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Sub-micron particle number size distribution characteristics at two urban locations in Leicester

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ABSTRACT

The particle number size distribution (PNSD) of atmospheric particles not only provides information about sources and atmospheric processing of particles, but also plays an important role in determining regional lung dose. Owing to the importance of PNSD in understanding particulate pollution two short-term campaigns (March–June 2014) measurements of sub-micron PNSD were conducted at two urban background locations in Leicester, UK. At the first site, Leicester Automatic Urban Rural Network (AURN), the mean number concentrations of nucleation, Aitken, accumulation modes, the total particles, equivalent black carbon (eBC) mass concentrations were 2002, 3258, 1576, 6837 # cm^{-3}, 1.7 μg m^{-3}, respectively, and at the second site, Brookfield (BF), were 1455, 2407, 874, 4737 # cm^{-3}, 0.77 μg m^{-3}, respectively. The total particle number was dominated by the nucleation and Aitken modes, with both consisting of 77%, and 81% of total number concentrations at AURN and BF sites, respectively. This behaviour could be attributed to primary emissions (traffic) of ultrafine particles and the temporal evolution of mixing layer. The size distribution at the AURN site shows bimodal distribution at ~22 nm with a minor peak at ~70 nm. The size distribution at BF site, however, exhibits unimodal distribution at ~35 nm. This study has for the first time investigated the effect of Easter holiday on PNSD in UK. The temporal variation of PNSD demonstrated a good degree of correlation with traffic-related pollutants (NO_X, and eBC at both sites). The meteorological conditions, also had an impact on the PNSD and eBC at both sites. During the measurement period, the frequency of NPF events was calculated to be 13.3%, and 22.2% at AURN and BF sites, respectively. The average value of formation and growth rates of nucleation mode particles were 1.3, and 1.17 cm^{-3} s^{-1} and 7.42, and 5.3 nm h^{-1} at AURN, and BF sites, respectively. It can suggested that aerosol particles in Leicester originate mainly from traffic and domestic heating emissions.

1. Introduction

Atmospheric aerosol particles are ubiquitous and have negative impacts on human health, air quality and global climate change (Lohmann and Feichter, 2005; Pope and Dockery, 2006; Stevens and Feingold, 2009). Epidemiological studies have revealed that the existence of a relationship between fine particle concentration and respiratory and cardiovascular diseases (Pope, 2000). Numerous studies have since proposed that ultrafine particles (UFP – particles < 100 nm) are more toxic compared to larger particles of same composition and the adverse health effects caused by UFP number concentrations have been indicated to be stronger than those by the fine particle mass concentrations (Peters et al., 1997; Penttinen et al., 2001; Li et al., 2002; Nel, 2005). Nevertheless, current air quality standards are based on the particle mass concentrations. The mass concentrations of the particles < 100 nm, which really govern the total particle number concentrations in urban areas are insignificant (Seinfeld and Pandis, 1998). Thus, current air quality measurements might be insufficient to permit assumptions to be drawn concerning the association between particle number and the detrimental health effects. It is, therefore, vital to measure the particle number size distributions in order to fully understand the environmental effects of atmospheric ultrafine particles (Peters et al., 1997; Penttinen et al., 2001). Air quality at many urban background sites is influenced by road traffic emissions with diurnal patterns found to be strongly influenced by the primary traffic exhaust emissions in the urban areas (Tuch et al., 2003; Wehner and Wiedensohler, 2003; Hussein et al., 2004; Stanier et al., 2004; Rodríguez and Cuevas, 2007; Pérez et al., 2010). Traffic emissions are considered to be one of the most significant sources of UFP number concentrations in the urban atmosphere, with the other significant

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source originating from are particile formation (NPF) (Shi et al., 2001; Stanier et al., 2004; Brines et al., 2015). In addition, NPF events can occur widely under various meteorological and atmospheric conditions (Kulmala et al., 2004; Dal Maso et al., 2005). NPF can be a second source of UFPs in urban areas (Brines et al., 2015). Several NPF studies in rural and urban areas have revealed that NPF is generally favoured under high insolation and wind speed, low relative humidity, and low pre-existing particle surface area (Kulmala et al., 2004; Kulmala and Kerminen, 2008). As such, the increased background concentration of UFPs in polluted areas seems to decrease the NPF. Nevertheless, NPF events are still observed in many polluted urban areas and some of the studies have revealed that strong correlation between the NPF and the levels of vapour-phase H_2SO_4 which is mainly produced by the chemical oxidation of SO_2 with the hydroxyl radical during daytime (Jeong et al., 2004; Kulmala et al., 2012; Zhu et al., 2013; Wang et al., 2014).

eBC is typically formed by incomplete combustion of fossil fuels, biofuel and biomass, and is emitted from traffic. Several studies have shown that a strong relationship between black carbon and road traffic emissions (Fruin et al., 2008; Pérez et al., 2010; Boogaard et al., 2011; Invernizzi et al., 2011; Reche et al., 2011; Butterfield et al., 2015) and biomass burning emissions (Ingrid Sundvor et al., 2012; Butterfield et al., 2015). Moreover, numerous studies have revealed that exposure to road traffic emissions is best assessed by combining measurements of particle number and eBC concentrations (Harrison et al., 2004; Smargiassi et al., 2005; Rodríguez and Cuevas, 2007), since these parameters need to be controlled by air quality limit values.

In spite of its importance, aerosol size distributions at urban and road sites in UK have been reported at relatively few sites (see Table 1). To our knowledge, there are no studies regarding particle number size distribution measurements and analysis of NPF events in detail in a UK urban area such as Leicester. Information on the behaviour of particle number size distributions is still sparse. There is also a lack of knowledge about particle number size distributions generally in the UK. The objective of the present study is to characterize the NPF events and its impact on PNSD by taking measurements at two sites within the urban area of Leicester. This study also investigates the effect of Easter school holiday on PNSD. Daily and weekly variations of PNSD, and the difference between daily patterns of weekday and weekends of PNSD are explored. The influence of traffic emissions and meteorological conditions on PNSD are also investigated.

The study was carried out between March 2014 and June 2014 over which time particle number size distributions (PNSD) were measured concurrently with black carbon mass concentration (eBC), total particle number (TNC), and NO_X at two sites in Leicester. This study reports on the first results of PNSD measurements which were taken as part of the air quality monitoring network established across North West Europe as part of the JOint Air QUality INitiative (JOAQUIN, www.joaquin.eu), an INTERREG IVB funded European project, which aims at supporting health-oriented air quality policies in North-West Europe. More information can be found in the Joaquin report and publications (Joaquin, 2015; Cordell et al., 2016; Hofman et al., 2016; Hama et al., 2017).

Table 1
Summary of previous PNSD studies at various UK sites.

Site	Size range (nm)	Study
Urban background site (London)	10–415	Rodríguez and Cuevas (2007)
Road site (London)	14.6–661.2	Dall'Osto et al. (2011)
Urban background site (London)	8–700	von Bismarck-Osten et al. (2013)
Road site (London)	19.2–600	von Bismarck-Osten et al. (2013)
Urban background site (Birmingham)	10–1000	Harrison et al. (1999)
Road site (Birmingham)	9.47–359	Shi et al. (2001)
Road site (Manchester)	4–160	Longley et al. (2003)
Rural site (Harwell)	11–450	Charron et al. (2007)
Urban background site (Cambridge)	10–2500	Kumar et al. (2008)
Road site (Leicester)	5–1000	(Agus et al., 2007)
AURN urban background (Leicester)	10–1093	This study
BF urban background (Leicester)	10–1093	This study

2. Experimental

2.1. Sampling sites

The measurements were a part of the JOAQUIN project (www.joaquin.eu). Sampling was conducted during the spring (March–June 2014) at two sites located at the University of Leicester (Fig. 1). More detailing about the characteristics and locations of the sampling sites can be found in Hama et al. (2017). In this study hourly traffic density data was provided by Leicester City Council was used. For a detailed overview of the monitoring sites and the JOAQUIN project, the reader is referred to the final report (Joaquin, 2015).

2.2. Instrumentation

Table 2 summarizes the availability of monitors for PNSD, eBC, TNC, NO_X and O_3 per site. Measurements at the AURN and BF sites were carried out with the devices in the mobile measurement trailer.

In this study the particle size distribution was measured by a Scanning Mobility Particle Spectrometer (Grimm SMPS + C 5420 with L-DMA). TNC was measured with a TSI model 3783 water-based condensation Particle counter (CPC). This instrument measures the number of particles in the size range of ~7 to 1000 nm. To measure the eBC (Petzold et al., 2013) mass concentration a MAAP (model 5012, Thermo-Scientific) was used. NO_X were measured by a Thermo 42i NO-NO_2-NO_X monitor. The concentrations of O_3 were measured by UV absorption at AURN site. The reference method for the determination of concentrations of O_3 are described in European Standard EN14625, more information can be found in this website (https://uk-air.defra.gov.uk/networks/monitoring-methods?view=eu-standards). For a detailed overview of the instruments and monitors that were used in this study and also about data quality assurance in the JOAQUIN project, the reader is referred to Hama et al. (2017).

Meteorological data were obtained from a mobile laboratory van during this study. Moreover, Meteorological data were also provided by the Air Quality Group from the Leicester City Council, located 4.9 km northwest the AURN site. The mean, median, and max values of wind speed (WS), wind direction (WD), temperature (T), and the relative humidity (RH) form March to May 2014 at both sites are shown in Table 3.

2.3. Data processing and analysis

Particle number concentrations for different size ranges were calculated by the particle size distribution from the Grimm SMPS measurement. The particle number concentrations were categorised into $10 \le d \le 1093$ nm (N_{total}), $100 \le d \le 1093$ nm (N_{acu}), $25 \le d < 100$ nm (N_{Aitken}) and $d < 25$ nm (N_{nuc}), for total, accumulation mode, Aitken mode and nucleation mode, respectively. Data analyses have been carried out using the Openair software package (Carslaw and Ropkins, 2012; Carslaw, 2015) using R software (R Core Team, 2014) and sometimes Igor software. The Openair is freely available as an R package.

All data were screened for irregularities. Continuous air quality data collected during instrument errors or maintenance were removed from the analysis. For PNSD, particle losses to the surface of the sampling system and the measuring device can occur via diffusion. Therefore,