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Intra-urban variation of ultrafine particles as evaluated by process related land use and pollutant driven regression modelling

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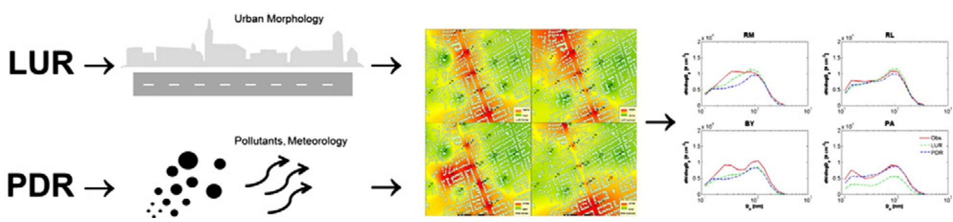
HIGHLIGHTS

- Two regression type models for urban ultrafine particle concentrations are compared.
- Land use regression model is driven by urban morphological parameters exclusively.
- Other regression type model uses pollutant and meteorological input parameters.
- Both models resolve spatial differences of ultrafine particle number concentrations.
- Both models adequately predict particle number size distributions <100 nm.

GRAPHICAL ABSTRACT



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ABSTRACT

The microscale intra-urban variation of ultrafine particle concentrations (UFP, diameter $D_p < 100$ nm) and particle number size distributions was studied by two statistical regression approaches. The models were applied to a 1 km^2 study area in Braunschweig, Germany. A land use regression model (LUR) using different urban morphology parameters as input is compared to a multiple regression type model driven by pollutant and meteorological parameters (PDR). While the LUR model was trained with UFP concentration the PDR model was trained with measured particle number size distribution data. The UFP concentration was then calculated from the modelled size distributions. Both statistical approaches include explanatory variables that try to address the 'process chain' of particle emission, dilution and deposition.

LUR explained 74% and 85% of the variance of UFP for the full data set with a root mean square error (RMSE) of 668 cm^{-3} and 1639 cm^{-3} in summer and winter, respectively. PDR explained 56% and 74% of the variance with RMSE of 4066 cm^{-3} and 6030 cm^{-3} in summer and winter, respectively. Both models are capable to depict the spatial variation of UFP across the study area and in different outdoor microenvironments. The deviation from measured UFP concentrations is smaller in the LUR model than in PDR.

The PDR model is well suited to predict urban particle number size distributions from the explanatory variables (total particle number concentration, black carbon and wind speed). The urban morphology parameters in the LUR model are able to resolve size dependent concentration variations but not as adequately as PDR.

1. Introduction

Particulate air pollution in urban areas is associated with significant impacts on human health (e.g. Brook et al., 2010; Heal et al., 2012;

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WHO, 2013; HEI, 2013). Recent epidemiological and toxicological research indicates that the number of ultrafine particles (UFP, $D_p < 100$ nm) is one important metric to assess the health effects of urban particulate air pollution. UFP are emitted as primary particles mainly from combustion processes (traffic, industry) or they form as secondary particles from different aerosol physical and chemical processes (e.g. condensation from hot traffic tailpipe emissions, nucleation from precursor gases, cf. Giechaskiel et al., 2005; von Bismarck-Osten et al., 2013). The UFP concentration can be estimated from measurements of the particle number size distribution. At street canyon or near-traffic sites the number of UFP generally accounts for the majority of total particle number concentration, i.e. >80% to 90% (Morawska et al., 2008; Weber et al., 2013). Since high traffic intensity is considered as an essential source for UFP and fine particles in general, the proximity of the residence to roads could be identified as a major determinant of health effects caused by particulate air pollution (Hoffmann et al., 2006, 2007; Jerrett, 2011). Thus, accurate assessment of exposure to traffic-related air pollution is important for epidemiological studies as well as for the estimation of health impacts of current and future road networks (e.g. Gilbert et al., 2005).

Despite the abundance of particles in urban environments the concentrations of UFP are characterised by considerable intra-urban variability, such as decrease of UFP with growing distance to roads (Weber, 2009; Krudysz et al., 2009). Hence, urban morphology and complexity (i.e. the road network with different traffic intensity on road sections, the three-dimensional structure of buildings, which influences climate modifications and dispersion) can lead to specific spatial distribution patterns of urban UFP concentrations.

Statistical aerosol models have been used recently to calculate total particle number concentrations in urban environments (e.g. Weber et al., 2013) or to study specific size ranges of the number size spectrum (Clifford et al., 2011; Mølgaard et al., 2012, 2013; von Bismarck-Osten et al., 2015). The types of statistical models that we define as pollutant and meteorological parameter driven model (PDR) are built from multiple linear/non-linear regression approaches using different sets of explanatory variables, such as gaseous pollutant concentrations (e.g. NOx), meteorological quantities (wind speed, temperature, humidity, solar radiation), or traffic-related variables to predict particle concentrations (Clifford et al., 2011; Mølgaard et al., 2012; Weber et al., 2013).

