Science of the Total Environment 433 (2012) 418–426

Contents lists available at SciVerse ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Comparison of particulate number concentrations in three Central European capital cities

T. Borsós [a,*], D. Řimnáčová [b], V. Ždímal [b], J. Smolík [b], Z. Wagner [c], T. Weidinger [d], J. Burkart [e], G. Steiner [e], G. Reischl [e], R. Hitzenberger [e], J. Schwarz [b], I. Salma [a]

[a] Institute of Chemistry, Eötvös University, Budapest, Hungary
[b] Department of Aerosols and Laser Studies, Institute of Chemical Process Fundamentals, Prague, Czech Republic
[c] E. Hála Laboratory of Thermodynamics, Institute of Chemical Process Fundamentals, Prague, Czech Republic
[d] Department of Meteorology, Eötvös University, Budapest, Hungary
[e] Faculty of Physics, University of Vienna, Vienna, Austria

ARTICLE INFO

Article history:
Received 27 March 2012
Received in revised form 3 June 2012
Accepted 13 June 2012
Available online xxxx

Keywords:
Urban environment
Ultrafine aerosol
Diurnal variation
Number size distribution
New particle formation

ABSTRACT

Number size distributions of atmospheric aerosol particles in the mobility diameter range from 10 to 1000 nm were determined in Budapest, Prague and Vienna for a one-year-long period. Particle number concentrations in various size fractions, their diurnal and seasonal variations, mean size distributions and some properties of new particle formation events were derived and compared. Yearly median particle number concentrations for Budapest, Prague and Vienna were 10.6×10^3, 7.3×10^3 and 8.0×10^3 cm^{-3}. Differences were linked to the different pollution levels of the cities, and to diverse measurement environments and local conditions. Mean contributions of ultrafine particles (particles with a mobility diameter <100 nm) to the total number concentration were 80%, 84% and 74% for Budapest, Prague and Vienna, thus these particles represent an overwhelming share of all particles in each city. Seasonal variation of particle number concentrations was not obvious. Diurnal variations of particles with a diameter between 100 and 1000 nm ($N_{100-1000}$) exhibited similar shape for the cities, which was related to the time-activity pattern of inhabitants and regional influences. The structure of the diurnal variation for ultrafine particles was also similar. It contained a huge morning peak in each city which was explained by emissions from vehicular traffic. The second peak was shifted from afternoon rush hours to late evenings as a result of the daily cycling in meteorological parameters. The character of the measurement site also influenced the diurnal variation. Diurnal variation of the mean ratio of ultrafine particles to $N_{100-1000}$ clearly revealed the presence and importance of new particle formation and subsequent growth in urban environments. Nucleation frequencies in Budapest and Prague were 27% and 23%, respectively on a yearly time scale. They showed a minimum in winter for both places, while the largest nucleation activity was observed in spring for Budapest, and in summer for Prague.
© 2012 Elsevier B.V. All rights reserved.

1. Introduction

It is increasingly recognized that aerosol particle number concentration can have significant implications on public health in addition to mass concentrations of particulate matter (Wichmann and Peters, 2000; Morawska, 2010; JRC-EASAC 64792, 2011). Aerosol particles are present in large numbers in polluted urban air. Particles with a diameter smaller than 100 nm dominate the number concentrations. These are called ultrafine particles. They represent excess health risks relative to fine (d<2.5 μm) or coarse particles (d>2.5 μm) of the same or similar chemical composition (Li et al., 2003; Nel et al., 2006). Clearance mechanisms of the respiratory system are able to remove huge numbers of deposited ultrafine particles only with limitations. In addition, such particles can enter directly the bloodstream or the cellular interstitial fluid from the lung, and can be transported to various sensitive organs such as the heart or the central nervous system (Oberdörster et al., 2005). They can also change some properties and functions of the blood. Several research studies were devoted to particle number concentrations in cities or various urban environments (e.g., Ruuskanen et al., 2001; Wehner and Wiedensohler, 2003; Bukowiecki et al., 2003; Hussein et al., 2004; Mönkkönen, 2005; Stolzenburg et al., 2005; Jeong et al., 2006; Moore et al., 2007; Qian et al., 2007; Putaud et al., 2010; Salma et al., 2011a, 2011b). Traffic was found to be the most important source of ultrafine particles (Shi et al., 1999; Morawska et al., 2008). Biomass burning from local combustion sources, and atmospheric nucleation contribute substantially. Diurnal variations of the intensities of these sources, particularly in road traffic, exhibit analogies for large cities, and, therefore, important similarities can be expected with regard to the ultrafine particles. Particle number concentrations are influenced by regional atmospheric and meteorological phenomena as well. For these reasons, it

* Corresponding author. Tel.: +36 202424525.
E-mail address: bteebi@gmail.com (T. Borsós).

