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Lung deposited surface area in Leicester urban background site/UK: Sources and contribution of new particle formation

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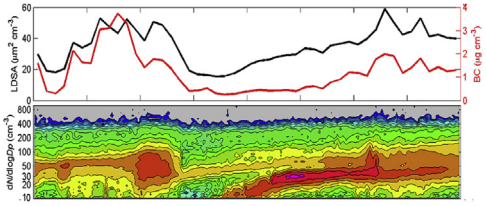
HIGHLIGHTS

- Clear seasonal variation of Lung Deposited Surface Area (LDSA) concentrations is observed with higher values in winter.
- Calculated LDSA seems to be a good surrogate to equivalent black carbon mass concentration.
- Traffic emissions appear to be the main source of LDSA in Leicester.
- LDSA concentrations are nearly doubled during new particle formation episodes.

GRAPHICAL ABSTRACT



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ABSTRACT

Lung Deposited Surface Area (LDSA) has been identified as a potential metric for the correlation of a physical aerosol particle properties with health outcomes. Currently, there is little urban LDSA data. As a case study, we investigated measurements of LDSA (alveolar) concentrations in a mid-size European city. LDSA and associated measurements were carried out over 1.5 years at an urban background site in Leicester, UK. Average LDSA concentrations in the cold (November–April) and warm (May–October) seasons of UK were 37 and 23 $\mu m^2\ cm^{-3}$, respectively. LDSA correlates well ($R^2 = 0.65–0.7$, $r = 0.77–0.8$) with traffic related pollutants, such as equivalent black carbon (eBC) and NO_X. We also report for the first time in the UK the correlation between an empirically derived LDSA and eBC. Furthermore, the effect of wind speed and direction on the LDSA was explored. Higher LDSA concentrations are observed at low wind speeds (1–2 m s^{-1}), owing to local traffic emissions. In addition, the diurnal variation of LDSA showed a second peak in the afternoon under warm and relatively clean atmospheric conditions, which can be attributed to photochemical new particle formation (NPF) and growth into the Aitken mode range. These NPF events increased the average background LDSA concentrations from 15.5 to 35.5 $\mu m^2\ cm^{-3}$, although they might not be health-relevant. Overall, the results support the notion that local traffic emissions are a major contributor to observed LDSA concentrations with a clear seasonal pattern with higher values during winter.

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1. Introduction

The surface area concentration is an important property of atmospheric aerosol particles which links aerosol loading to its health effects. Over recent years a number of epidemiological studies have shown that atmospheric particle surface area concentration may have a stronger correlation with negative health effects than particle mass or number concentration (Brown et al., 2000, 2001; LeBlanc et al., 2010; Nel et al., 2006; Nurkiewicz et al., 2009; Nygaard et al., 2004; Oberdörster et al., 2005; Sager and Castranova, 2009; Singh et al., 2007; Stoeger et al., 2006; Tran et al., 2000). Toxicological studies have found that ultrafine particles may have an increased toxicity compared to larger particles with the same composition (Johnston et al., 2000; Karlsson et al., 2009), and that the surface area concentration might be the most relevant physical measurement of ultrafine particle exposure (Maynard and Maynard, 2002; Moshammer and Neuberger, 2003; Oberdörster, 2000). All these studies reveal that the surface area concentration of atmospheric particles in various environments is a suitable property to represent the negative human health effects of aerosol exposure.

Instruments based on unipolar diffusion charging of particles or lung-deposited surface area (LDSA) concentrations (NSAM model 3550 and AeroTrak TM 9000, TSI Inc), (Bau et al., 2012) can measure a value related to particle surface area concentration and are more sensitive to ultrafine particles than gravimetric methods (e.g., Gravimetric Filter Method and Mass PM Analyzer, Mohr et al., 2005). Such instruments may offer the ability to explore source-related ultrafine particle exposure as they deliver high temporal and potentially spatially disaggregated measurements, which addresses requirements around the nature of ultrafine particles to coagulate quickly and create significant concentration gradients (Imhof et al., 2005; Peters et al., 2009). In these instruments a corona discharge produces unipolar ions, which can diffuse towards the particles and an electrometer is used to measure the total charge that transfers from the ions to the particles; the amount of charge is related to the active surface area concentration (Asbach et al., 2009; Baltensperger et al., 2001).

LDSA concentrations have been reported at urban background sites in Upper Austria (Moshammer and Neuberger, 2003), Minneapolis (Wilson et al., 2007), Los Angeles (Ntziachristos et al., 2007), Lisbon (Albuquerque et al., 2012; Gomes et al., 2012), Italy (Buonanno et al., 2010, 2012; Geiss et al., 2016; Spinazzè et al., 2015), Switzerland (Eeftens et al., 2015; Fierz et al., 2011), Barcelona (Reche et al., 2015), Helsinki (Kuuluvainen et al., 2016). However, the concentrations of LDSA in UK cities have not been reported in any previous studies and it is important to know the levels of the LDSA in UK as a reference for future studies.

The aim of the present study is to explore extra-annual cycles of LDSA in a mid-size urban environment. In addition, the association of the calculated LDSA with equivalent black carbon mass concentration (eBC) was investigated to assess the relationship of LDSA

Fig. 1. AURN and Brookfield sites at Leicester University campus, sampling locations.