per road segment. Hatzopoulou et al. (2017) looked into the amount of visits needed per road segment to characterise its average concentration and found an increase in model R<sup>2</sup> with an increasing number of visits. 20% of the road segments in our data set consist of 10 s or less. Excluding these road segments from model development increases our model R<sup>2</sup> to 0.20 (results not shown). This model however does not improve predictions to short-term stationary measurements (R<sup>2</sup> remains 0.36) and home outdoor measurements (R<sup>2</sup> of 0.56 compared to 0.57 for all road segments).

Of note, LUR models were developed using linear regression and adjusted by adding an AR-1 term to the model to correct for spatial autocorrelation. The AR-1 term assumes regular time and space intervals and that the autocorrelation remains constant over time. This method is unlikely to be optimal, but is considered the best option in several mobile monitoring campaigns (Farrell et al., 2016; Weichenthal et al., 2014; Zwack et al., 2011c). Other mobile campaigns include a Local Indicator of Spatial Analysis (LISA) (Hankey and Marshall, 2015), extend the averaging period (Fruin et al., 2008) or disregard the issue (Patton et al., 2014). Performing sensitivity analyses on the autoregressive models did not yield significant different results from the original models (Table B.1) and also in our previous campaign (Kerckhoffs et al., 2016) and in a study by Weichenthal et al. (2014).



## 5. Conclusions

Models based upon mobile and short-term stationary monitoring provided fairly high correlated predictions of UFP concentrations at 1500 randomly selected addresses in three Dutch cities. Mobile and short-term models explained 57% and 46% of the variability in measured average home outdoor UFP concentrations at external sites. In contrast, mobile BC models did not explain home outdoor BC concentrations at the external sites well ( $R^2 = 14\%$ ). We found a high correlation ( $R^2 = 0.80$ ) between predicted UFP levels based on the mobile LUR model and a previously developed mobile LUR model (with another spatial extent and in a different year) at 1500 random addresses. Because of on-road measurements predicted UFP concentrations made by the mobile models were on average 30% higher than predicted by the stationary models. Distance to the road and land-use/traffic predictors did not explain the overprediction. Overall, our study supports that robust LUR models for UFP can be developed based on mobile monitoring.

## Supplement information

The Supporting information is divided into three subsections. [Appendix A](#) contains general information concerning both UFP and BC. [Appendix B](#) contains supporting information about UFP and [Appendix C](#) about BC.

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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.2017.08.040>.

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