Robustness of Intra Urban Land-Use Regression Models for Ultrafine Particles and Black Carbon based on Mobile Monitoring

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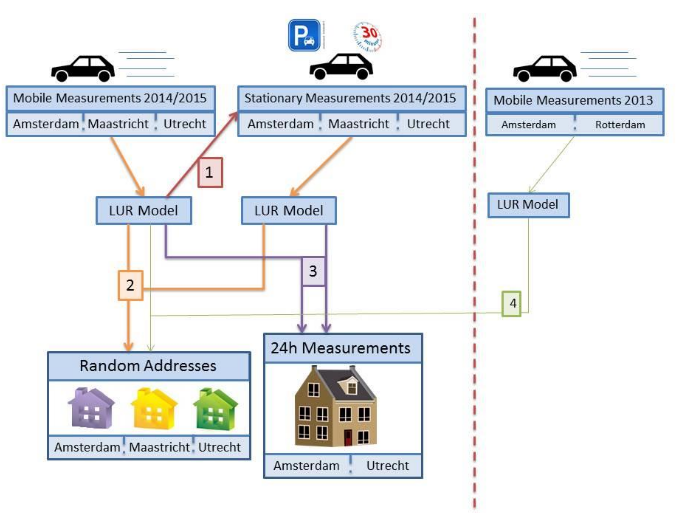
Abstract

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 (3x30min, 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 (3x24hour 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.

Keywords: Mobile Monitoring, UFP, BC, LUR models, Spatial Variation



TOC Art.

1. Introduction

Traffic is considered a major source of intra-urban air pollution[1,2]. Multiple studies have linked traffic proximity and traffic related air pollution to increased risks of adverse health effects[3,4]. With about 75% of the population living in urban environments in Europe[5], 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)[1,6,7]. 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 especially within city limits, it can capture the high variability of UFP and BC in a complex urban terrain[8,9].

Several land use regression (LUR) models for UFP and BC have been developed using mobile measurements in North America[10–16] and Europe[17,18], with promising results for effective exposure assessment. Mobile monitoring campaigns that developed LUR models used bikes[10,11], cars[12,13,16,18], public transport[17] or walking with backpacks[14,15] 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 ~ 0.88) of model predictions with LUR models based on short-term stationary measurements (30min) from a combined (mobile and stationary) measurement campaign in two cities in The Netherlands[18]. 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 24h) 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 TOC Art and supporting information table A.1. Four of them (on the left of the red dotted line) were collected and retrieved from the EXPOsOMICS 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[18–20]. Data from the EXPOsOMICS campaign[21] in the Netherlands consists of mobile, short-term stationary, and home outdoor 24h 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[22].

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 5,236 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 9:15AM and stopped before 4:00PM. 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[23]. 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[24]), urban background sites, industrial areas, sites near urban green, regional background sites and sites near rivers or canals[22]. 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-hour measurements of UFP and $PM_{2,5}$ 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[22] and Eeftens et al.[25] 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[26,27]. $PM_{2,5}$ absorbance samples were measured using Harvard Impactors and were found to be highly correlated with Black carbon[25]. 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[18], 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[11,12,15]. 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 meters long and accumulated 25 seconds 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[18,28]. To reduce the noise of the instrument Hagler et al[28] 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 sec). When the BC measurement changed during a road segment, an average was calculated.

2.3 Data Processing

UFP values of 500 particles/cm^3 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[18–20] 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.[29] and Ranasinghe et al[30]. 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 2km outside the city border of Utrecht, 40km to Amsterdam and 140km 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[22] and the previous mobile monitoring campaign[18]. 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 minutes 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 minute) 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[11,12,15,18], 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 50m to 1000m buffers); ii) land use (e.g. port, industry, urban green, airports) and population / household density in buffers from 100 to 5000m. 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[18,19]. 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[9,11,14,31,32]. 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 TOC art. 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 24hour 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[18]. 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[18] (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 figure 1 and appendix table A.3 and figure 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 30min, while mobile measurements are averaged over a road segment (about 25sec), thus partly explaining the lower variability in stationary measurements. In figure 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).

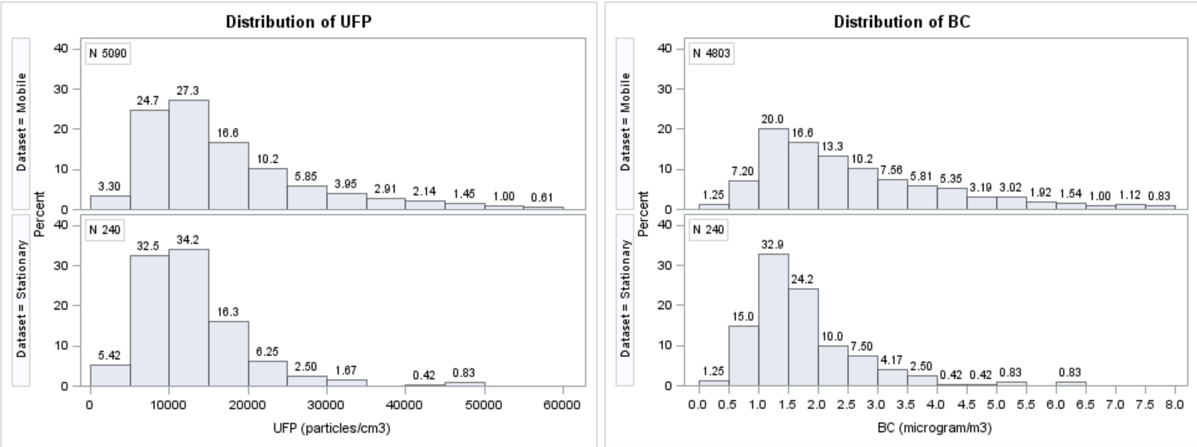


***Figure 1**. Distribution of mobile and stationary UFP/BC measurements in 2014/2015.*
The number of mobile measurements does not match the total of road segments (n=5,236), as the figure for UFP is cropped to a maximum 60,000 particles per cm^3 and 10 µg/m^3 for BC (Max UFP=209140 particles per cm^3, max BC=38 µg/m^3). Numbers above bars are their respective percentages of segments within that bin.

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 500m 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 %).

	UFP (particles/cm^3)	
Variable	**Short-Term**	**Mobile AR-1**
Intercept	7,784 *(582)*	8,072 *(968)*
Population Density:		
Population density in a 5000m buffer	4,720 *(977)* [a]	
Residential land area in a 5000m buffer		7,763 *(1,155)*
Traffic:		
Traffic intensity on the nearest road	2,499 *(860)*	2,244 *(756)*
Heavy traffic intensity on the nearest road		989 *(536)*
Traffic intensity in a 50m buffer	3,459 *(782)*	
Length of major roads in a 50m buffer	2,873 *(998)*	
Length of major roads in a 100m buffer		4,588 *(524)*
Land Use:		
Area of industry in a 500m buffer	854 *(450)*	
Port area in a 5000m buffer		3,457 *(995)*
Urban green area in a 500m buffer		-1,001 *(494)*
R^2 of model	0.46	0.15 [b]
Number sites used for model development	240	5,236

***Table 1.** Mobile and Short-Term Stationary UFP Models.*
[a] Regression slopes and standard error (between brackets), multiplied by the difference between 10th and 90th percentile for all predictors. [b] 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 ~ 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 (figure 2a), which is more than two times higher than the mobile model is able to explain its own measurements (15%).

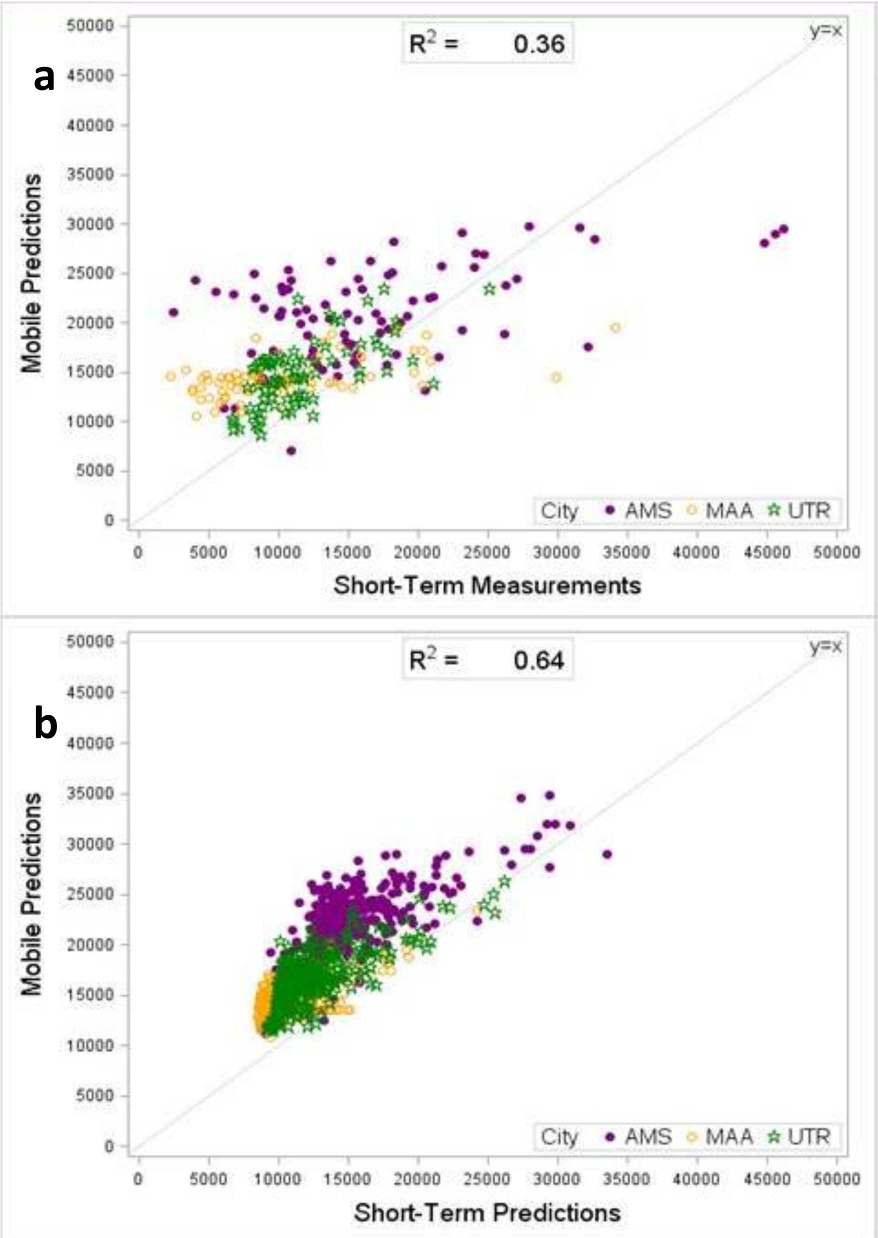


Figure 2. (a) *Predicted concentration levels (particles/cm^3) at stationary sites based on mobile LUR model compared to stationary measurements.* ***(b)*** *Comparison of predicted concentration levels based on mobile and stationary LUR models at 1,500 random addresses in Amsterdam (AMS), Utrecht (UTR) and Maastricht (MAA).*

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

Figure 3 shows the correlation between predicted UFP concentrations for 42 home outdoor measurement sites and their respective average of 3 times 24h-measurements, based on the mobile (figure 3a) and short-term stationary model (figure 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 (Figure B.1).

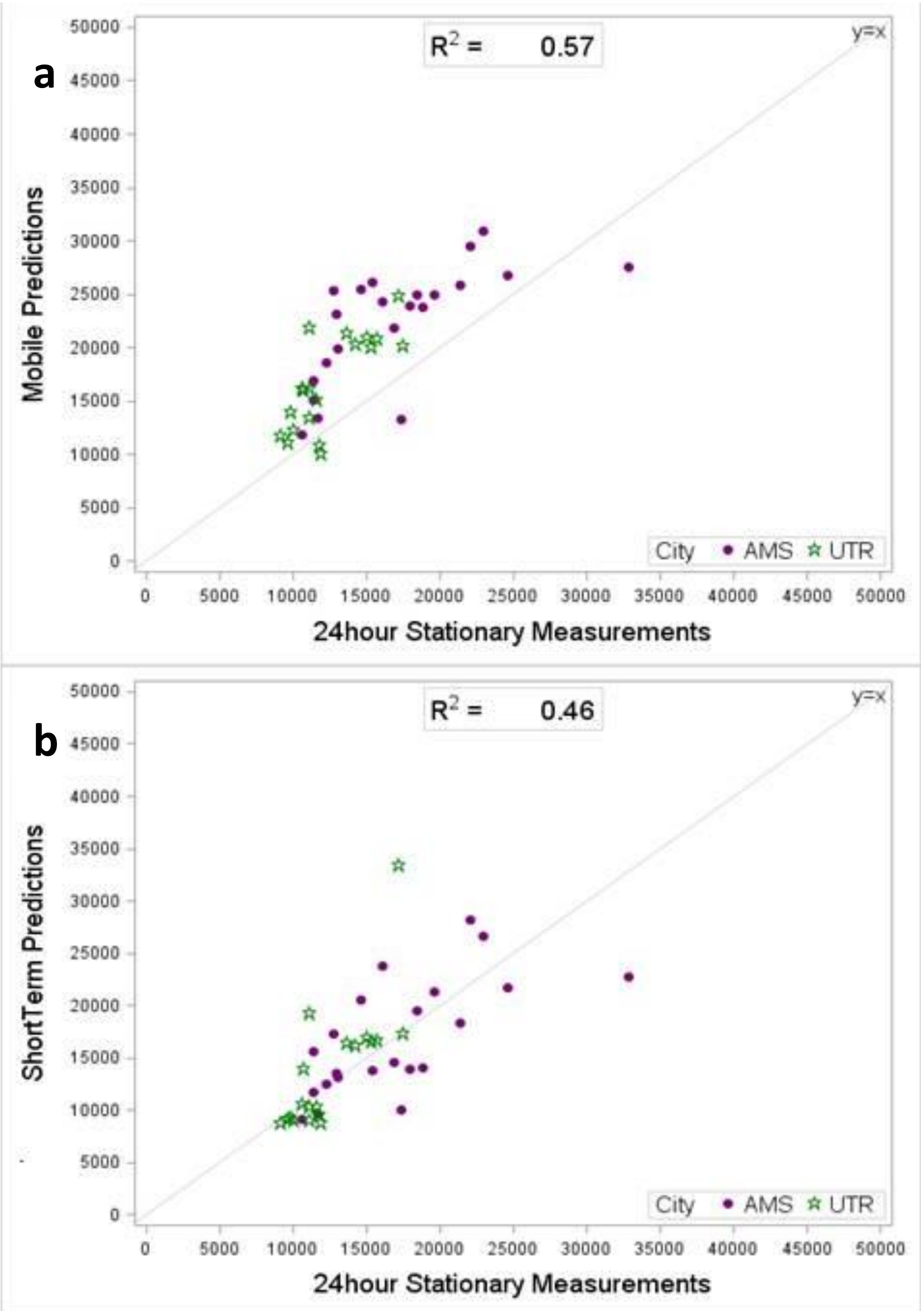


Figure 3. *Predicted concentration levels (particles/cm^3) at home outdoor sites (n=42) based on mobile models **(a)** and short-term stationary models **(b)** compared to the average of 3 x 24h measurements at home addresses.*

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 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 (Figure 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 (Figure 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; Figure C.1).

Where the UFP mobile LUR was able to predict measurements with longer averaging periods (3x24h) 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 figure 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 stationary models, consistent with our previous work[18]. Predictions made on randomly selected addresses were on average about 5000 particles/cm^3 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 24h home outdoor measurements. The 2014/2015 mobile model overestimated the home outdoor measurements by 27% (about 4100 particles/cm^3 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 (figure 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^3. 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 (figure B.3).

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

Table 2. *Differences between the 2013 and 2014/2015 mobile measurement campaigns.*
[a] Difference between predicted concentration levels based on mobile and short-term stationary LUR models, tested on 500 random addresses in Amsterdam.

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 figure 4 (R^2=0.80). Predictions made by the two short-term stationary models were also highly correlated as shown in figure 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 5000m 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.

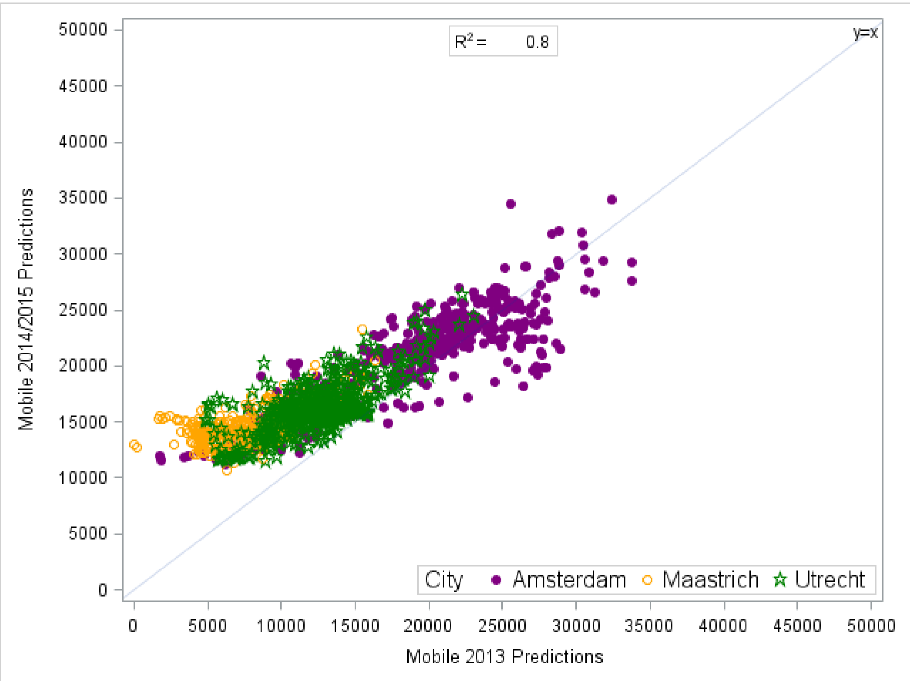


Figure 4. *Mobile predictions (particles/cm^3) based on 2013 measurement campaign versus mobile predictions based on measurement presented in this paper (2014/2015), on 1500 random addresses in Amsterdam, Utrecht and Maastricht.*

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 included (R^2=0.51; figure 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[18], 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 (3x24 hours) 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[18,19]. 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[18,19,22]. This is due to the significant decrease in total variance from temporal averaging. LUR models based on longer term UFP monitoring campaigns[34–38] 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 25sec), 36% to short-term stationary measurements (3x30min) and 57% to home outdoor measurements (3x24h). 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[10,12,15].

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 (Figure 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) [18]. 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; Figure 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 5000m 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-minute 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[39] 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[10] also needed at least 1min 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[1,6,7,24,40–44]. However, no studies have measured actual difference between measuring on-road and near the side of the road. Ragettli et al[45] 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[41] 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^3, while an average of about 73.000 particles/cm^3 was measured walking along the building side of the pavement (difference about 13,000 particles/cm^3 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 meters. Short-term stationary sites were mostly located within 2 to 10m from the edge of the road. Within these distances the mobile models are not able to scale down concentration levels to residential addresses[18]. 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 9:15AM and 4:00PM, 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 24hour average concentrations. Other studies only sampled during rush hour[10,11,14] or only sampled in the summer season[10,11,15], which respectively would cause some overestimation and underestimation[1,46] 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.

4.5 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[19]. 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.[47] 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).

4.6 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[30,46]. 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[30]. This is reflected in our study in very low R^2 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[47,48], which explains the consistent selection of explanatory variables and good external dataset prediction.

Several mobile monitoring studies suggest to use a minimum temporal resolution[12,49,50] or minimum number of visits[11] to adequately assign average concentrations per road segment. Hatzopoulou et al[47] looked into the amount of visits needed per road segment to characterise its average concentration and found an increase in model R^2 with an increasing number of visits. 20% of the road segments in our data set consist of 10 seconds or less. Excluding these road segments from model development increases our model R^2 to 0.20 (results not shown). This model however does not improve predictions to short-term stationary measurements (R^2 remains 0.36) and home outdoor measurements (R^2 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[11,14,51]. Other mobile campaigns include a Local Indicator of Spatial Analysis (LISA)[10], extend the averaging period[52] or disregard the issue[13]. 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[18] and in a study by Weichenthal et al[14].

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: General Information

Table A.1: Overview of different data sets.

Data set	Year	Location	Number	Resolution	Instruments
Mobile Measurements	2014/2015	Amsterdam, Maastricht and Utrecht	5236 Road Segments	~25seconds	CPC (UFP) and Aethalometer (BC)
	2013	Amsterdam and Rotterdam	2964 Road Segments	~20seconds	CPC (UFP) and Aethalometer (BC)
Short-Term Stationary Measurements	2014/2015	Amsterdam, Maastricht and Utrecht	240 Sites	3 times 30 minutes	CPC (UFP) and Aethalometer (BC)
Randomly selected addresses	/	Amsterdam, Maastricht and Utrecht	1500 Addresses	/	/
Home Outdoor Measurements	2014/2015	Amsterdam and Utrecht	42 Homes	3 times 24 hours	MiniDisc (UFP) and Harvard Impactors (BC)

Table A.2. Spatial predictor variables with units, a priori defined directions of effect and buffer sizes in the mobile and short-term stationary data sets.

Predictor variable	Units	Direction of effect	Buffer (m)	Mobile			Short-term Stationary		
				10th percentile	Mean	90th Percentile	10th percentile	Mean	90th Percentile
Industry area			**100**	0	1400	0	0	440	0
			300	0	13633	37122	0	7237	6463
	m^2	+	**500**	0	41895	166610	0	29522	110419
			1000	0	197751	711267	0	174742	619476
			5000	2305345	5591752	8349138	1931709	5353372	8062690
Port area			**100**	0	247	0	0	208	0
			300	0	2473	0	0	1786	0
	m^2	+	**500**	0	8142	0	0	7167	0
			1000	0	45285	0	0	52363	0
			5000	0	2428278	9225706	0	2116359	8617504
Airport area	m^2	+	**5000**	0	136650	0	0	25216	783
Urban green area			**100**	0	490	0	0	963	0
			300	0	7711	22125	0	10843	37904
	m^2	-	**500**	0	32633	132893	0	37932	152378
			1000	0	190110	579390	0	192426	551366
			5000	1372895	5233285	9714816	1122221	4590130	9281079
Natural and forested areas			**100**	0	222	0	0	167	0
			300	0	2103	0	0	1588	0
	m^2	-	**500**	0	7244	0	0	6077	0
			1000	0	52377	165299	0	51926	144940
			5000	1334268	4944915	8182331	1328510	5172157	8159990
Residential land area			**100**	0	26434	31375	0	25954	31375
			300	66208	226402	282618	47422	222740	282618
	m^2	+	**500**	231301	593328	785191	189582	579130	785191
			1000	842139	2082317	3050349	518003	1967973	3005396
			5000	15002050	28689475	46595685	11680371	27225930	46124341
Population density			**100**	7	245	524	15	270	561
			300	311	2117	4370	480	2096	4429
	n	+	**500**	1236	5469	11421	956	5208	10991
			1000	4805	19270	42504	2533	17452	39812
			5000	85535	251242	539468	71083	227169	531610
Household density			**100**	3	132	292	6	142	324
			300	144	1136	2476	200	1111	2553
	n	+	**500**	563	2940	6507	453	2773	6309
			1000	2079	10416	24315	1012	9380	22482
			5000	39485	134927	307588	32832	121927	303001
Traffic intensity on nearest road	$Veh.day^{-1}$	+		82	8656	25785	30	4090	14943
Traffic intensity on nearest major road	$Veh.day^{-1}$	+		5736	18232	36470	5649	18579	34240

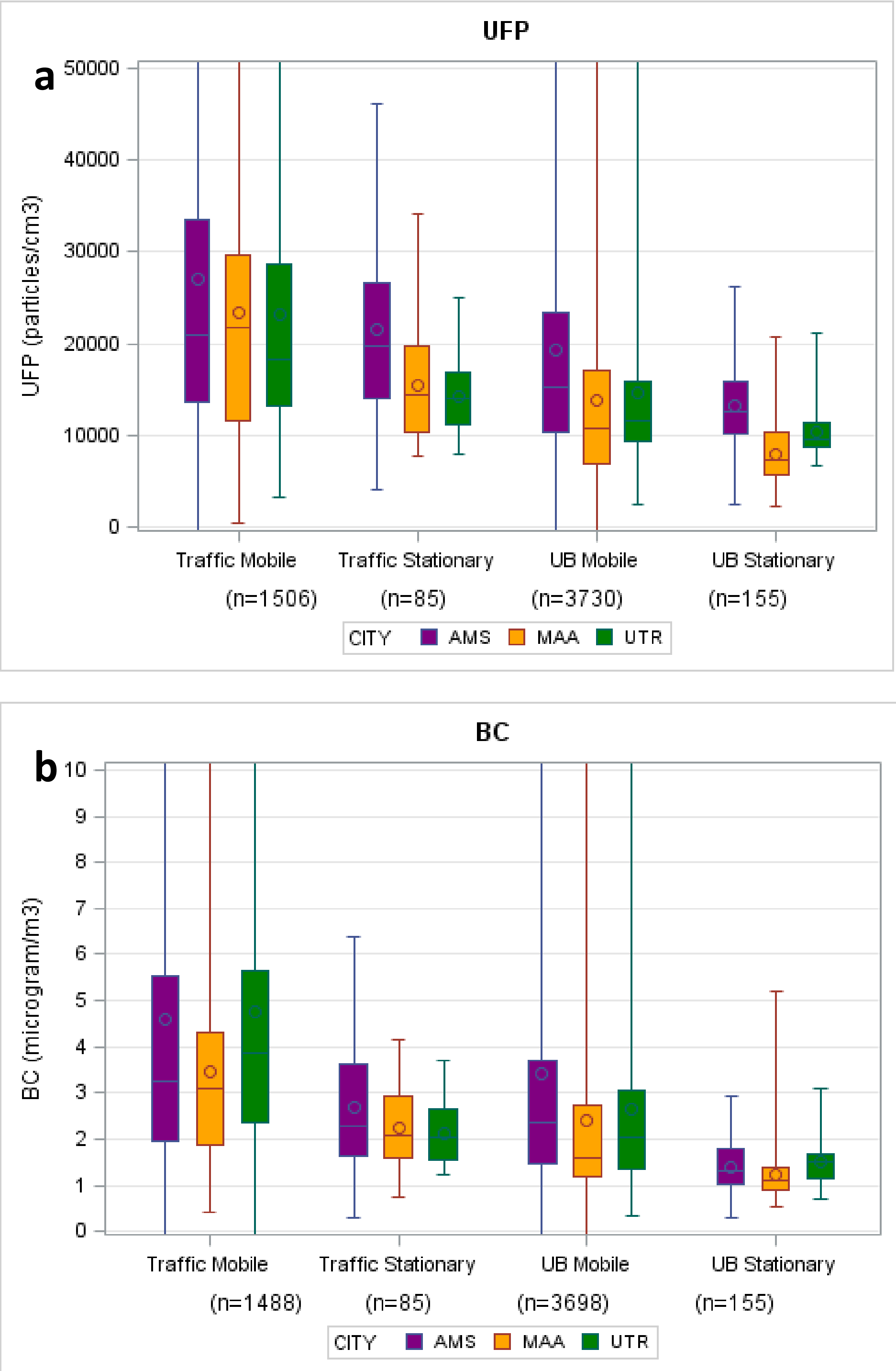
Heavy-duty traffic intensity on nearest road	$Veh.day^{-1}$	+		0	324	1005	0	125	420
Heavy-duty traffic intensity on nearest major road	$Veh.day^{-1}$	+		67	982	1950	48	1206	1769
Road length of all roads	m	+	**50**	102	258	404	98	190	308
			100	488	838	1200	363	716	1059
			300	3997	6359	8494	3180	5925	8209
			500	9994	16603	21857	7734	15605	21532
			1000	33179	60248	80412	27773	55926	78365
Road length of all major roads	m	+	**50**	0	79	203	0	42	174
			100	0	194	507	0	107	390
			300	0	1030	2259	0	664	1760
			500	0	2600	4951	0	2080	4164
			1000	3161	9707	15328	2165	8471	14235
Traffic intensity on all roads (sum of (traffic intensity * length of all segments))	$Veh.day^{-1}m$	+	**50**	40082	1257727	3400359	7963	757087	2207469
			100	185957	3417972	8691268	77072	2255473	5463763
			300	3136239	22142804	47797106	1315772	15681335	36067592
			500	11762571	58068646	120273812	5579975	46420718	98239045
			1000	63847576	226759175	452445437	31635299	196221848	417062321
Traffic intensity on all major roads (sum of (traffic intensity* length of all segments))	$Veh.day^{-1}m$	+	**50**	0	1127175	3336962	0	688087	2185933
			100	0	2998942	8304429	0	1942440	4963848
			300	0	18940815	44304127	0	12729584	31697479
			500	4808393	49735352	108364418	0	38751720	89534363
			1000	44458796	196518241	410822257	18085365	168852113	377180297
Heavy-duty traffic intensity on all roads (sum of (heavy-duty traffic intensity* length of all segments))	$Veh.day^{-1}m$	+	**50**	440	54021	141389	0	36679	92230
			100	3122	155742	378676	568	131698	213495
			300	72173	1121670	2637212	41345	760615	1541019
			500	328397	3078547	7798998	157767	2233684	5463944
			1000	2075563	13058286	26737180	1077166	10937816	23631745
Heavy-duty traffic intensity on major roads (sum of (heavy-duty traffic intensity*length of all segments)	$Veh.day^{-1}m$	+	**50**	0	48165	134636	0	33903	92230
			100	0	137648	357616	0	120977	210302
			300	0	988569	2470260	0	651430	1335757
			500	106797	2727455	7255375	0	1931699	5094488
			1000	1133803	11678977	25328256	624627	9707896	21738224
Inverse Distance to nearest road [a]	m^{-1}	+		na	na	na	0.073	0.488	0.756
Inverse Distance to nearest major road [a]	m^{-1}	+		na	na	na	0.002	0.065	0.135

[a] Variables were not used for mobile model development, due to values being zero.

Table A.3: Distribution of UFP and BC in mobile and short-term stationary data set.

Pollutant	Type of Measurements	No. of observations	Mean	10^{th} percentile	Median	90^{th} percentile
UFP (in particles /cm^3)	Mobile	5236	18623	6850	14013	35512
	Stationary	240	12910	6453	11062	20663
BC (in µg/m3)	Mobile	5186	3.32	1.07	2.29	6.40
	Stationary	240	1.73	0.85	1.50	2.94

Figure A.1: Distribution of UFP and BC in mobile and short-term stationary data set.



Appendix B: Ultrafine Particles

Table B.1. UFP Land-Use Regression Models based upon Mobile Measurements with and without AR-1 term and local exhaust plumes.

Variable	Original (With AR-1 term and with peaks)	With peaks, Without AR-1 term	With AR1- term, Without Peaks	Without AR-1 term and without peaks	Fixed City effect model (Without AR-1 term)
Intercept	8072 (968)	9002 (578)	8296 (596)	8603 (413)	Amsterdam 7858 (1060) Maastricht 8950 (658) Utrecht 8719 (641)
Population Density:					
Residential Land Area in a 5000m buffer	7763 (1155) [a]	5591 (703)	4493 (710)	4182 (504)	5955 (758)
Traffic:					
Traffic Intensity on the Nearest Road	2244 (756)	3727 (656)	2755 (504)	2876 (462)	3760 (656)
Heavy Traffic Intensity on the Nearest Road	989 (536)	1790 (499)	878 (381)	952 (355)	1754 (501)
Major Road Length in a 100m buffer	4588 (524)	5057 (465)	1727 (476)	2445 (473)	5095 (469)
Major Road Length in a 300m buffer			2069 (587)	1656 (533)	
Land Use:					
Port Area in a 5000m buffer	3457 (995)	3882 (586)	4195 (594)	4525 (415)	4599 (812)
Urban Green Land in a 500m buffer	-1001 (494)	-1018 (354)			-926 (361)
Urban Green Land in a 1000m buffer				-803 (306)	
Number Road Segments used for model development	5236	5236	5164	5164	5236
Model R^2	0.15 [b]	0.15	0.18 [b]	0.19	/
Pearson Correlation with Original Model on 1500 Random Addresses	/	0.99	0.97	0.98	/

[a] Regression slopes and standard error (between brackets), multiplied by the difference between 10^{th} and 90^{th} percentile for all predictors to allow comparison of the effect of predictors with different units and distribution on measured concentrations. Predictions in particles/cm^3.

[b] R^2 of model without AR-1 term.

Table B.2. UFP Land-Use Regression Models from the 2014-2015 campaign based upon Mobile Measurements.

Variable	Combined	Amsterdam
Intercept	8072 *(968)*	4053 *(3015)*
Population Density:		
Residential Land Area in a 5000m buffer	7763 *(1155)*[a]	8528 *(1873)*
Traffic:		
Traffic Intensity on the Nearest Road	2957 *(740)*	2817 *(1141)*
Heavy Traffic Intensity on the Nearest Road	989 *(536)*	
Heavy Traffic Intensity on the Nearest Major Road		1156 *(633)*
Traffic Intensity in a 100m buffer		
Major Road Length in a 100m buffer	4588 *(524)*	3100 *(1034)*
Land Use:		
Port Area in a 5000m buffer	3457 *(995)*	4911 *(1993)*
Urban Green Land in a 500m buffer	-1001 *(494)*	
Number Road Segments used for model development	5,236	1,991
R^2 of model compared to short-term stationary measurements	0.36	0.21

[a] *Regression slopes and standard error (between brackets), multiplied by the difference between 10th and 90th percentile for all predictors to allow comparison of the effect of predictors with different units and distribution on measured concentrations. Predictions in particles/cm^3.*

