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Statistical modelling of aerosol particle number size distributions in urban and rural environments – A multi-site study

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ABSTRACT

A multiple linear regression approach was used to model the urban atmospheric particle number size distribution (NSD) at 11 different sites (roadside, urban background, rural) in Central European cities for the time period 2008–2010. The same set of 13 model input parameters, consisting of temporal information (daytime, season) and meteorological measurement data, was used at each site.

NSD model performance indicates an average deviation between observations and model (Bias) in the order of <10% with respect to total particle number concentration. The most reliable predictions were achieved for roadside sites with correlation coefficients (R) of 0.75 on average and a normalized root mean square error ($RMSE_n$) of 0.79. Limited performance was observed for rural sites ($R = 0.61$; $RMSE_n \sim 1.2$). The transferability of the model approach to an independent urban background site was tested showing R of 0.5 and normalized $RMSE_n$ of 1.4. Although the physical relationship between particle NSD, ambient meteorological conditions and the temporal parameters are extremely complex, the model was able to reproduce the variation in particle concentrations. As a first in

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the field this study focused on modelling the entire number size distribution in contrast to size integrated particle number concentrations.

1. Introduction

A detailed knowledge of atmospheric particle formation processes, physico-chemical transformation and boundary layer dispersion of particles is of high scientific and public interest due to the risks of fine and ultrafine particles to human health, especially in urban agglomerations with high concentrations of airborne particles. This requires detailed quantification of different aerosol characteristics and metrics, such as particle emission rates (e.g. from traffic, industrial processes or domestic heating), relevant physical and chemical transformation processes and particle dispersion. While information on mass-based particulate matter metrics is abundant (Putaud et al., 2010), studies on particle metrics related to particle number concentration is more scarce. In cities, ambient particle number concentrations are greatly influenced by the sources and transformation processes of ultrafine particles (Wegner et al., 2012; Kumar et al., 2014), which are more difficult to capture and describe as a result of the shorter life-time of these particles.

Meteorological parameters directly or indirectly influence some of these processes. A number of studies for example focused on the relationship between particle concentrations and wind speed (Hussein et al., 2006; Agus et al., 2007), the behaviour of aerosols at different ambient temperatures (Olivares et al., 2007; Sabaliauskas et al., 2012), cloud formation processes induced by cloud condensation nuclei (Jones et al., 2007) or scavenging effects during precipitation events (Mircea et al., 2000). Other recent studies addressed physical and chemical processes such as gas-to-particle conversion (e.g. Ketzel and Berkowicz, 2004; Pöschl, 2005; Kulmala, 2005; Kumar et al., 2011).

The exposure of humans towards particles is of particular interest since particles, especially ultrafine particles (UFP) with diameters (D_p) < 100 nm, are associated to different health endpoints, i.e. cardiovascular and respiratory diseases (e.g. Schikowski et al., 2007; Hirano, 2009; Strak et al., 2010; Araujo, 2011). Despite a growing number of particle number concentration and size distribution measurements in urban areas around the world, there is still limited knowledge about the processes influencing the temporal variation of urban concentration levels on different spatial scales, i.e. the concentration variability between and across cities (e.g. Krudysz et al., 2009; von Bismarck-Osten et al., 2013).

The aerosol particle number size distribution (NSD) is an important metric since it provides a size dependent number concentration of aerosols that can be converted to other (exposure) metrics of interest such as integral or size interval-based particle number concentration, but also particle surface area or volume concentration. Although the measurement of the particle NSD requires additional instrumental efforts, continuous measurements have now become a routine matter in certain observation networks, for example ACTRIS (Aerosols, Clouds, and Trace gases Research InfraStructure Network), the UK Particle Numbers and Concentrations Network, and the German Ultrafine Aerosol Network (Birmili et al., 2009). Such efforts have become possible since the development and implementation of quality assurance measures for mobility particle size spectrometers (Wiedensohler et al., 2012; Schladitz et al., 2014).

The need to provide spatially and temporally complete exposure data highlights the importance of developing modelling approaches, which allow for an estimation of the NSD for specific regions, cities or site types (e.g. traffic vs. non-traffic exposed sites) by deriving characteristic patterns of particle concentrations and size distributions. This could contribute to improved human exposure assessment for particles and could facilitate the implementation of emission reduction targets for specific regions. Recently some studies focused on the prediction of particle number concentrations within the fine and ultrafine size spectrum (cf. Table 1 for an overview). These include both statistical forecasting approaches for certain size fractions of the aerosol spectrum (e.g. Mikkonen et al., 2011; Mølgaard