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# Comparative study of ultrafine atmospheric aerosol within a city

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## HIGHLIGHTS

- Median particle number concentrations varied by a factor of 40 within a city.
- Diurnal $N_{6-100}$ patterns were different for various urban sites.
- Median diameter of number size distributions decreased with anthropogenic impact.
- Nucleation strength factor was introduced to quantify the contribution of nucleation.

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## ABSTRACT

Particle number size distributions in a mobility diameter range of 6–1000 nm and size-resolved number concentrations were determined with a time resolution of 10 min for a near-city background, city centre, street canyon and road tunnel environments in Budapest. Median $N_{6-100}$ concentrations for the sites listed were $3.1 \times 10^3$, $9.3 \times 10^3$, $19.4 \times 10^3$ and $123 \times 10^3$ cm$^{-3}$, respectively. Contributions of the ultrafine (UF) particles (<100 nm) to the total particle number for all locations were rather large (up to 86%), and do not seem to vary substantially in time. Diurnal variations of the mean $N_{6-100}$ concentrations had different patterns for both the various urban sites, and for workdays and weekends. Nucleation strength factor (NSF) was introduced for the first time to quantify the relative importance of new particle formation with respect to all sources of UF particles. During the daytime in summer, nucleation in the near-city background was a major production process of UF particles with a daily mean relative contribution of 42%. In the city centre and street canyon, the daily mean relative contributions of nucleation to the UF particles were 30% and 23%, respectively. Median particle diameters for the background, city centre, street canyon and road tunnel environments were 61, 42, 35 and 42 nm, respectively, so they were jointly influenced with the anthropogenic impact and aerosol ageing. Monthly mean frequency of new particle formation and growth events in the background seems somewhat larger, while it appears smaller for the street canyon in comparison to the city centre.



## 1. Introduction and objectives

Ultrafine (UF) atmospheric aerosol particles (with an electric mobility diameter <100 nm) are usually present in relatively large number concentrations (daily medians up to $10^4$–$10^5$ cm$^{-3}$) and abundances (70–90% of all aerosol particles) in cities (Kulmala et al., 2004; Aalto et al., 2005; Putaud et al., 2010; Backman et al., 2012; Borsós et al., 2012). The particles are either emitted directly from high-temperature processes (such as automotive road traffic exhaust, industrial combustion processes, cooking and residential heating) or they are formed in the air by atmospheric nucleation. Importance of understanding the contribution of primary and secondary particles on regional and global spatial scales was recently outlined by Reddington et al. (2011).

Although studies are scarce, there is a body of evidence that UF particles represent specific and excess health risks relative to coarse or fine particles of the same or similar chemical composition (Oberdörster et al., 2005; JRC-EASAC, 2011). Their health effects are mainly caused by 1) the large number of insoluble particles deposited in the respiratory system, 2) their large surface area and 3) very small size. Clearance mechanisms of the respiratory system are only able to remove huge numbers of deposited particles with limitations, and the rest of particles contribute to releasing free radicals and causing inflammatory effects (Kreyling et al., 2006).

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The smallest UF particles can cross the cellular membrane of the respiratory epithelium, enter into the bloodstream or the cellular interstitial fluid. This modifies certain functions of the blood, and yields enhanced and systematic translocations of the deposited UF particles from the respiratory system to other organs including the liver, heart and nervous system, and can cause adverse health effects there (Oberdörster et al., 2005). The impacts are most severe on the elderly and people with compromised respiratory system such as the chronic obstructive pulmonary disease (Morawska et al., 2004; Geiser et al., 2005; Morawska, 2010). Ultrafine particles can impact the urban climate and heat island in city centres as well because they can grow to cloud condensation nuclei (CCN), and can influence the cloud formation and properties (Reddington et al., 2011). According to modelling studies, at least 50% of the CCN is related to the nucleation process on a global scale (Merikanto et al., 2009).

Several research studies were devoted to concentrations and physical properties of UF particles in cities (e.g., Väkevä et al., 2000; Woo et al., 2001; Wehner and Wiedensohler, 2003; Hussein et al., 2004; Aalto et al., 2005; Harrison and Jones, 2005; Stolzenburg et al., 2005; Jeong et al., 2006; Rodríguez et al., 2007; Avino et al., 2011; Salma et al., 2011a, 2011b; Dall'Osto et al., 2013). Methods to differentiate among the major production processes of particles were also proposed (Shi et al., 1999; Alam et al., 2003; Watson et al., 2006; Qian et al., 2007; Park et al., 2008; Costabile et al., 2009). There is, however, limited information available on the UF aerosol within a city. Despite that some properties or features of the UF aerosol are expected to differ in various urban environments. This is due to the relatively short residence time of most of these particles (Salma et al., 2011a), and the influential vicinity of their major production processes. Differences in the properties of UF particles could affect their impacts on public health and the environment. As far as mass size distributions are concerned, substantial differences in the distributions and some differences in the lung deposition for various urban sites within a city were indeed observed (Salma et al., 2002a, 2002b). The main objectives of this paper are to report average particle number concentrations, to interpret their temporal variation for some important size fractions and for derived properties such as nucleation strength factor; to determine representative and typical particle number size distributions for further modelling; and to discuss the frequency variation of new particle formation and growth events within a city.

## 2. Applied methods

### 2.1. Measuring methods

Aerosol particles were measured by a flow-switching type differential mobility particle sizer (DMPS, Aalto et al., 2001; Salma et al., 2011a) including a $^{241}$Am neutraliser, a Nafion semi-permeable membrane drier, a 28-cm long Hauke-type differential mobility analyser and a butanol-based condensation particle counter (CPC, model 3775, TSI, USA). The system operates in an electric mobility diameter range from 6 to 1000 nm in 30 size channels at two sets of flows. In the first flow mode, 20 channels are measured (from 6 to 200 nm), while in the second flow mode, 10 channels are acquired (from 200 to 1000 nm). A shoulder around 200 nm often appears in the size distributions as an artefact caused by switching the flow and high voltage parameters. Its extent usually remains below 10% although it occurs as a systematic deviation. Fortunately, no major aerosol formation and transformation processes are exclusively confined to the diameter range affected. The diameters obtained refer to the dry state of particles since the DMPS operates in dried sample flow (with a typical relative humidity, RH $<$ 20%). The measurements were performed with a time resolution of approximately 10 min. The measuring system and method fulfil the recommendations of the international technical standards (Wiedensohler et al., 2012).

### 2.2. Measurement environments and time intervals

The measurements were performed in Budapest, Hungary. Its population is 2 million. The major pollution sources include vehicular road traffic, residential heating and household activities. Long-range transport of some pollutants also plays a role (Salma and Maenhaut, 2006). Contributions of passenger cars and buses to the vehicle fleet registered in Budapest and Pest County are 87% and 0.46%, respectively (OKJ, 2010). Diesel-powered vehicles shared 18.2% and 97% of the national passenger car and bus fleets, respectively. Unleaded petrol is exclusively sold for road vehicles, and the diesel fuel marketed for road vehicles contains S in a concentration $<$10 ppm. The experimental work was realised at four selected sites which represent different urban environments, i.e., a near-city background, a city centre, a street canyon and a road traffic microenvironment.

The measurements in the near-city background were performed at the KFKI Atomic Energy Research Institute (latitude 47° 29′ 12.5″ N, longitude 18° 57′ 17.7″ E, altitude 424 m above sea level, a.s.l.) continuously from 1 June to 10 August 2011. The site is situated within a woody campus on the western border of Budapest. It is expected to represent the air masses entering the city since the prevailing wind direction is NW. The measurements in the city centre were accomplished at the Eötvös University's campus at Lágymányos (latitude 47° 28′ 29.8″ N, longitude 19° 03′ 44.6″ E, altitude 114 m a.s.l.) continuously from 3 November 2008 to 2 November 2009 (Salma et al., 2011a). The site is situated in a distance of 80 m from the bank of the river Danube. The street canyon measurements were performed in a building of the Eötvös University located in the city centre (5 Rákóczi Street, latitude 47° 29′ 39.4″ N, longitude 19° 03′ 36.3″ E, altitude 111 m above a.s.l) continuously from 28 March to 31 May 2011. The street is approximately 2.0 km long, 25–40 m wide, and has a typical height of 25–30 m. The street belongs to regular long street canyons (Hunter et al., 1992). For this type of canyons, the bulk perpendicular air flow skims over the canyon and usually produces a single vortex within it, which favours air pollution build-up. The major ventilation occurs in longitudinal direction. The measurements in the traffic microenvironment were carried out in the Castle District Tunnel (latitude 47° 29′ 54.5″ N, longitude 19° 02′ 24.6″ E, altitude 106 m a.s.l. at its eastern gate) continuously from 12 to 26 July 2010 (Salma et al., 2011b). The tunnel has a single, straight bore with a length of 350 m, a width of 9.3 m, and it varies from 7.9 to 10.7 m in height. It is situated in the city centre, and is oriented perpendicular to the river Danube with its closer, eastern gate in a distance of approximately 130 m from the river bank. The tunnel comprises two-lane road traffic, and involves a pedestrian lane and a service curb along the sides. The tunnel has an elevation of 1.8% toward the western gate, which promotes passive ventilation. The air movement is enforced by ventilation without filtering from about 8:00 to 18:00 local time on workdays. Ambient air is drawn from the outer sideway spaces near the gates and above the bore by a mine ventilator through shafts, and it is delivered into the bore through a portal at a distance of 169 m from the eastern gate. The nominal ventilation rate is 1900 $m^3$ $min^{-1}$. Heavy-duty vehicles are not allowed to enter the tunnel. The measuring instrument was set up in a spare ventilation hall of the tunnel in a distance of 226 m from the eastern gate.