Land Use Regression (LUR) models were developed as an alternative to dispersion models. They can be applied to predict local variation in traffic pollution and to obtain urban scale air pollutant concentrations without a detailed pollutant emission inventory (Briggs et al., 2000; Brauer et al., 2003, Beelen et al., 2013). LUR models are multiple linear regression approaches that assume independent residuals and use GIS-based explanatory variables to predict pollutant concentrations at certain locations (Hoek et al., 2008; Mercer et al., 2011). They have been widely applied in cities of North America, Europe, and Asia (e.g. Arain et al., 2007; Chen et al., 2010; Saraswat et al., 2013; Tang et al., 2013; Rivera et al., 2012; Abernethy et al., 2013). As one example, Henderson et al. (2007) developed an LUR model for UFP in the city of Amsterdam including traffic intensity on nearest road, distance to roads, population density and land use. However, only few studies included 3D data of buildings and street canyons into LUR modelling (Tang et al., 2013; Ghassoun et al., 2015). Eeftens et al. (2013) described an approach to include the effects of urban canyon geometry effects on NO_2 and NO_x concentrations using 3D building data and evaluated the improvement on LUR. Ho et al. (2015) developed LUR models for vertical distributions of $PM_{2.5}$ elemental composition and examined the influence of different heights from ground level on the predicted values of these pollutants.

Our hypothesis is that urban morphology parameters are capable to not only explain spatial distributions of urban UFP concentrations but also intra-urban variations in particle number size distributions. To the authors knowledge this is the first study using an LUR approach to predict particle number size distributions on the urban microscale, i.e. 10^1–10^3 m. Knowledge of the particle number size distribution offers the chance to deduce different particle exposure metrics from this data, i.e. concentrations in certain size ranges. The present LUR model conceptually tries to depict particle concentrations in urban microenvironments as the result of the particle 'process chain' composed of emission, dilution/dispersion and deposition. All these processes take place under certain boundary conditions, which are variable due to anthropogenic behaviour, weather conditions and surrounding built environment. Therefore the models are implemented with respective input parameters that are related to these three processes.

The present study establishes and compares two different types of statistical models which are capable of estimating urban UFP concentrations and particle number size distributions on the urban microscale. A PDR model was used to calculate the full particle number size distribution in a size range between $10 < D_p < 420$ nm (cf. Ruths et al., 2014). The UFP concentration was then calculated from the number size spectrum. An LUR is developed to estimate UFP concentrations at given locations but will also be tested for its capability to calculate the particle number size distribution.

The specific difference between both model approaches (LUR and PDR) is that the LUR model predicts UFP concentrations by using urban morphological parameters exclusively. Those parameters can be obtained from 2D GIS datasets like open street map (OSM), official 2D data sources or from 3D city models. In contrast, the PDR model is established using meteorological and pollutant measurement data as input.

2. Material and methods

2.1. Study area

The spatial variation of UFP is studied in the city of Braunschweig, which is located in the south-eastern part of the federal state Lower-Saxony, Germany. The 1 km^2 study area is characterised by roads with low traffic intensities (RL, <10,000 Vehicles d^{-1}), medium (RM, >10,000 Vehicles d^{-1}) and with high traffic intensities (RH, >30,000 Vehicles d^{-1}), residential areas (RE, <2000 Vehicles d^{-1}), backyards (BY) and urban park areas (PA). The area was divided into 31 cells representing different 'outdoor microenvironments' (OME, Fig. 1). OME are defined as microscale areas with quasi-homogeneous concentrations which can be used to derive characteristic concentrations (e.g. Ott et al., 2007). As a compromise between a good spatial resolution of the pollutant sampling points while keeping the measurement time of a mobile measurement tour as short as possible (to minimize trends of particle concentration on the diurnal course) the amount of sampling spots was reduced to 27 (Fig. 1). However, at least one spot per OME was measured.

2.2. Measurements and instrumentation

Mobile monitoring was conducted with a platform consisting of a bicycle connected to a 2-wheel trailer which contained the instruments in a weather protected box. The measurements were performed along a pre-specified route stopping at the same sampling spots on each measurement day to conduct a 3 min measurement. At each site the average concentration of total particle number concentration, the number size distribution and black carbon were measured. The measurements were carried out during 8 campaigns in winter (January–March 2013), and 7 campaigns in summer (June–August 2013) resulting in a total of 15 replicates at each sampling spot. Further details are reported in Ruths et al. (2014).

Particle number size distributions were measured with a mobile scanning mobility particle sizer (NanoScan SMPS, TSI 3910, TSI Inc.), which offers a size distribution in the range $10 < D_p < 420$ nm with 1 min time resolution. The NanoScan reports size resolved particle number concentrations in 13 size bins with midpoint diameters of 11.5, 15.4, 20.5, 27.4, 36.5, 48.7, 64.9, 86.6, 115.5, 154, 205.4, 273.8 and 365.2 nm.