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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 × 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 × 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 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 (Kerckhoffs et al., 2016; Montagne et al., 2015; Klompmaker et al., 2015). Data from the EXPOsOMICS 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 $PM_{2,5}$ 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. (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). $PM_{2,5}$ 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^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 (Kerckhoffs et al., 2016; Montagne et al., 2015; Klompmaker 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