0048-9697/$ – see front matter © 2012 Elsevier B.V. All rights reserved.
doi:10.1016/j.scitotenv.2012.06.052

seems sensible to compare different properties of particle number concentrations for cities that belong to the same region, type of climate, and are of comparable size.

Budapest, Prague and Vienna are capital cities of Hungary, Czech Republic and Austria. Populations of Budapest, Prague and Vienna are 1.7 million, 1.3 million and 1.7 million inhabitants, respectively. The location of the cities is shown in Fig. 1. They are situated in Central Europe in a relatively narrow band oriented in parallel with one of the prevailing wind directions that is NW in the region. The distance between Prague and Vienna is 250 km, 220 km between Vienna and Budapest, and 440 km between Prague and Budapest. Only unleaded petrol is used for road vehicles in the countries, and diesel fuel marketed for road vehicles contains sulphur in a concentration <10 ppm, according to EU specification. Geographical, meteorological and other conditions facilitate the comparison of average particle number concentrations for various size fractions, their typical time variation, mean particle number size distributions, and some properties of new particle formation events. The objective of this study was to investigate these parameters for a one-year-long time period in order to reveal differences and similarities, as well as common or specific features.

2. Measurement sites

The measurements in Budapest were performed at the campus of the Eötvös University in Lágymányos on a second floor balcony, ca. 10 m above ground. The site (47° 28′ 29″ N, 19° 03′ 43″ E, 115 m above mean sea level, amsl) is located near the central part of the city, at a distance of 80 m from the bank of the river Danube, and is influenced by the wind channel formed above the river (Salma et al., 2011a). The major pollution sources include vehicular road traffic and residential heating in the Budapest agglomeration area. Long-range transport of some pollutants also plays a role (Salma et al. 2001, 2004; Salma and Maenhaut, 2006). The contributions of passenger cars and buses to the total 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.

The measurements in Prague were performed at the campus of the Institute of Chemical Process Fundamentals in Suchdol, at the roof level, 5 m above ground. The site (50° 7′ 35″ N, 14° 23′ 5″ E, 277 m amsl) is located in a suburban residential area in the NW of Prague, and is slightly elevated above the city. Air masses usually arrive from the countryside. Stationary sources (local coal-based heating, heating plants, etc.) are spread throughout the locality (Řimnáčová et al., 2011). Automotive sources also contribute to the aerosol levels. Passenger cars and buses make up 88% and 0.40%, respectively, of the vehicles (excluding motorcycles) registered in Prague. Diesel-powered vehicles accounted for 27% of all passenger cars, while 83% of buses ran on diesel.

Fig. 1. Location of the measurement sites in Central Europe.

The measurements in Vienna were performed at the Physics Building of the University of Vienna. The site (48° 13′ 18″ N, 16° 21′ 23″ E, 190 m amsl) is situated in the central part of the city, at a distance of 100 m from the nearest road. It is, however, separated from the road traffic by interconnected buildings and courtyards, and can, therefore, be regarded as an urban background site (Hitzenberger et al., 2006). The instruments were set up in a roof laboratory at a height of approximately 35 m above ground level. In 2009, passenger cars and buses contributed 73% and 0.2% to the total Austrian vehicle fleet, and 55% of passenger cars had diesel engines, while municipal buses ran on LPG. No data are available on the share of diesel and LPG powered buses (Statistik Austria, 2012).

3. Measurement methods and time intervals

The measurement system in Budapest included a flow-switching type differential mobility particle sizer (DMPS) and a meteorological station. The DMPS consists of a Nafion membrane drier, a 28-cm long differential mobility analyser (DMA) and a butanol-based condensation particle counter (CPC, TSI model 3775, USA) (Aalto et al., 2001; Salma et al., 2011a). The system operates at two sets of flows. In the first flow mode, the sample air and sheath air flows are 2 and 20 L min^{-1}, respectively, while in the second flow mode, these rates are 0.3 and 3 L min^{-1}, respectively. Particle number concentrations are recorded in 30 size channels, and the covered particle diameter range is from 6 to 1000 nm. In the first flow mode, 20 channels are measured (from 6 to 200 nm), while in the second flow mode, data from 10 channels are acquired (from 200 to 1000 nm). For the purpose of this comparative study, a size interval from 10 to 1000 nm was utilised. The measurements were performed with a time resolution of approximately 10 min. The sampling line was a Cu tubing, with an internal diameter of 4 mm and length of approximately 1.5 m. There was no upper size cut-off inlet applied to the sampling line. Basic meteorological data were obtained from the Urban Climatological Station of the Hungarian Meteorological Service operated within the university campus.

The measurements in Prague were based on a scanning mobility particle sizer (SMPS, TSI model 3034, USA) and an aerodynamic particle sizer (APS, TSI model 3321, USA). Both spectrometers sampled synchronously through a common TSP inlet (MiniPM inlet, BGI, USA) with a flow rate of 6 L min^{-1}. The SMPS sampled with a flow rate of 1 L min^{-1} isokinetically from the main 2-metre-long vertical tubing, which provided a flow of another 5 L min^{-1} to the APS. Samples were taken at an ambient relative humidity without additional conditioning. The SMPS contains a DMA (TSI model LDMA 3081, USA) and a build-in butanol-based CPC (TSI model CPC 3010, USA) in a single cabinet. The SMPS was operated at a closed-loop sheath flow rate of 4 L min^{-1} and recorded particles over a range of mobility diameters from 10 to ca. 500 nm, resolved into 54 channels. The APS measures particles in the aerodynamic diameter (AD) range from 0.5 to 20 μm. In order to combine the data of both spectrometers into a single size distribution, the AD data of the APS were converted into mobility diameters using particle density of 1500 kg m^{-3} and including the slip correction factor (Khlystov et al., 2004). For the purpose of this comparative study, a size interval from 10 to 1000 nm was utilised. The time resolution of the measurements was 5 min. Meteorological data were obtained from an Automated Imission Monitoring station of the Czech Hydrometeorological Institute operated within the campus.

The measurement system used in Vienna consisted of a SMPS consisting of a Vienna-type DMA operated in a closed-loop arrangement (Jokinen and Mäkelä, 1997; Giebl et al., 2002; Burkart et al., 2011), and a butanol-based CPC (TSI model 3760A, USA). It operates at an aerosol flow rate of 1.5 L min^{-1} and a sheath air flow rate of 7.5 L min^{-1}. Particle concentrations were determined from 10 to 926 nm in 87 channels with a time resolution of 10 min. The sampling line had an internal diameter of 6 mm and a length of approximately 0.5 m. No drier was used in the sample flow. There was no upper size cut-off inlet applied

to the sampling line. Meteorological data were obtained from the station of the Austrian Central Institute for Meteorology and Geodynamics located ca. 1.5 km N of the measurement site.

Daily mean concentration data for PM_{10} mass and O_3 were also included in the comparison. They were adopted from the public data sets of the official municipal air quality monitoring stations closest to the measurement sites. The PM_{10} mass data were obtained by the beta-radiation absorption method, and the O_3 concentrations were determined by the chemo-luminescence method in all cities.

Data sets collected over the period from 3 November 2008 to 2 November 2009 in Budapest and Prague were evaluated. In Vienna, the data sets available for the comparison were obtained one year earlier than for the other two sites, thus from November 2007 to October 2008. However, the one-year-long time span of the data sets enabled us to perform the comparisons.

The data compared could be subject to some systematic effects due to differences in experimental methods and instrumentation. Particle number concentrations and size distributions were measured with DMPS or SMPS systems. Their principle of operation is practically identical. The sampling losses were estimated to be negligible compared to the differences caused by site and meteorological differences. The ability of the DMPS to determine the total number of particles accurately was tested with another CPC operated in parallel, and the mean DMPS-to-CPC concentration ratio was within the ±10% uncertainty interval specified for the CPCs by the manufacturer. Similar differences for counting particles with a diameter >300 nm were observed during an intercomparison field test (Putaud et al., 2010), and consistency in the number concentration data was estimated to be approximately ±15%. For the PM_{10} mass and O_3 concentrations, the systematic experimental differences between the cities are expected to be smaller than this due to the same type of instrumentation used. As far as sizing of particles is concerned, larger uncertainties were experienced for the diameter range >300 nm (Van Dingenen et al., 2004). More importantly, the measurements in Budapest were accomplished under controlled humidity conditions (relative humidity, RH<20%), while the instruments used in Prague and Vienna were operated at ambient RHs. While this is not a problem in winter, when the sample is heated up in the lines and dried, it may lead to a substantial increase of the measured particle size in summer, because a warmer and humid sample is led to a colder space of the measuring container causing an increase of relative humidity of the sample followed by condensation of water vapour on the surface of the hygroscopic aerosol particles. Therefore, the direct comparison of the size distributions for the cities via modal diameters or geometric standard deviations was avoided, and only overall shapes or modal structures were examined instead.

4. Results and discussion

4.1. Meteorology

Mean meteorological parameters at the three measurement sites for the four seasons are summarised in Table 1. The mean values of certain parameters for the cities generally vary within a factor of 1.5, while the air temperature and, in particular, RH values were considerably closer to each other. The parameters tend to vary monotonically with the geographical latitude. The microclimate of the cities and meso-scale meteorology could also play a modifying role. As far as wind speed (WS) is concerned, no tendency was found. The highest mean WS was found in Vienna. This can probably be explained by the location of the meteorological station at the edge of the Alps and the effect of the Danube (the "Wiener Pforte"). In contrast, the lowest mean WS was observed in Prague where the instrumentation was set up in a park located on a hill-side. The higher mean temperature for winter in Vienna can be linked to the warmer winter in Central Europe in 2007/08 with respect to 2008/09. Mean meteorological parameters were in accordance with geographically typical values, and they lay within a reasonably small interval. It all suggests that the comparison of the three measurement sites is feasible and can supply valuable information on the cities.

Table 1
Mean meteorological parameters at the measurement sites for the four seasons. The data sets for Vienna were obtained from Luftmessnetz Wien.

Parameter [unit]	Winter	Spring	Summer	Autumn
Global radiation [W m^{-2}]				
Prague	37	167	174	75
Vienna	42	177	225	85
Budapest	55	262	261	101
Temperature [°C]				
Prague	−0.3	9.2	19	9.9
Vienna	3.2	11	20	11
Budapest	1.6	14	22	13
Relative humidity [%]				
Prague	82	68	69	78
Vienna	78	67	68	77
Budapest	75	52	57	70
Wind speed [m s^{-1}]				
Prague	2.1	2.3	1.4	1.7
Vienna	3.8	3.6	3.1	3.0
Budapest	3.0	3.2	2.9	2.7

4.2. Atmospheric concentrations

Ranges and medians of the daily atmospheric concentrations of all aerosol particles (*N*), ultrafine particles (*UF*), PM_{10} mass and ozone in Budapest, Prague and Vienna for a one year period are summarised in Table 2. The site in Budapest exhibited the largest median aerosol concentrations, while the Prague site showed the lowest values. The ratios of Budapest to Prague, Budapest to Vienna, and Vienna to Prague for the median total particle number concentrations were 1.45, 1.33 and 1.10, respectively, while for the median ultrafine particle number concentrations, these values were 1.40, 1.42 and 0.98, respectively. The corresponding ratios for the PM_{10} mass were 1.45, 1.45 and 1.00, respectively. The particle number and mass concentrations followed the same trend. This can be explained by the fact that their common major emission source is associated with the road traffic in all three cities. The relationship is, however, not simple because different mechanisms can be responsible for the production of ultrafine particles (which control the total particle number concentration) and of larger (>100 nm) particles (which control the mass concentration), and the mechanisms do not necessarily co-vary in space and time (Harrison and Jones, 2005). The differences in the median aerosol concentration data can also be linked to both different pollution levels of the cities, and to the fact that the actual character of the measurement environments was somewhat diverse. The measurements in Budapest were performed near the city centre in a larger open area, while the measurement site in Prague was situated in an open suburban location in the upwind direction of the city centre. The measurement site in Vienna was close to the city centre but separated

Table 2
Medians and their range of daily atmospheric concentrations for particle numbers in the diameter ranges of 10–1000 nm (*N*) and 10–100 nm (*UF*), PM_{10} mass, and ozone. The data sets for Vienna were obtained from Luftmessnetz Wien.

Measurement site		$N\times10^{-3}$ [cm^{-3}]	$UF\times10^{-3}$ [cm^{-3}]	PM_{10} mass [μg m^{-3}]	O_3 [μg m^{-3}]
Budapest	Maximum	28	18.8	148	89
	Median	10.6	8.4	29	34
	Minimum	3.0	2.2	8	3
Prague	Maximum	48	48	179	111
	Median	7.3	6.0	20	53
	Minimum	1.62	1.28	4	2
Vienna	Maximum	19.3	14.7	85	114
	Median	8.0	5.9	20	53
	Minimum	1.64	1.24	5	2

from the road traffic by buildings, and the measurement was performed at an elevated height. Despite the southernmost location of Budapest, the median concentration of O_3 was smaller than for the other two sites by a factor of 0.64 each. A simple explanation is that tropospheric O_3 concentrations in moderately polluted cities are known to be related inversely to general air quality on long time scales. It has to be noted that all three sites belong to the relatively clean urban areas of Europe as far as particle number concentrations are concerned. Daily mean values up to 50×10^3 cm^{-3} were observed at many European urban sites (Aalto et al., 2005; Putaud et al., 2010).

The mean contribution (and its standard deviation) of ultrafine particles to the total number of particles (*UF*/*N* ratio) was (80 ± 8)% for Budapest, (84 ± 10)% for Prague and (74 ± 9)% for Vienna. Strong association between these two size fractions is also evident from their large correlation coefficients which were larger than 0.90 for all sites. The yearly mean *UF*/*N* ratios are similar to each other. This is likely caused by the comparability of the major source types and formation processes in the cities, and by the similarities in the accumulation-mode particle number concentrations in the larger region of Central Europe. The contributions of the *UF*/*N* ratios are close to the overall European mean ratio of 0.76 (Putaud et al., 2010), and are comparable to, but somewhat smaller than the range of 88–94% reported for some (central) urban environments (Jeong et al., 2006).

Ranges of the daily median concentrations for the Prague site seem to be considerably wider than for the other two sites, except for the ultrafine particle number concentrations. The maxima are larger, and the minima are smaller in Prague. The larger variability can be explained by the actual (suburban) location of the measurement site upwind of the central part of the city. This makes the site more sensitive to the changes in the wind direction. The smallest concentrations occurred when the wind blew from the countryside (westerly winds), while the largest number concentrations occurred when the air masses arrived from the city centre. The larger maxima can also be associated with the vicinity and intermittent intensity of some fixed local pollution sources such as residential heating. The other two sites are located near the city centre which makes the dependency on the wind direction less pronounced.

4.3. Diurnal and seasonal variations

For the three cities, the diurnal variations of the mean number concentration of ultrafine particles and particles with a diameter of 100–1000 nm ($N_{100-1000}$), averaged by the time of day are shown in Fig. 2a and b, respectively. The latter size range is utilised to approximate the accumulation size fraction. Atmospheric residence times for most ultrafine particles (more exactly, for the Aitken-mode particles) in the boundary layer in cities are relatively short ($<10^0$ h), and therefore, their presence in the air can be directly associated with their local emission sources or formation mechanisms. At the same time, accumulation mode particles have much longer residence times (up to 10^1 days) in the boundary layer, so they represent a larger territory.

The diurnal curves in Fig. 2a resemble the time-activity pattern of inhabitants, particularly in the pattern of road traffic flow. These curves contain two peaks. The morning peak reaches its maximum between 7:00 and 9:00 local time, and is associated with the road traffic emissions during rush hours. It is relatively narrow and its time evolution closely follows the vehicle intensity at each site. This also means that the meteorological conditions influencing the particle concentrations change with a smaller rate and less over the time interval of interest than road traffic. The peak for Budapest is substantially larger than for the other two sites as the result of the generally larger influence of traffic there.

The second peak does not appear during the afternoon rush hours (usually between 16:30 and 18:30) but shows up in the late evening between 21:00 and 22:00 instead in all three cities. The morning and evening peaks are separated by a broad (local) minimum in Vienna and Budapest, while the concentrations between the two peaks remained elevated in Prague throughout the daytime. This can be explained by the differences in the character of the measurement sites. The shift of the second peak into the late evening hours can be mainly explained by the effect of meteorology. In particular, daily cycling of the boundary layer mixing height, and of the intensity of mixing, which are strong under anti-cyclonic conditions, decrease the concentration levels in the afternoons, and this effect can compensate the increased contribution of direct emissions until sunset. The effect of burning and heating activities at residences and homes can also play a role. The derived diurnal behaviour is consistent with the time variations observed in other European cities (Aalto et al., 2005; Moore et al., 2007; Avino et al., 2011). It was demonstrated earlier that new particle formation (which usually occurs around midday) also contributes substantially to the ultrafine particle number concentrations in cities (e.g., Salma et al., 2011a, Fig. 3). The effect of nucleation on the mean particle concentration is, however, not evident in Fig. 2a. It is explained by a relatively large contribution of direct emissions in the cities (see Section 4.4), which masks the contribution of nucleation events.

It is seen in Fig. 2b that the diurnal variations of $N_{100-1000}$ also followed a periodic pattern over the days, and that they have a rather similar shape for the cities. They consist of a morning and an evening peak with a deep and broad local minimum between them. The extent of the change was larger for Budapest and Vienna, and smaller for Prague, where the station is more influenced by background air masses.

Diurnal variations of the mean ratio of ultrafine particle concentrations to $N_{100-1000}$, averaged by the time of day for the three cities are shown in Fig. 3. The curves imply that the concentrations of ultrafine particles were larger than the approximated regional concentrations ($N_{100-1000}$) by mean factors of 2.4, 3.6 and 4.4 for Vienna, Budapest and Prague, respectively. The values are consistent with the *UF*/*N* ratios discussed in Section 4.2. The curves show a clear absolute minimum around 4:00, and they also contain the morning and evening peaks that can also be seen in Fig. 2a and b. In addition, a third peak appeared between 12:00 and 16:00 in all cities. For the Prague and Budapest sites, the peak was huge and broad, while for the Vienna site, it was much smaller but still visible. This peak was caused by several effects. During the nucleation days (see later), the concentration levels of ultrafine particles were increased substantially over the midday, while the concentration of the accumulation fraction was relatively small and more or less constant over this time interval. As a result, the corresponding individual $UF/N_{100-1000}$ ratios became larger, and caused a significant increase in the overall mean ratio. The relationship between nucleation events and the third (midday) peak was also confirmed by deriving the $UF/N_{100-1000}$ concentration ratios separately for days with obvious nucleation and for all days. The mean ratios for the nucleation days were larger by factors of 2.3 and 2.5 for Budapest and Prague, respectively, than the ratios obtained for all days. The explanation of the midday peak suggests in an indirect way that atmospheric nucleation may be more intense and/or frequent in Budapest and Prague than in Vienna.

Variations of the monthly median total number concentrations for the three cities are shown in Fig. 4. For Vienna, months for which a considerable share of the measured data (approximately >20%) was missing were omitted from the evaluation. It is worth noting that there was no evident seasonal dependency in the particle number concentration for Budapest (Salma et al., 2011a) and Prague, and that is likely valid for Vienna as well (seasonal patterns of total particle concentrations are given by Burkart et al., 2011). A general tendency for less pronounced seasonal variation in the PM mass has also been observed for the involved cities. The lack of obvious seasonal patterns in number concentrations can be linked to 1) improved combustion technologies, 2) to the fact that their major emission

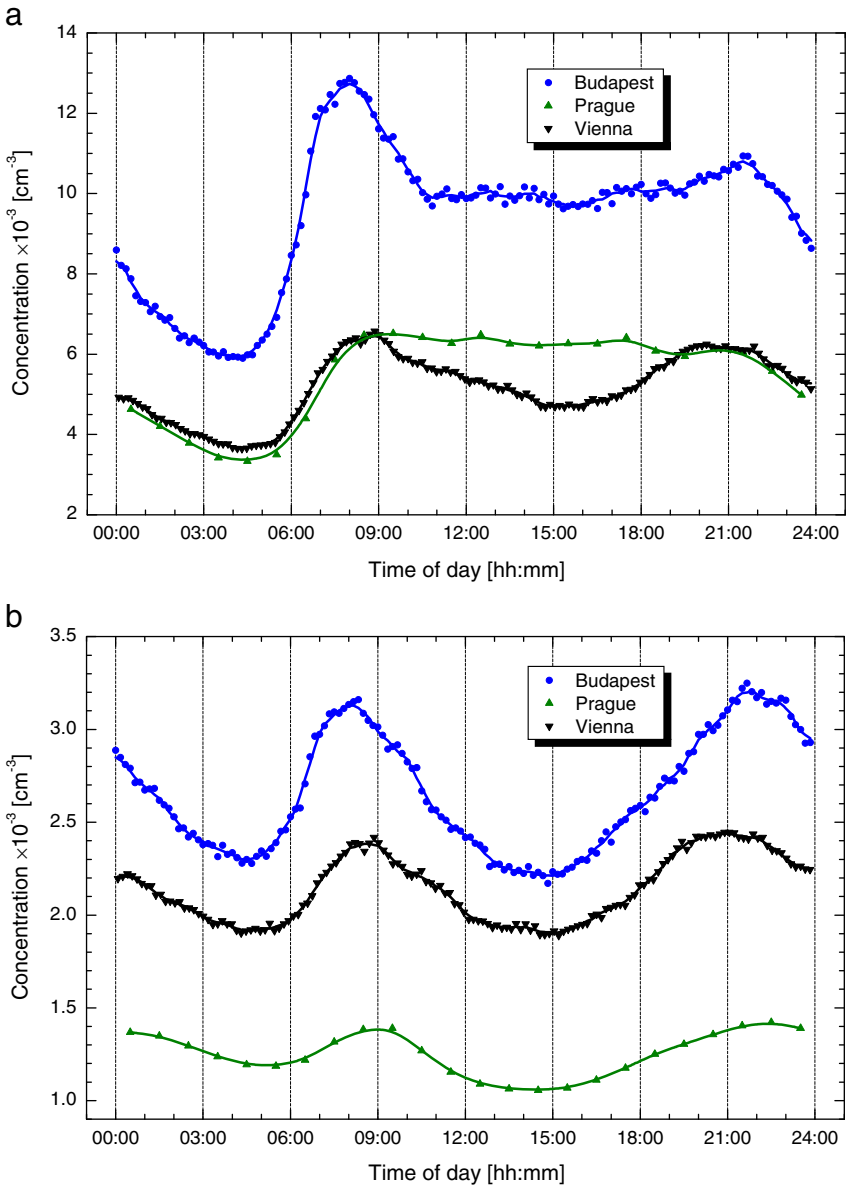

Fig. 2. Diurnal variation of mean ultrafine particle number concentration (a) and of particles with a diameter between 100 and 1000 nm (b) for Budapest, Prague and Vienna.

source (road traffic) is distributed evenly throughout the year, and 3) to the smaller atmospheric residence time of ultrafine particles. For some other European cities, marked seasonal dependency was recorded (Aalto et al., 2005).

4.4. Number size distributions and new particle formation events

Monthly mean number size distributions of aerosol particles in the diameter range of 10–1000 nm for the three measurement sites are shown in Fig. 5 for January and Fig. 6 for July. It is evident that the number size distributions are rather dynamic. They reflect continual emission, formation, transformation and transport processes. The averaged distributions obtained were, therefore, substantially broadened. The shape of the size distributions for Budapest was similar for all months. They contained a broad peak which could be usually deconvoluted into Aitken and accumulation modes of similar areas. The presence of separate Aitken and accumulation modes in the size distributions was more evident for Prague and Vienna. The shape of the size distributions for these cities changed from month to month more markedly than for Budapest. The relative contribution of the two modes had no clear tendency. The exact positions of the modes are subject to systematic effects (as discussed in Section 3), therefore, their comparison was not performed.

The measurement days were classified into four groups: 1) days with evident new particle formation (NPF) event, 2) with obviously no event, 3) undefined days, and 4) days with missing data (missing data for >4 h a day). The classification was performed according to a modified scheme of Dal Maso et al. (2005). A statistical overview on the groups is given in Table 3 for Budapest and Prague. There was no such information available for Vienna because of the considerable number of missing data. New particle formation occurred on 83 days in Budapest and 74 days in Prague during the campaign, which represent 27% and 23% of the relevant days, respectively. The NPF frequency exhibited a remarkable seasonal variation for both cities as it can be seen in Fig. 7. For Budapest, the frequencies were 7.3% and 44% for winter and spring, while it was 28% and 29% for summer and autumn, respectively. For Prague, the frequencies for winter, spring, summer and autumn were 3.9%, 34%, 41% and 10%, respectively. Seasonal variation of new particle formation is known to be related to local features, e.g., to solar radiation, air temperature, relative humidity,

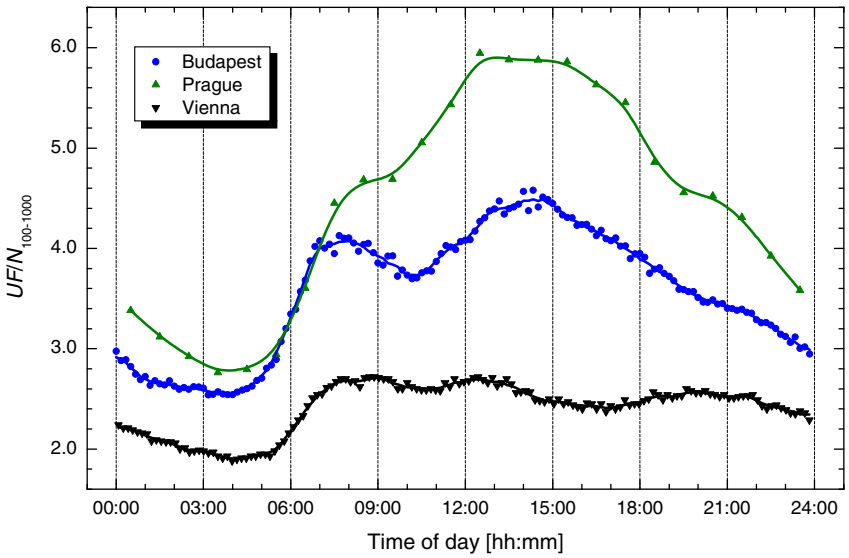

Fig. 3. Diurnal variation of the mean ratio of ultrafine particle concentration to particles with a diameter of 100–1000 nm.

biogenic emissions, local wind speed, height of the boundary mixing layer, as well as concentration and size distribution of the atmospheric aerosol particles (Väkevä et al., 2000; Kulmala et al., 2004; Kiendler-Scharr et al., 2009). Nucleation activity may also be associated with some limiting or triggering pre-existing processes which can depend on the actual region of the cities. Particle growth rates were smaller in Prague than in Budapest. For Prague, the growth rate ranged from 0.1 to 15.0 nm h^{-1} with a mean and standard deviation of (4.3 ± 2.8) nm h^{-1}. The range for Budapest is similar, from 2.0 to 13.3 nm h^{-1}, but the mean value of (7.7 ± 2.4) nm h^{-1} is significantly larger than for Prague. The difference can be linked to the different pollution levels and somewhat different character of the measurement sites (urban background in Budapest and suburban background in Prague).

5. Summary and conclusions

Number size distributions of aerosol particles were measured in urban environments in Budapest, Prague and Vienna continuously for one year. Atmospheric concentrations for all particles and ultrafine particles, and some other derived properties were determined and compared. The largest median number concentration occurred in Budapest, while Prague had the smallest levels. The difference was linked to the different pollution levels of the cities and to the diverse measurement environments. The average concentrations belong to the lower half of concentration levels typical for other European cities. