# Time-resolved number concentrations and size distribution of aerosol particles in an urban road tunnel

Imre Salma<sup>1</sup>), Tibor Borsós<sup>1</sup>), Pasi P. Aalto<sup>2</sup>) and Markku Kulmala<sup>2</sup>)

<sup>1)</sup> Institute of Chemistry, Eötvös University, P.O. Box 32, H-1518, Budapest, Hungary

<sup>2)</sup> Department of Physics, P.O. Box 64, FI-00014 University of Helsinki, Finland

Received 1 Dec. 2010, accepted 7 Mar. 2011 (Editor in charge of this article: Hannele Korhonen)

Salma, I., Borsós, T., Aalto, P. P. & Kulmala, M. 2011: Time-resolved number concentrations and size distribution of aerosol particles in an urban road tunnel. *Boreal Env. Res.* 16: 262–272.

We used flow-switching type differential mobility particle sizer in a diameter range of 6–1000 nm and vehicle counting detectors with a time resolution of 10 min for 15 days in July 2010 in the Castle District Tunnel, Budapest. The total particle number concentrations varied from  $5.1 \times 10^3$  to  $465 \times 10^3$  cm<sup>-3</sup> with a median of  $143 \times 10^3$  cm<sup>-3</sup>. The median was greater by a factor of 12 than that for the urban ambient air. The mean (± SD) contribution of ultrafine particles to the total particle number was  $85\% \pm 1\%$ , which is significantly greater than for the urban ambient air. Diurnal variations in the number concentrations in the tunnel on workdays exhibited different time pattern than the traffic, which was explained by ventilation. Number size distributions of particles were resolved into Aitken and accumulation modes with the overall median diameters of 33 and 86 nm, respectively. The mean Aitken mode/accumulation mode concentration ratio was 3.6.

## Introduction

Aerosol particles are present in large concentrations in polluted urban environments. Ultrafine aerosol particles (with a mobility diameter < 100nm) are the dominant constituent in terms of particle number at these locations. Impact of ultrafine particles on human health has been increasingly recognised, and is regarded to be harmful. Ultrafine particles represent specific health risks relative to coarse or fine particles of the same or similar chemical composition. They can enter directly into the bloodstream from the lungs, and can be deposited in various sensitive organs in the body such as the heart or central nervous system (Oberdörster et al. 2005, Morawska 2010). Number concentrations in cities are often divided into Aitken- and accumulation-mode concentrations. In Budapest, number median mobility diameters of the Aitken and accumulation modes typically vary between 20 and 60 nm, and 90 and 120 nm, respectively (Salma et al. 2011). Residence time for the Aitken mode in the planetary boundary layer is relatively short, e.g., in central Budapest it is < 1 h. Therefore, the appearance and presence of ultrafine particles in the air in increased concentrations can be directly related to their major sources. Ultrafine aerosol particles are either emitted directly from hightemperature processes or they are formed in the air as secondary particles. Their major production types in urban environments include automotive road traffic emissions (Morawska et al. 2008), heating, household burning emissions and atmospheric nucleation (Kulmala et al. 2004, 2005). Combustion sources emit greater (i.e., Aitken

mode) particles than those formed by nucleation because particles from high-temperature sources grow already inside or immediately after leaving the source due to coagulation and condensation of semi-volatile substances when the exhaust or flue-gas are cooled and diluted (Ristovski et al. 1998, Shi and Harrison 1999, Charron and Harrison 2003). Primary ultrafine particles have high relevance for urban air quality and human health. Road traffic is their major source in many urban microenvironments (Morawska et al. 1998), and high exposures to particle numbers are realised in these locations. Up to 50% of daily ultrafine exposure of commuters in Los Angeles occurs inside vehicles (Zhu et al. 2007). Properties and effects of the vehicular emissions are often studied in road tunnels for several reasons (Kristensson et al. 2004). Road tunnels represent a segregated microenvironment with dominating vehicular emissions, closed character, restricted dispersion, lack of sunlight, and specific meteorological and boundary conditions. Various tunnel studies dealt with gaseous emissions and aerosol mass concentrations. Particle numbers only became a subject of the investigations in recent years. Their results were summarised for both fixed-site and on-vehicle based measurements by Westerdahl et al. (2005) and Knibbs et al. (2009). Health significance of road tunnels was also studied, and undesirable respiratory and cardiovascular implications were reported (Svartengren et al. 2000, Larsson et al. 2007, Mills et al. 2007).

Production, growth and properties of ultrafine atmospheric aerosol particles in central Budapest were studied for one year (Salma et al. 2011) with the primary focus on new particle formation and subsequent growth. It was concluded that in central Budapest vehicular emissions often determine the particle number concentrations. As a continuation of this research, we performed a fixed-site measurement campaign in the major road tunnel in Budapest, i.e., in the Castle District Tunnel to investigate specifically the particle emissions from road traffic. Number concentrations in the tunnel or other traffic microenvironments were not studied earlier according to our knowledge. The main objective of this air quality research was to quantify the levels and time variation in particle number concentrations in the tunnel. Our goal was achieved by investigating the diurnal and weekly time variation of the concentrations and time-resolved number size distributions with respect to traffic flow.

### Methods

#### Measurements

The measurements were carried out in the Castle District Tunnel (47°29.6'N, 19°2.5'E, altitude of the eastern gate 106 m a.s.l.), which is situated in central Budapest. The tunnel has been in operation since 1857, and it was reconstructed in 1973. It is 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. Its location is perpendicular to the Danube River, and the closer, eastern gate is located approx. 130 m from the river bank. The tunnel comprises altogether 2-lane road traffic, and involves one pedestrian lane and one service curb along the two sides. The tunnel has an inclination of 1.8% towards the western gate, which often leads to passive ventilation. The air movement is enforced by ventilation without filtering from about 08:00 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 as fresh air into the bore through a portal at a distance of 169 m from the eastern gate. The nominal ventilation rate is ca. 1900 m<sup>3</sup> min<sup>-1</sup>. No jet fans are installed inside the bore for further longitudinal ventilation. Typical wind speeds in the side lanes near the western gate within the tunnel when ventilation is switched off and on were 1.0 and 2.3 m s<sup>-1</sup>, respectively. No information is available on the radial concentration gradient but it is reasonable to assume that it is small after the ventilation port since the air is turbulently mixed there. Surfaces of the bore are washed twice a year. Vehicles using the tunnel are not subject to a toll. Total number of vehicles passing the tunnel on an average workday is approximately  $20 \times 10^3$ . Maximum permitted vehicle speed is 50 km h<sup>-1</sup>. The traffic flow and speed of vehicles is considerably lowered by congestions that extend from the cross points outside the gates into the tunnel during the morning (approx. 07:00-09:30) and afternoon

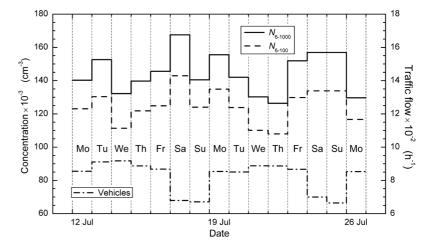
(approx. 14:30–18:00) rush hours on workdays. Heavy-duty vehicles and lorries are not allowed to enter the tunnel. Pedestrian crossing is not recommended by municipal authorities. Number of vehicles passing the tunnel in both directions was obtained every 10 min from online loop-counting devices used for controlling traffic lights near the western gate. Passenger cars and buses comprised 87% and 0.46% of the vehicle fleet registered in Budapest and the Pest County, while diesel-powered vehicles shares in the national passenger car fleet and bus fleet were 18.2% and 97%, respectively (OKJ 2010). The mean lifetimes of passenger cars and buses registered in Budapest and the Pest County were 9.9 and 13.9 years, respectively. Unleaded petrol has exclusively been used for road vehicles in Hungary since 1999 (Salma et al. 2000, Salma and Maenhaut 2006). Since July 2005, diesel fuel for road vehicles in Hungary has contained sulphur in a concentration of less than 10 ppm which is in accordance with the EU specification (Directive 2009/30/EC of the European Parliament and of The Council 2009). No equipment is mounted for monitoring the air quality or alarming purposes in the tunnel. During daylight in 1999, the mean (± SD) aerosol mass concentrations in the tunnel for PM<sub>10-2.0</sub> and  $PM_{20}$  size fractions were 430 ± 170 µg m<sup>-3</sup> and  $149 \pm 56 \,\mu \text{g m}^{-3}$ , respectively (Salma *et al.* 2001, 2005). The coarse mode was pronounced in mass size distributions, even for elements of typically anthropogenic origin. This was explained by settling, association of coarse and fine particles, and their repetitive joint resuspension.

We installed the measuring instrument in a spare ventilation hall of the tunnel at a distance of 226 m from the eastern gate. The sampling inlet was set up at a height of 1.3 m above ground with no upper size cut-off device applied. No attempt was made to create isokinetic sampling conditions due to the small particle diameters of interest. The sampling line was made of a Cu tubing with an internal diameter of 4 mm and length of approx. 1.5 m. The measurements were performed using a flow-switching type differential mobility particle sizer (DMPS, Aalto *et al.* 2001). The main parts of the DMPS include a <sup>241</sup>Am neutralizer, a Nafion semi-permeable membrane drier, a 28-cm-long Hauke-type differential mobility differential mobility particles yet and yet and yet.

ferential mobility analyser and a butanol-based condensation particle counter (CPC, model 3775, TSI, USA). The DMPS records particle number concentrations in an electric mobility diameter range of 6-1000 nm in 30 channels. The diameters refer to dry state of particles since the DMPS operates in the dried sample flow. Time resolution of the measurements was approximately 10 minutes. The maximum detectable number concentration of the CPC was  $5 \times 10^5$  cm<sup>-3</sup>, which was not approached during the measurements. The performance of the DMPS to measure the total number of particles accurately was tested with another CPC of the same type, and an excellent agreement was achieved. The actual instrument applied was described in detail by Salma et al. (2011). The measurements were carried out continuously from Monday, 12 July 2010 to Monday 26 July 2010, thus for 15 days.

#### Data treatment

The measured data were mathematically inverted online after each measurement cycle (Wolfenbarger & Seinfeld 1990). The inverted concentrations were utilised to generate contour plots showing jointly time variations in particle diameter and normalised particle number concentrations for a unity logarithmic diameter. The inverted data were also used for calculating particle number concentrations in the diameter ranges from 6 to 1000 nm  $(N_{6-1000})$  and from 6 to 100 nm  $(N_{6-100})$  with a time resolution of ca. 10 min. The former size-fraction represents total number of aerosol particles while the latter fraction can be associated with ultrafine particles. Number concentrations of particles with a diameter above 1000 nm is expected to be negligible in cities in relation to the concentrations of these size fractions. We are aware that numbers of coarse particle can be also non-negligible for some specific aerosol types. Daily and overall average concentrations were calculated for the size fractions. Diurnal variation in the concentrations, averaged by the time of day separately for workdays and weekends were also obtained. All number size distributions were fitted by lognormal functions using the DoFit algorithm (Hussein et al. 2005) to obtain modal concentrations,



**Fig. 1**. Daily median particle number concentration in size fractions of 6–1000 nm ( $N_{6-1000}$ ) and 6–100 nm ( $N_{6-100}$ ), and the daily mean traffic flow in the Castle District Tunnel from 12 to 26 July 2010.

number median mobility diameters (NMMDs) and geometric standard deviations (GSDs) for the modes identified. Various concentration data sets and traffic flow data were subjected to correlation analysis.

Experimental results of the present study were sometimes compared with the corresponding ambient data for central Budapest. The ambient data were obtained at the Lágymányos campus of the Eötvös University (47°28'29''N, 19°03'43''E, 115 m a.s.l.) between 3 November 2008 and 2 November 2009 (Salma et al. 2011). This site is located downwind from the tunnel at a distance of about 3.1 km from its eastern gate, and 80 m from the river bank. The prevailing wind direction in Budapest is NW. It brings clean air into the central part, dilutes the polluted air there, and leaves almost unrestrictedly in the S-SE direction. It is expected that both the external air at the tunnel and ambient air at the Lágymányos campus are influenced or linked by a wind channel which is formed above the river.

## **Results and discussion**

#### Concentrations

Daily mean traffic flow in the tunnel was rather constant during the workdays; on the weekends it was smaller by 25%–30% (Fig. 1). Variations in the daily median concentrations on weekdays did not follow changes in the traffic flow. Total particle number concentrations varied from  $126 \times 10^3$  cm<sup>-3</sup> (Thursday, 22 July 2010) to  $167 \times 10^3$  cm<sup>-3</sup> (Saturday, 17 July 2010) with a median of  $143 \times 10^3$  cm<sup>-3</sup>. The concentrations were greater by factors of 33, 5.7 and 11.9 than the corresponding minimum, maximum and median values obtained for the ambient air in central Budapest over one year. (It is worth mentioning here that no obvious seasonal variation in the daily median number concentration was identified in the ambient data set.) The daily median concentration of ultrafine particles in the tunnel varied from  $108 \times 10^3$  cm<sup>-3</sup> (Thursday, 22 July 2010) to  $143 \times 10^3$  cm<sup>-3</sup> (Saturday, 17 July 2010) with a median of  $123 \times 10^3$  cm<sup>-3</sup>. For the workdays, the daily medians varied irregularly from day to day. The average traffic flow and composition of vehicle fleet (e.g., relative frequency of buses), which influence the concentration levels, were more or less constant on a daily scale. The concentration levels realised within the tunnel were also affected by ambient concentrations and micrometeorological circumstances near and within the tunnel via passive and enforced ventilation. The latter quantities changed irregularly, and they likely caused the observed variability. High concentration levels were reached on Saturdays. This was caused by the joint effect of ventilation that was switched off and smaller traffic flow on weekends. The median daily concentrations were in the range  $41 \times 10^{3}$ -760  $\times 10^{3}$  cm<sup>-3</sup> reported for other urban tunnels, and were usually smaller than the typical average values (Knibbs et al. 2009). The lower concentration level can be also connected