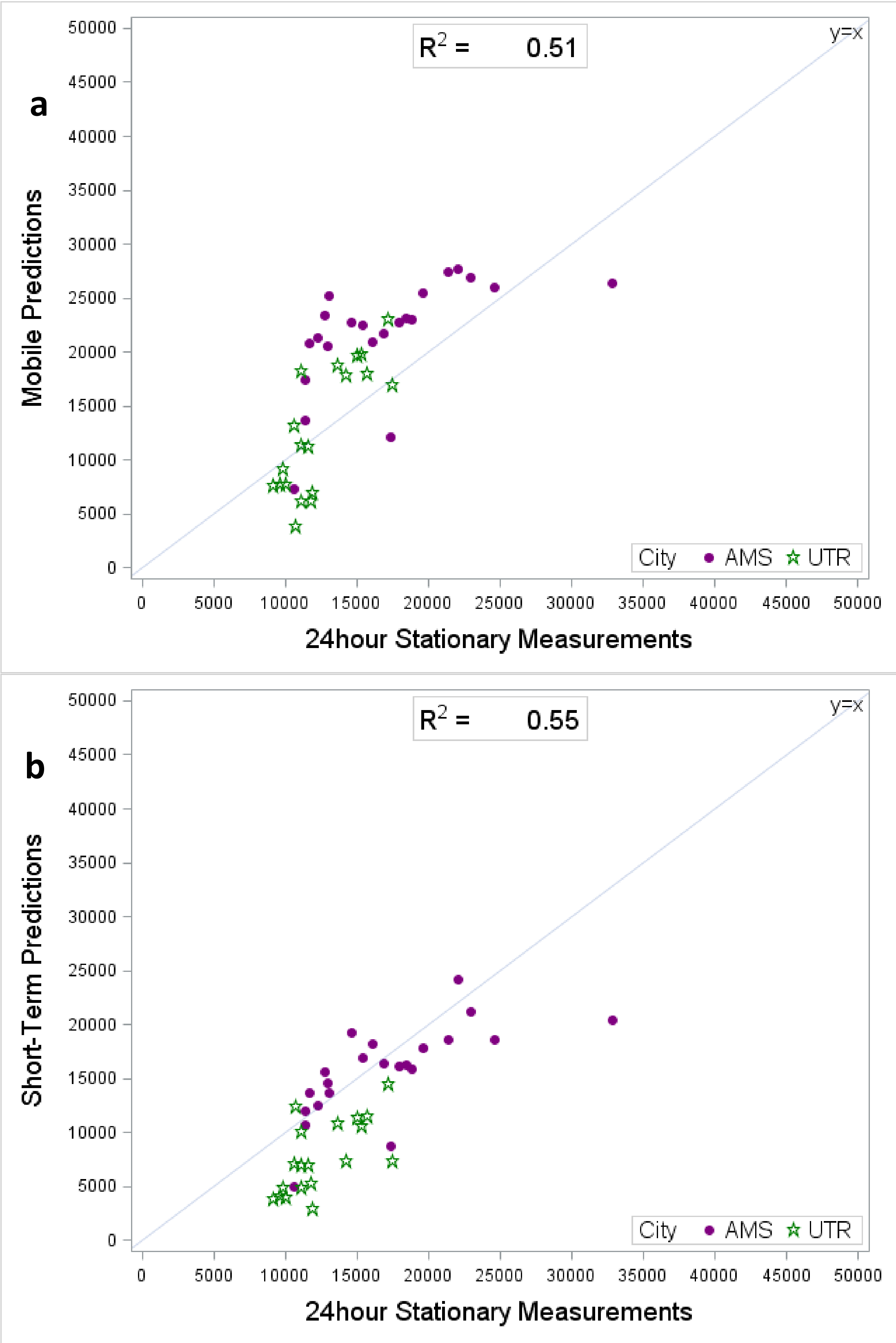
Table B.3. UFP Land-Use Regression Models from the 2013 campaign based upon Mobile Measurements.

Variable	Combined	Amsterdam
Intercept	5656 (2675)	-1254 (2974)
Population Density:		
Population density in a 5000m buffer	8064 (1947)[a]	8323 (2865)
Traffic:		
Traffic Intensity on Major Roads in a 100m buffer	1928 (1095)	5722 (1641)
Traffic Intensity in a 500m buffer	2917 (1514)	
Traffic Intensity in a 1000m buffer		7694 (2919)
Major Road Length in a 50m buffer	6868 (1071)	3884 (1567)
Land Use:		
Port Area in a 500m buffer		2102 (633)
Port Area in a 1000m buffer	2499 (1248)	
Airport Area in a 5000m buffer	4669 (1185)	
Natural Land in a 5000m buffer	-2557 (1357)	
Number Road Segments used for model development	2964	1427
R^2 of model	0.13	0.18

[a] *Regression slopes and standard error (between brackets), multiplied by the difference between 10^{th} and 90^{th} percentile for all predictors to allow comparison of the effect of predictors with different units and distribution on measured concentrations. Predictions in particles/cm^3.*

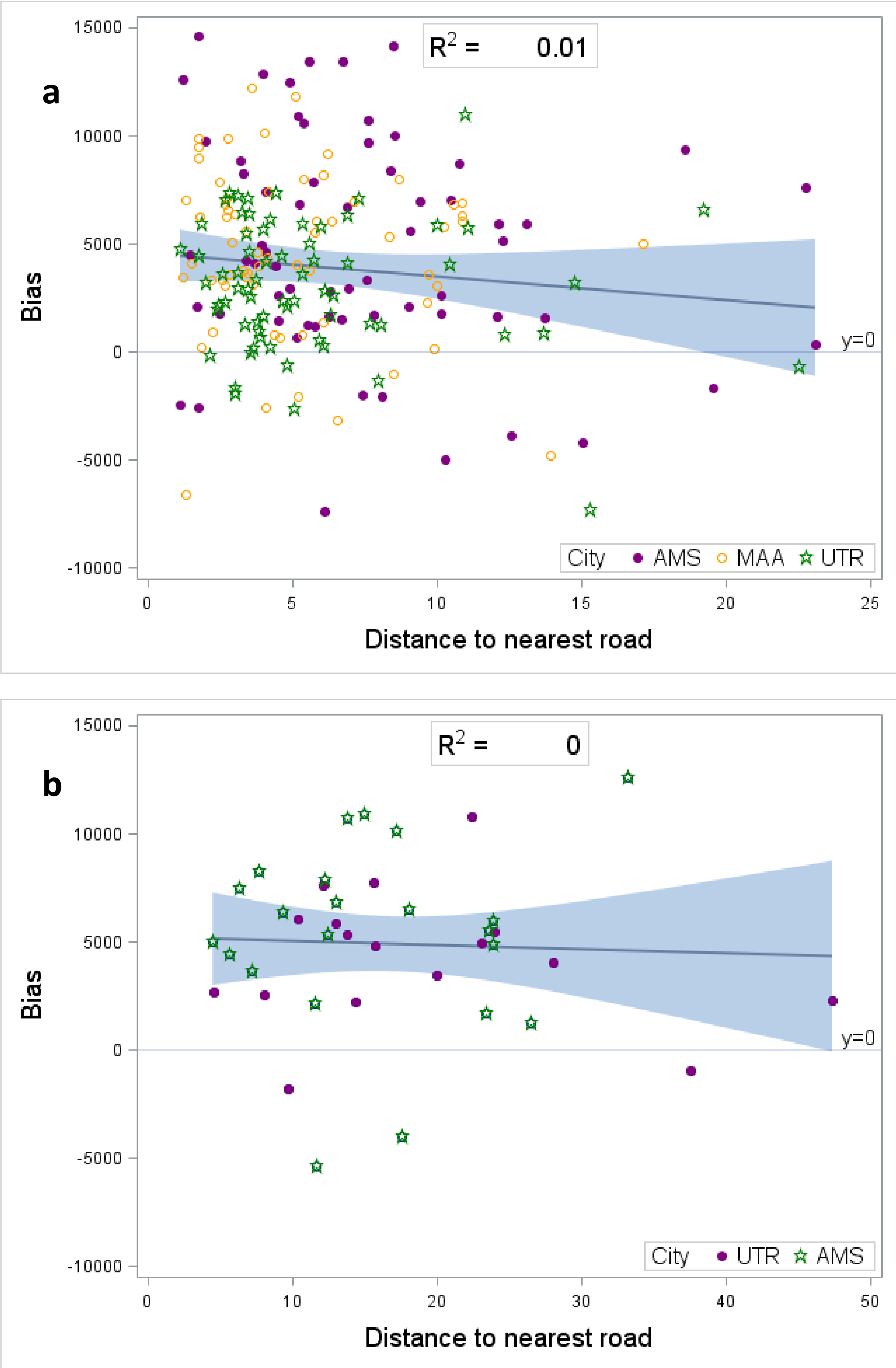
[b] *R^2 of short-term stationary model is between brackets.*

Figure B.1. Predicted concentration levels at home outdoor sites (n=42) based on mobile UFP 2013 model (a) and short-term stationary UFP 2013 model (b) compared to 3 x 24h measurements from 2014/2015.



UFP levels in particles/cm^3

Figure B.2. Bias of predicted UFP counts on the short-term stationary sites (a) and home outdoor 24h sites (b) vs. distance of the measurement site to the nearest road.



Bias UFP levels in particles/cm³

Figure B.3. Mobile UFP predictions compared to short-term stationary UFP predictions on 1500 random addresses, with Bland Altman plots and for both measurement campaigns.

2013 Campaign:

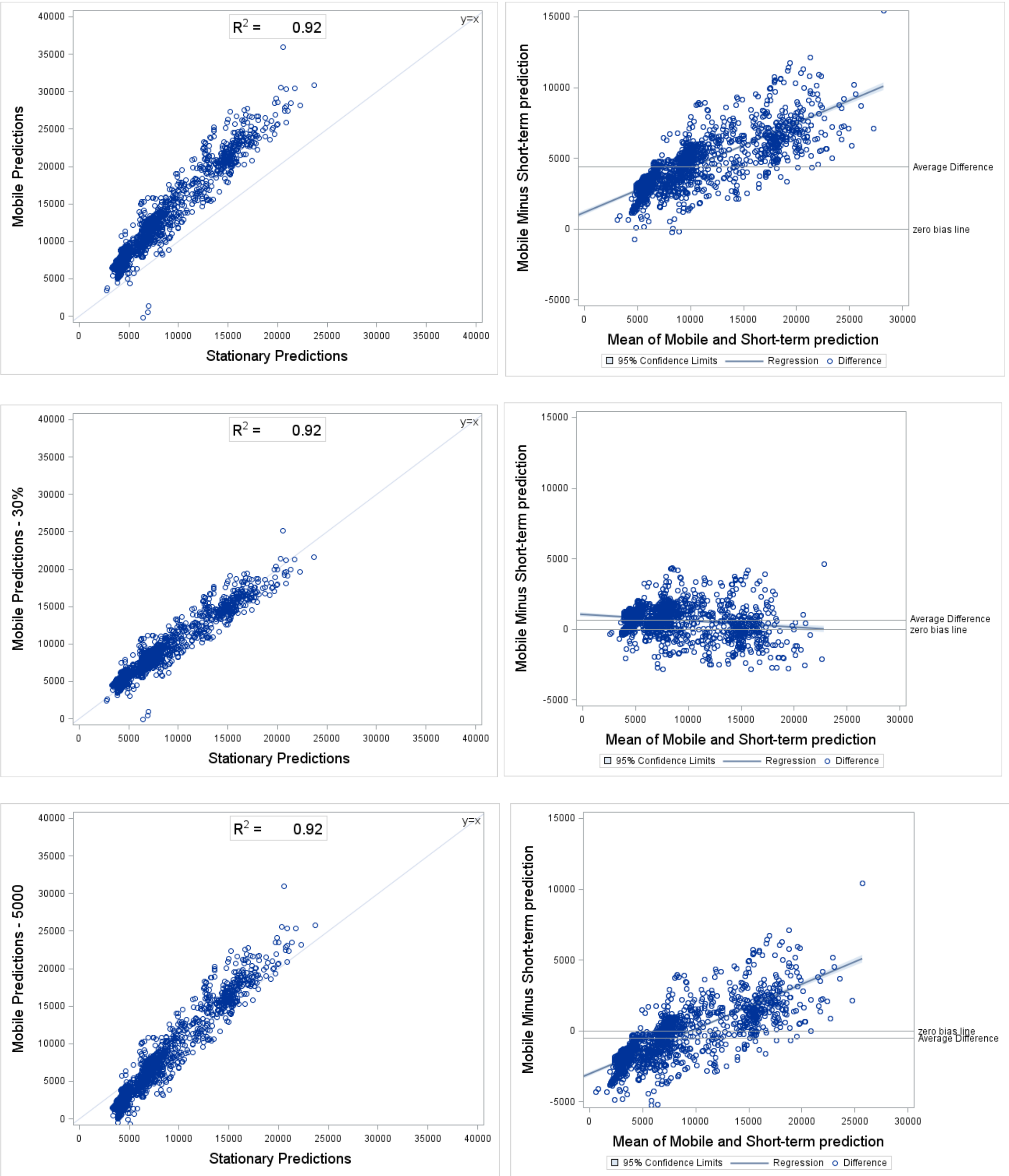


Figure B.3. Continued.

2014/2015 Campaign:

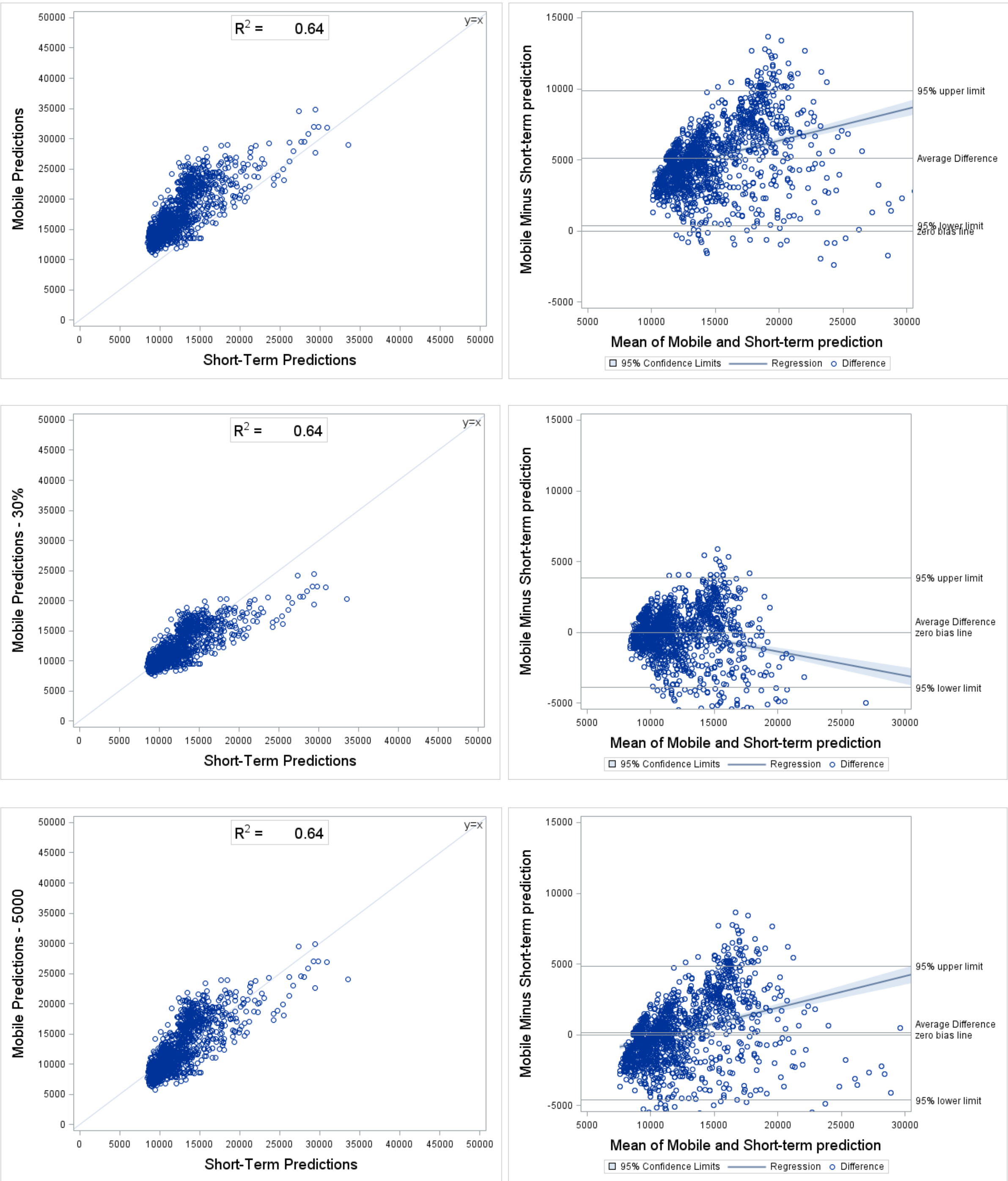
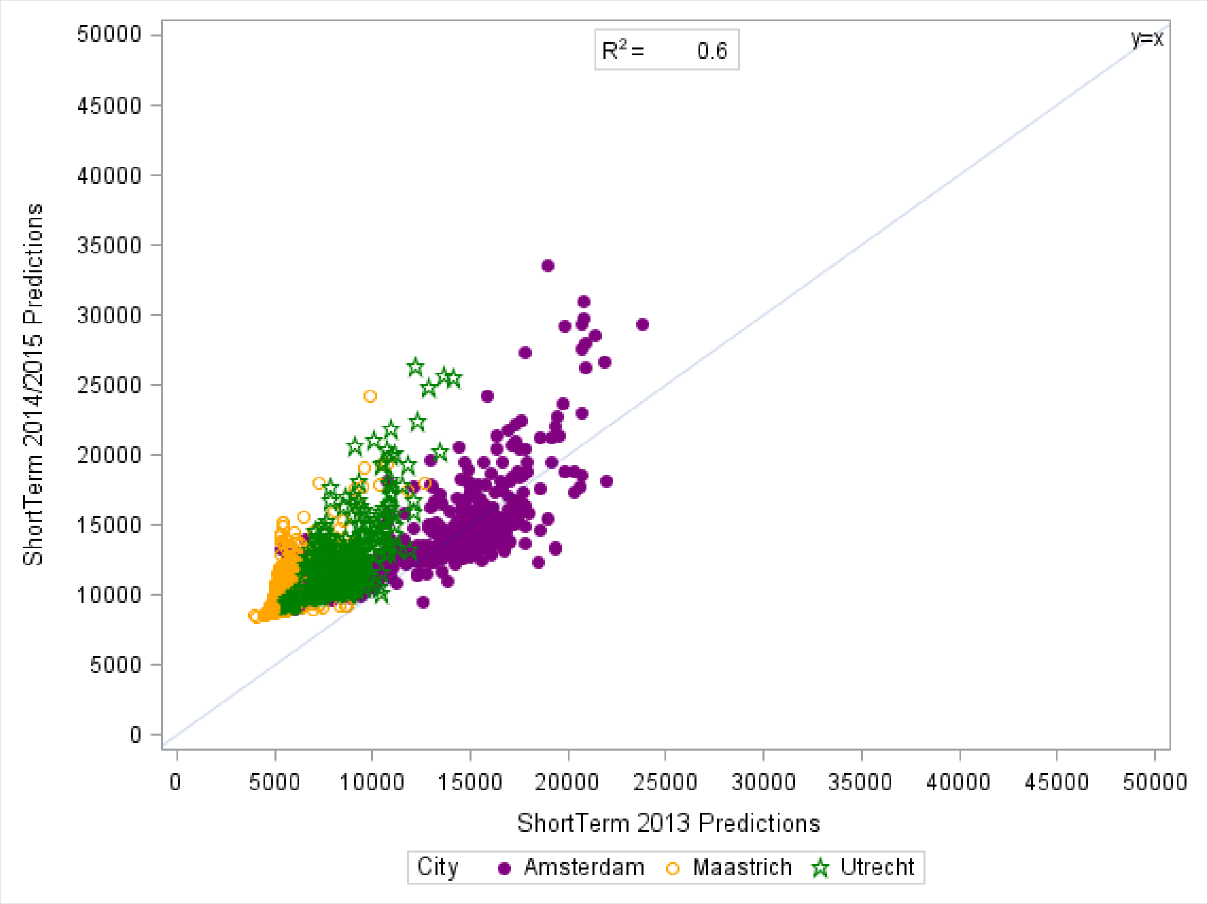
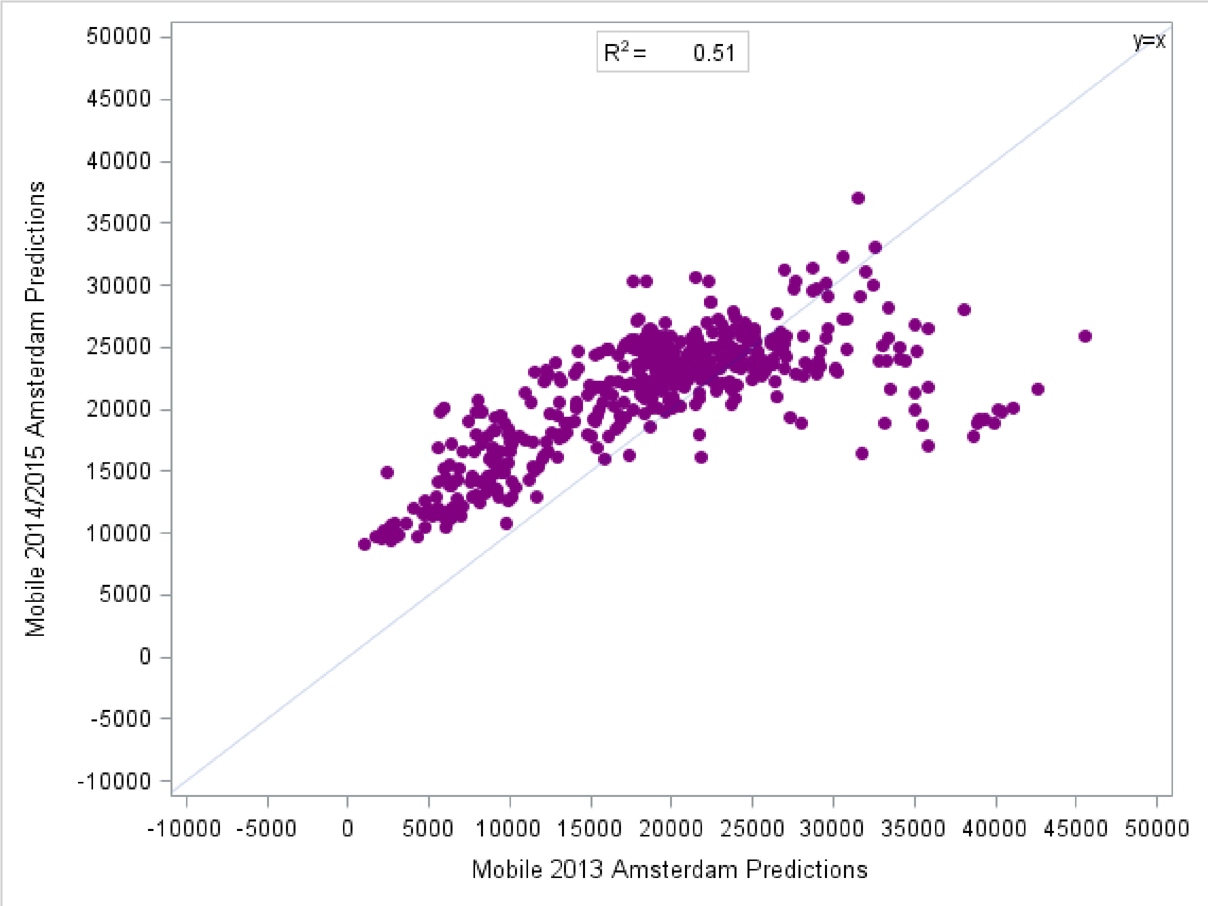


Figure B.4. Short-Term Stationary UFP predictions based on measurements from the 2013 campaign versus Short-Term Stationary predictions based on the measurements in 2014/2015.



Predictions in particles/cm³.

Figure B.5. Comparison of predicted UFP counts based on mobile LUR models in 2013 and 2014/2015 in Amsterdam.



Predictions in particles/cm^3.

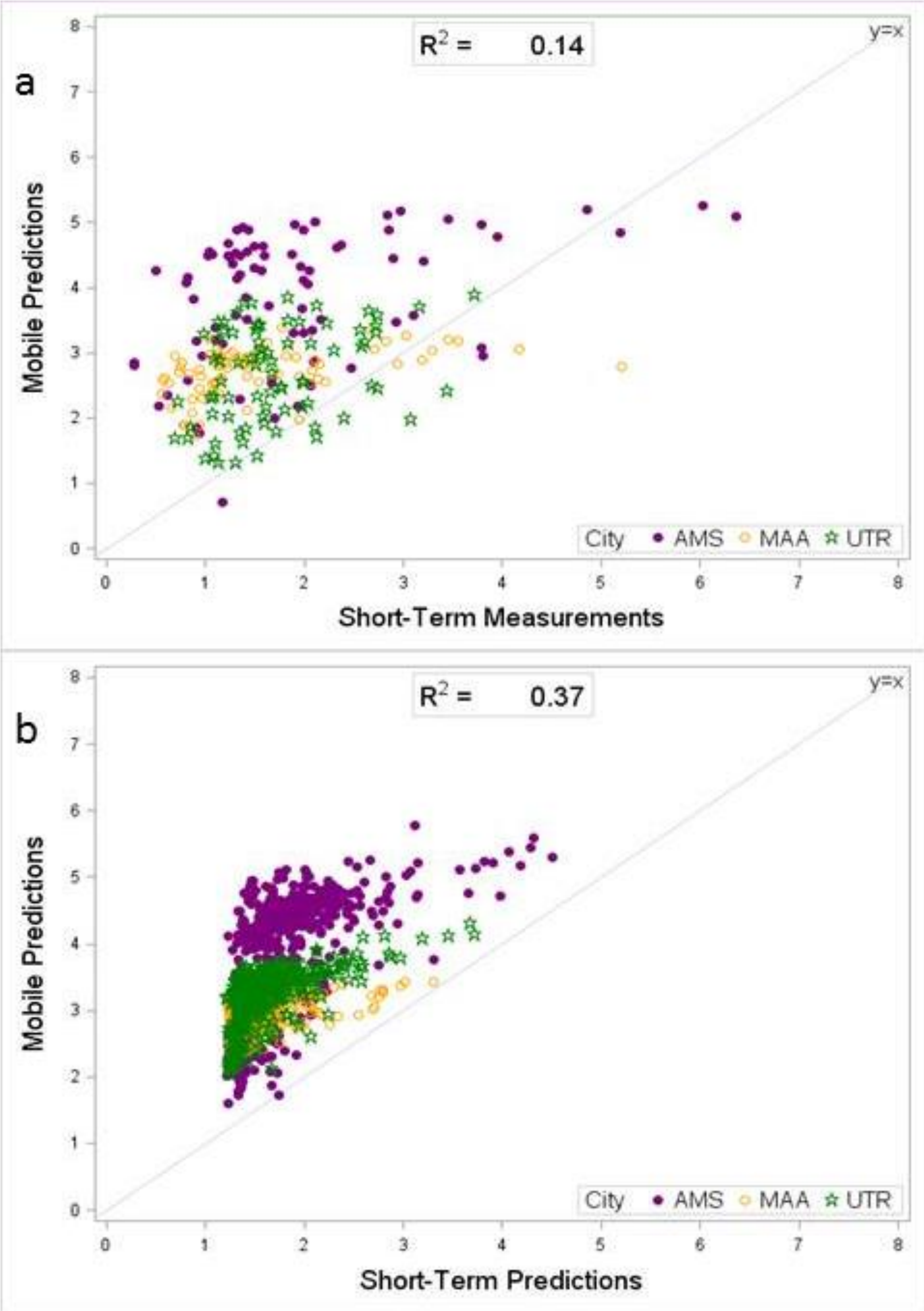
Appendix C: Black Carbon

Table C.1. Mobile and Short-Term Stationary BC Models.

	BC (in $\mu g/m^3$)	
Variable	**Short-Term**	**Mobile AR-1**
Intercept	1.20 *(0.07)*	1.00 *(0.28)*
Population Density:		
Household density in a 1000m buffer	0.33 *(0.14)*	
Residential land area in a 5000m buffer		2.43 *(0.29)*
Traffic:		
Traffic intensity on the nearest road	0.29 *(0.12)*	0.36 *(0.10)*
Traffic intensity in a 50m buffer	0.63 *(0.12)*	
Length of major roads in a 100m buffer	0.37 *(0.13)*	0.27 *(0.10)*
Land Use:		
Urban green in a 1000m buffer		-0.35 *(0.16)*
R^2 of model	0.44	0.10 [b]
Number sites used for model development	240	5,169

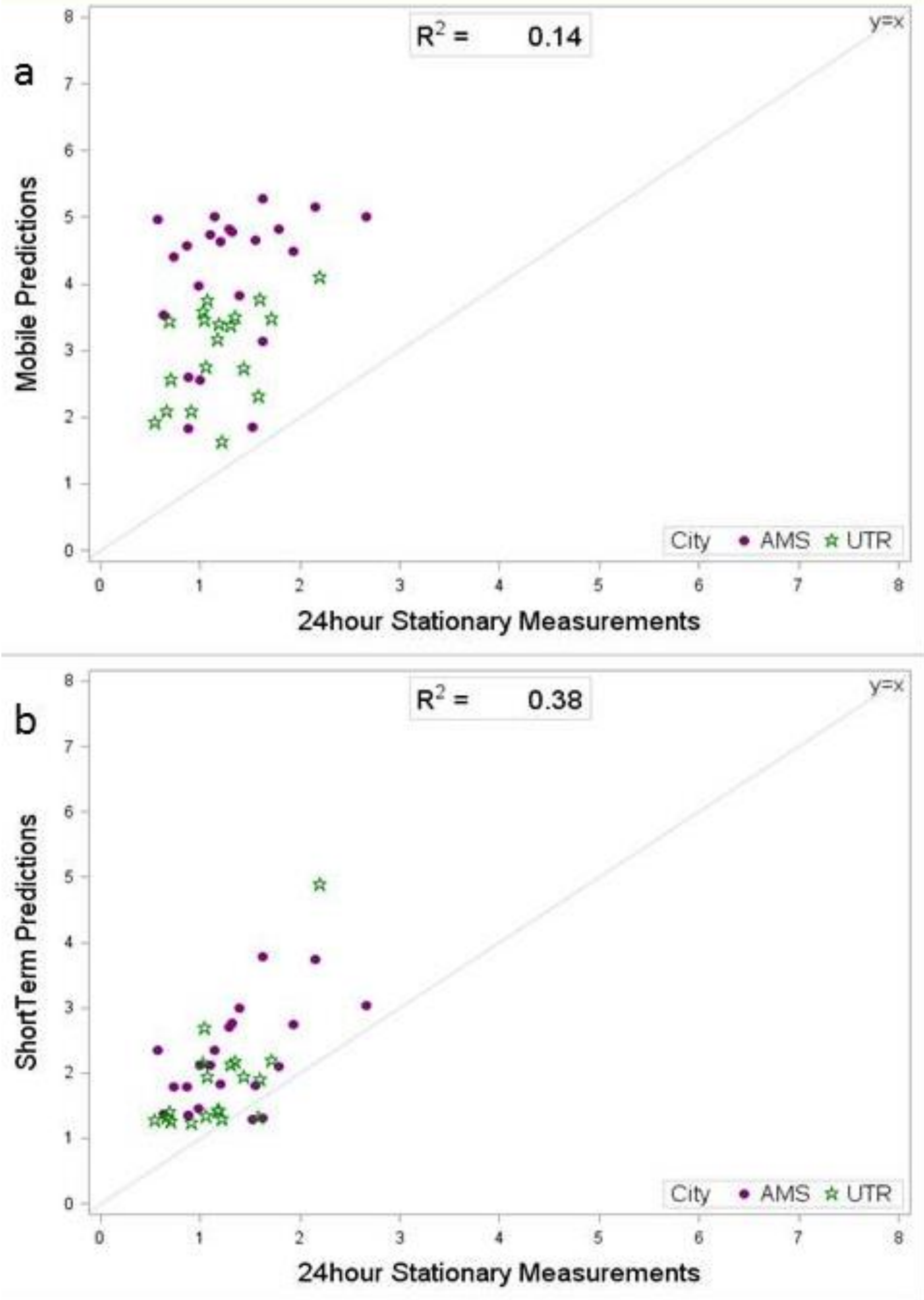
[a] Regression slopes and standard error (between brackets), multiplied by the difference between 10th and 90th percentile for all predictors. [b] R^2 of model without AR-1 term.

Figure C.1. (a) Predicted concentration levels 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 1,500 random addresses in Amsterdam, Utrecht and Maastricht.



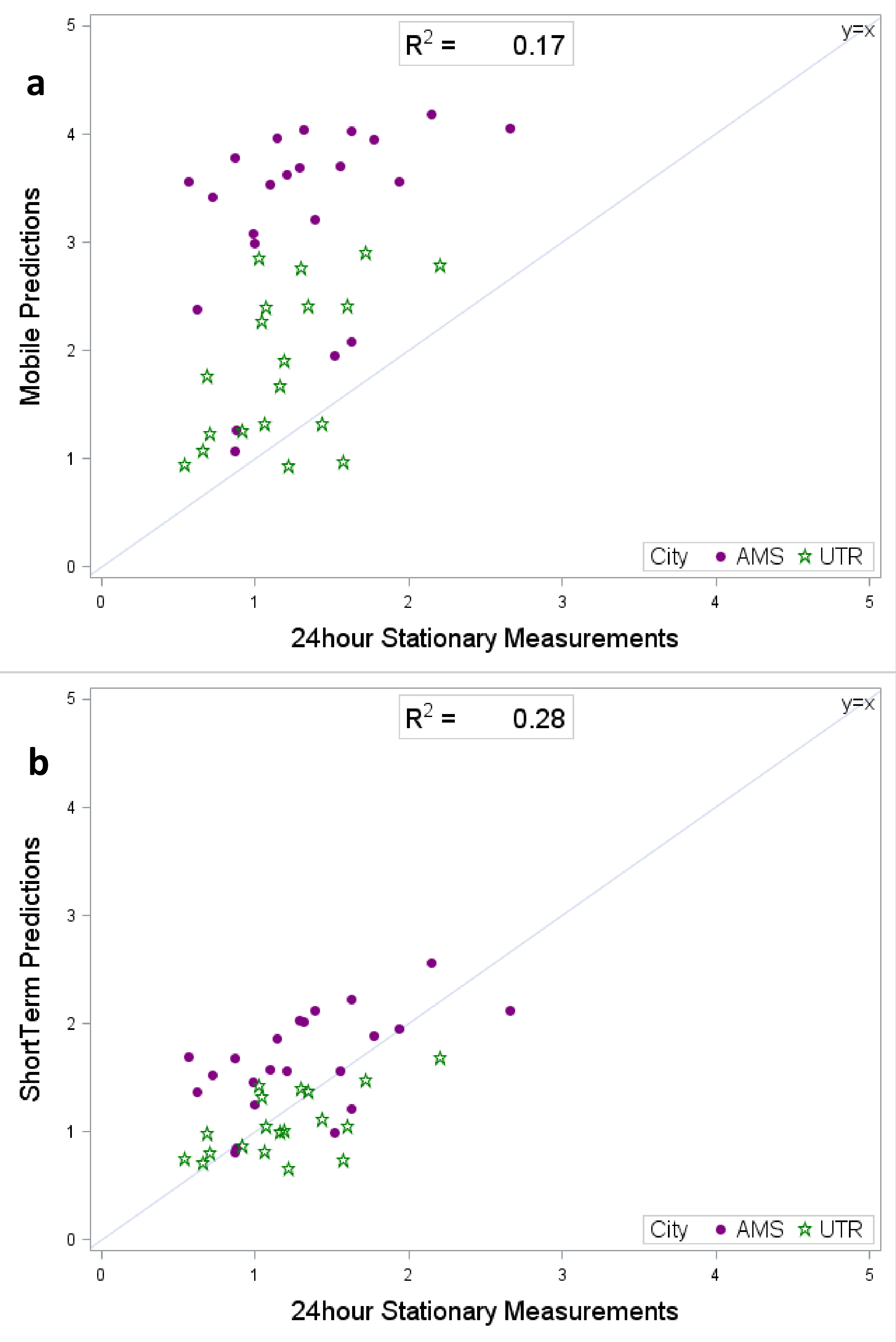
BC predicted levels in µg/m^3.

Figure C.2. Predicted concentration levels at home outdoor sites (n=42) based on mobile models (a) and short-term stationary models (b) compared to 3 x 24h measurements.



BC predicted levels in µg/m³.

Figure C.3. Predicted concentration levels at home outdoor sites (n=42) based on mobile BC 2013 model (a) and short-term stationary BC 2013 model (b) compared to 3 x 24h measurements from 2014/2015.



BC predicted levels in µg/m^3.