to the fact that the Castle District Tunnel is a short tunnel. At the same time, we are aware that there are important differences (e.g., in the dimensions, operational circumstances, engineering systems, vehicle densities, ventilation systems, cleaning frequencies) among various urban tunnels. Moreover, the measuring methods, techniques and approaches applied in tunnel studies were as diverse as the tunnels themselves. Therefore, conclusions on the general air quality of various tunnels can not be always deduced by direct comparison of experimental results.

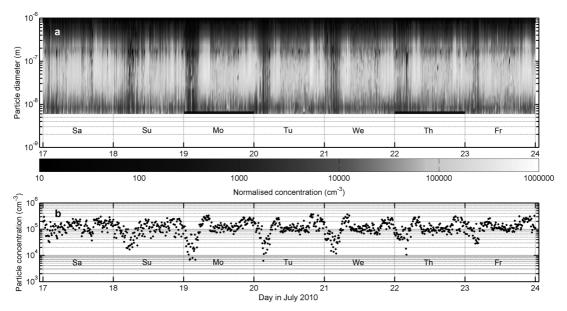
It is more realistic, however, to consider measured 10-min concentrations because the travellers, cyclists or pedestrians spend a comparable time interval in tunnels, and actual concentrations over this period can be much greater or much smaller than the daily medians. The particle number concentrations cover a range of two orders of magnitude (Table 1). The largest measured total particle number concentration of  $465 \times 10^3$  cm<sup>-3</sup> was reached at 18:50 on Tuesday, 20 July 2010, and it was about 40-times greater than the yearly median for the ambient urban air. Strong association between  $N_{6,100}$ and  $N_{6-1000}$  mentioned above was confirmed by Pearson's correlation coefficient equalling 0.992 (p < 0.0001). This points to their common major emission source which is unambiguously road traffic. Contribution of ultrafine particles to the total number concentrations on a daily basis ranged from 61% to 98% (mean  $\pm$  SD = 85%  $\pm$  1%) (see Table 1). The minimum is slightly greater than the corresponding minimum of 58% for the ambient urban air, while the maximum and mean  $\pm$  SD are significantly greater than the corresponding ambient values of 92% and

 $79\% \pm 6\%$ , respectively. Smaller ratios typically occurred at the very beginning of the day (until 04:00) when the traffic flow was the lowest (see below). This can be explained by the fact that vehicles mainly emit particles in the ultrafine size range, and that their contribution to the total particle number concentration is greater in the tunnel than in urban areas due to the restricted ventilation and dispersion. The median particle number concentration levels within the tunnel and in the ambient air were utilised to estimate roughly the relative exposure in the tunnel with respect to the city centre. Pedestrians walking through the tunnel for 5-7 min are generally exposed to particle numbers that are equivalent to an exposure obtained for ca. 1 h 15 min while walking in the city centre. Exposure of those who walk through the tunnel during the concentration peak periods (worst-case scenario) corresponds to approx. 4-h walking in the city. The real situation can be somewhat better because there is a relationship between the concentration levels within and outside the tunnel, in particular for periods when the ventilation is activated.

The smallest traffic flow (at night) was considerably smaller on workdays than on weekends, and the median number of vehicles that passed the tunnel on workdays was 1.41 greater than that for weekends (Table 2). The daily changes in traffic flow were greater for workdays than for weekends. Both the smallest and the largest 10-min concentrations occurred on workdays, at night and in the morning, respectively. The workday/weekend ratio of the median number concentrations for both total particles and ultrafine particles were below the value of 1 (approx. 0.85). This means that the concentration levels were smaller on workdays than on week-

Table 1. Range, median, mean and standard deviation of the measured total particle concentration, ultrafine particle concentration and traffic flow in the Castle District Tunnel. Contributions of ultrafine particles to the total number of particles are also shown.

	Traffic flow (h <sup>-1</sup> )	Total particles (× 10 <sup>-3</sup> cm <sup>-3</sup> )	Ultrafine particles (× 10 <sup>-3</sup> cm <sup>-3</sup> )	Concentration ratio (%)
Minimum	87	5.1	3.7	61
Median	960	143	123	85
Maximum	1560	465	392	98
Mean	839	159	134	85
SD	390	86	71	5



**Fig. 2**. (a) Contour plot of normalised particle number concentrations showing two major traffic emissions over working days corresponding to morning and afternoon rush hours in the Castle District Tunnel from 17 to 23 July 2010. (b) Variation of total particle number concentration with time: a typical pattern consisting of a minimum and two modest maxima over working days is visible.

ends. It can be explained by the effect of ventilation which is activated on workdays between 08:00 and 18:00, and by the different traffic flow on workdays and on weekends.

No new particle formation and subsequent particle growth was identified during one week from Saturday, 17 July 2010 to Friday, 23 July 2010 (*see* Fig. 2). It was somewhat expected because new particle formation in Budapest was shown to be favoured by low pre-existing aerosol concentration levels, low condensation sink, and high solar radiation (Salma *et al.* 2011). These conditions do not occur in the tunnel at all. It can be seen that considerable variation in the number concentration occurred during the

 Table 2. Ratio of the traffic flow, total particle number concentrations and ultrafine particle number concentrations on workdays to those on weekends for the Castle District Tunnel.

	Traffic flow	Total particles	Ultrafine particles
Minimum	0.59	0.29	0.25
Median	1.41	0.84	0.86
Maximum	1.24	1.24	1.17

days, and that the variation seemed to follow a periodic pattern with one deep minimum and two modest maxima during working days. In order to study this cyclic variation in more detail, we produced average diurnal plots separately for workdays and weekends.

#### **Diurnal variations**

The traffic increased monotonically and rapidly from 05:00 to 07:00 on workdays, and reached its first maximum between 07:00 and 08:00 (Fig. 3). The traffic remained elevated until the second maximum which appeared at around 17:00, after which the number of vehicles decreased monotonically until about 04:00 next morning. The maximum of the traffic flow was limited by congestions that spread from the outside cross points near the gates into the tunnel. The mean traffic flow from 00:00 to 05:00 was greater on weekends than on workdays. This can be related to more overnight entertainment activities on weekends than on workdays. On weekends, the morning growth in traffic was slower, and the first maximum was shifted towards noon.

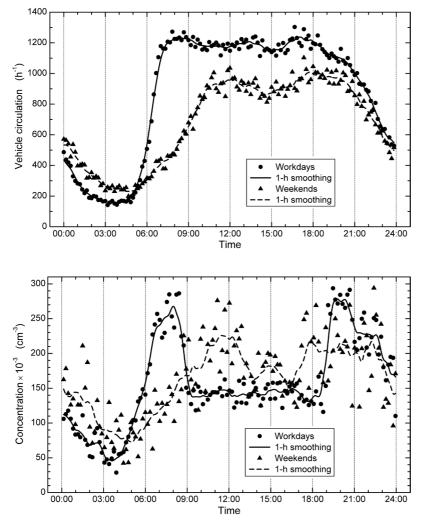
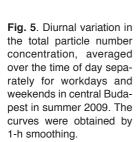


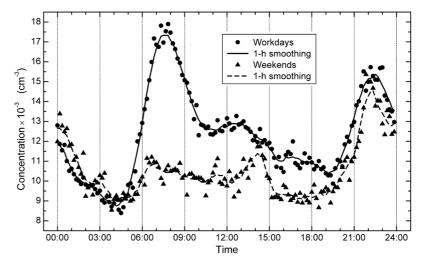
Fig. 3. Diurnal variation in traffic flow in both directions through the Castle District Tunnel, averaged for the time of day separately for workdays and weekends. The curves were obtained by 1-h smoothing.

Fig. 4. Diurnal variation in the total particle number concentration, averaged over the time of day separately for workdays and weekends in the Castle District Tunnel. The curves were obtained by 1-h smoothing.

It reached a considerably lower level than on workdays. The second maximum was broader, and it was shifted to about 18:00. Between the two maxima, a deeper minimum appeared at approximately 15:00. Diurnal variation in the traffic flow was related to the daily activity pattern of the inhabitants (Jeong *et al.* 2006, Park *et al.* 2008), and suggested that the traffic through the tunnel had significance on the urban scale.

The shapes of the curves of diurnal variations in the mean total particle number concentrations, averaged by the time of day separately for the workdays and weekends are rather different (Fig. 4). The mean number concentration between 00:00 and 05:00 were smaller on workdays than on weekends. This is a consequence of the mentioned above differences in traffic flow on workdays and on weekends. The mean concentration on workdays increases monotonically and rapidly from 05:00 to 08:00 when it reaches its first maximum of approximately  $270 \times 10^3$ cm<sup>-3</sup>. After the maximum, the mean concentration drops within an hour to about  $140 \times 10^3$  cm<sup>-3</sup> as a result of ventilation, which starts at 08:00. This concentration level is maintained constant until ventilation is on. To assess the particle number concentrations in the fresh-ventilated air, diurnal variation in the mean total particle number concentrations in the ambient air in central Budapest, averaged separately for workdays and weekends (involving holidays) in summer was derived (Fig. 5). It revealed that the median





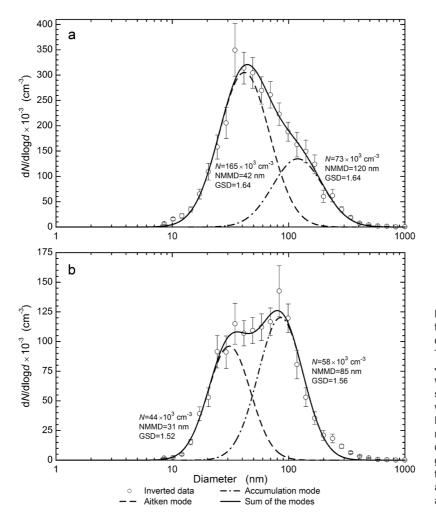
concentration of the total particle number in the ambient air in central Budapest over the period of 09:00–18:00 was between  $15 \times 10^3$  and  $11 \times 10^3$ cm<sup>-3</sup>. Its actual value depended on the weather conditions and types. It was expected that this concentration would not decreased substantially during ventilation since there was no filtration device involved. The fresh air introduced from outside dilutes the particle number concentration within the tunnel by approximately half (Fig. 4). When ventilation is switched off (at 18:00), in 1.5 hours the concentrations increased again and reached a second maximum of a similar extent as that in the morning. After 20:30, the concentrations decreased again because of the decreasing traffic flow which during that time was already substantial. On weekends, the mean total number concentrations varied similarly to the variation in the traffic flow showing a slower increase until a maximum at noon, and a second broader maximum between 18:00 and 22:30. The maximum level of the mean concentration on weekends reached approximately  $220 \times 10^3$  cm<sup>-3</sup>. The daily average concentrations on weekends were eventually greater than those on workdays. Diurnal variations in ultrafine particles for workdays and weekends exhibited similar behaviour and tendencies as for the total particles.

It is also evident from the diurnal curve for workdays that ventilation could have considerably decrease the concentrations if had it been activated approx. 2 h earlier and switched off approx. 3 h later, or if it had been controlled by some pollution (e.g.,  $NO_x$  and/or CO) monitoring instrument. Ventilation operated around midday and late evening on weekends could lower the daily exposures of travellers as well.

As expected, the correlation between the total particle number concentration and traffic flow, or between the ultrafine particle number concentration and traffic flow based on the 10-min data sets were insignificant, because of the role of ventilation. The correlation coefficient was 0.650 (p < 0.01) for the period from 20:00 to 07:30 for workdays, during which interval the number concentrations were related to vehicle emissions.

#### Size distributions

Number size distributions of aerosol particles usually exhibited a wide peak, which could be resolved by fitting into Aitken and accumulation modes. The statistical uncertainty of the fitted modal concentrations was sometimes large when the two modes were close to each other. This caused large scatter in the modal concentrations, and hindered some further evaluations. Overall median NMMDs for the Aitken and accumulation modes derived from the individual data were 33 and 86 nm, respectively. The values are comparable to the corresponding average diameters of 26 and 93 nm for the ambient urban air in Budapest, respectively (Salma et al. 2011). The greater median diameter of the Aitken mode in the tunnel can be related to several factors. The



**Fig. 6.** Number size distribution of aerosol particles in the Castle District Tunnel on Monday, 19 July 2010 at (**a**) 06:22 when ventilation was switched off, and (**b**) 14:19 when ventilation was on. Modal concentrations (*N*), number median mobility diameters (NMMD) and geometric standard deviations (GSD) for Aitken and accumulation modes are also shown.

smallest particles are affected by coagulation scavenging and wall losses more extensively than the larger particles, and these effects can shift the Aitken mode to greater median diameters. Semivolatile compounds that are present in greater concentrations in the tunnel also condense on the surface of the Aitken-mode particles, which also contributes to the shift. The mean ratio of the Aitken-mode concentration to the corresponding accumulation-mode concentration calculated on the basis of the 10-min data sets were 3.6, 3.8 and 2.1 for all days, workdays and weekends, respectively. The ratios are substantially greater than those for the ambient urban air (of 1.14), which is explained by a much greater contribution of road traffic (Aitken-mode) emissions in the tunnel. Furthermore, the Aitken mode was

substantially greater than the accumulation mode during 06:00-08:30 and 19:00-22:00 on workdays (Fig. 6a). This is explained by the largest share of the fresh emissions during the morning and afternoon peak periods when the ventilation is switched off. The accumulation-mode concentration was similar to or slightly greater than the Aitken-mode concentration during 09:00-18:30 on workdays (Fig. 6b). This is due to the fact that the ventilated air contains older emissions and aged particles, which are larger than the freshly emitted particles. This could be inferred from the mean Aitken mode to accumulation mode concentration ratios which for the ambient air and the air in the tunnel were 1.14 and 3.8, respectively. As a result, ventilation primarily diluted the concentration of Aitken-mode particles (see Fig. 6).

## Conclusions

A total number and length of road tunnels is expected to increase in many cities. Their impacts on people's health and environment should therefore be considered. For a relatively short road tunnel in Budapest, a median total particle number concentration of  $143 \times 10^3$  cm<sup>-3</sup> was obtained, which is about 12 times greater than the corresponding ambient level. The maximum of the 10-min measured concentrations reached  $465 \times 10^3$  cm<sup>-3</sup>. The daily median concentrations varied irregularly from day to day for the workdays due to changing ambient concentration levels and micrometeorological circumstances. The daily average concentrations on the weekends were greater than on the workdays. This is explained by the use of ventilation which is switched off during weekends. Mean contribution of ultrafine particles to the total particle number was 85%. The value is greater than that for the ambient air, which is related to the fact that vehicles mainly emit ultrafine particles. Diurnal variation of the traffic flow through the tunnel was rather periodic on workdays, and different than that on weekends. This was caused by the different time-activity pattern of the inhabitants on workdays and weekends. Diurnal variation of the number concentration was influenced by traffic flow, ventilation, ambient concentration levels and micrometeorological circumstances. Ventilation had a substantial impact on the air quality in the tunnel. It diluted the number concentration by a factor of approx. 2. It also suppressed the relative contribution of the Aitken mode with respect to the accumulation mode in size distributions. The present study emphasizes that tunnels are rather important microenvironments where substantial exposures to particle numbers occur. Pedestrians, cyclists, travellers in open vehicles, in vehicles with open windows or without filtration devices are subject to greater exposures. Traffic congestions in tunnels amplify the adverse effects. Traffic arrangements should prevent or decrease excess exposures. The study also demonstrates that the operation regime of the ventilation should be modified in several respects. Reducing vehicle emissions, however, seems to be a more efficient approach. The duration of the field campaign was limited in time, but it already revealed several important properties and actual features of aerosol particles in terms of particle numbers, which have public-health relevance, and it pointed out to the need for further extended investigations.

Acknowledgement: Financial support of the Hungarian Scientific Research Fund (contract K84091) is appreciated.

## References

- Aalto P., Hämeri K., Becker E., Weber R., Salm J., Mäkelä J. M., Hoell C., O'Dowd C.D., Karlsson H., Hansson H.-C., Väkevä M., Koponen I.K., Buzorius G. & Kulmala M. 2001. Physical characterization of aerosol particles during nucleation events. *Tellus* 53B: 344–358.
- Charron A. & Harrison R.M. 2003. Primary particle formation from vehicle emissions during exhaust dilution in the roadside atmosphere. *Atmos. Environ.* 37: 4109–4119.
- Directive 2009/30/EC of the European Parliament and of The Council 2009. *OJ EU* L140: 88–113.
- Hussein T., Dal Maso M., Petäjä T., Koponen I.K., Paatero P., Aalto P.P., Hämeri K. & Kulmala M. 2005. Evaluation of an automatic algorithm for fitting the particle number size distributions. *Boreal Env. Res.* 10: 337–355.
- Jeong C.-H., Evans G.J., Hopke P.K., Chalupa D. & Utell M. J. 2006. Influence of atmospheric dispersion and new particle formation events on ambient particle number concentration in Rochester, United States, and Toronto, Canada. J. Air Waste Manage. Assoc. 56: 431–443.
- Knibbs L.D., DeDear R., Morawska L. & Mengersen K. 2009. On-road ultrafine particle concentration in the M5 East road tunnel, Sydney, Australia. *Atmos. Environ.* 43: 3510–3519.
- Kristensson, A., Johansson C., Westerholm R., Swietlicki E., Gidhagen L., Wideqvist U. & Vesely V. 2004. Realworld traffic emission factors of gases and particles measured in a road tunnel in Stockholm, Sweden. *Atmos. Environ.* 38: 657–673.
- Kulmala M., Vehkamäki H., Petäjä T., Dal Maso M., Lauri A., Kerminen V.-M., Birmili W. & McMurry P. 2004. Formation and growth rates of ultrafine atmospheric particles: a review of observations. J. Aerosol Sci. 35: 143–176.
- Kulmala M., Petäjä T., Mönkkönen P., Koponen I.K., Dal Maso M., Aalto P.P., Lehtinen K.E.J. & Kerminen V.-M. 2005. On the growth of nucleation mode particles: source rates of condensable vapor in polluted and clean environments. *Atmos. Chem. Phys.* 5: 409–416.
- Larsson B.-M., Schistedt M., Grunewald J., Sköld C.M., Lundin A., Blomberg A., Sandström T., Eklund A. & Svartengren M. 2007. Road tunnel air pollution induces bronchoalveolar inflammation in healthy subjects. *Eur. Respir. J.* 29: 699–705.
- Mills N.L., Törnqvist H., Gonzalez M.C., Vink E., Robinson S.D., Söderberg S., Boon N.A., Donaldson K.A., Sand-

ström T., Blomberg A. & Newby D.E. 2007. Ischemic and thrombotic effects of dilute diesel-exhaust inhalation in men with coronary heart disease. *New Engl. J. Med.* 357: 1075–1082.

- Morawska L., Thomas S., Bofinger N., Wainwright D. & Neale D. 1998. Comprehensive characterization of aerosols in a subtropical urban atmosphere: particle size distribution and correlation with gaseous pollutants. *Atmos. Environ.* 32: 2467–2478.
- Morawska L., Ristovski Z., Jayaratne E.R., Keogh D.U. & Ling X. 2008. Ambient nano and ultrafine particles from motor vehicle emissions: Characteristics, ambient processing and implications on human exposure. *Atmos. Environ.* 42: 8113–8138.
- Morawska L. 2010. Airborne particles and health. *Air Qual. Clim. Change* 44: 13–15.
- Oberdörster G., Oberdörster E. & Oberdörster J. 2005. Nanotoxicology: an emerging discipline evolving from studies of ultrafine particles. *Environ. Health Perspect*. 113: 823–839.
- OKJ 2010. Országos Közúti Gépjárműállomány. Ministry of National Development, Budapest.
- Park K., Park J.Y., Kwak J.-H., Cho G.N. & Kim J.-S. 2008. Seasonal and diurnal variations of ultrafine particle concentration in urban Gwangju, Korea: Observation of ultrafine particle events. *Atmos. Environ.* 42: 788–799.
- Ristovski Z., Morawska L., Bofinger N.D. & Hitchins J. 1998. Submicrometer and supermicrometer particulate emission from spark ignition vehicles. *Environ. Sci. Technol.* 32: 38–45.
- Salma I., Maenhaut W., Dubtsov S., Zemplén-Papp É. & Záray, Gy. 2000. Impact of phase-out of leaded gasoline ont he air quality of Budapest. *Microchem. J.* 67:

127-133.

- Salma I., Maenhaut W., Zemplén-Papp É. & Záray Gy. 2001. Comprehensive characterisation of atmospheric aerosols in Budapest: physicochemical properties of inorganic species. *Atmos. Environ.* 35: 4367–4378.
- Salma I., Ocskay R., Raes N. & Maenhaut W. 2005. Fine structure of mass size distributions in urban environment. Atmos. Environ. 39: 5363–5374.
- Salma I. & Maenhaut W. 2006. Changes in chemical composition and mass of atmospheric aerosol pollution between 1996 and 2002 in a central European city. *Environ. Pollut.* 143: 479–488.
- Salma I., Borsós T., Weidinger T., Aalto P., Hussein T., Dal Maso M. & Kulmala M. 2011. Production, growth and properties of ultrafine atmospheric aerosol particles in an urban environment. *Atmos. Chem. Phys.* 11: 1339–1353.
- Shi J.P. & Harrison R.M. 1999. Investigation of ultrafine particle formation during diesel exhaust dilution. *Environ*. *Sci. Technol.* 33: 3730–3736.
- Svartengren M., Strand V., Bylin G., Järup L. & Pershagen, G. 2000. Short-term exposure to air pollution in a road tunnel enhances the asthmatic response to allergen. *Eur. Respir. J.* 15: 716–724.
- Westerdahl D., Fruin S., Sax T. & Fine, P.M. 2005. Mobile platform measurements of ultrafine particles and associated pollutant concentrations on freeways and residential streets in Los Angeles. *Atmos. Environ.* 39: 3597–3610.
- Wolfenbarger J.K. & Seinfeld J.H. 1990. Inversion of aerosol size distribution data. J. Aerosol Sci. 21: 227–247.
- Zhu Y., Eiguren-Fernandez A., Hinds W.C. & Miguel A.H. 2007. In-cabin commuter exposure to ultrafine particles on Los Angeles freeways. *Environ. Sci. Technol.* 41: 2138–2145.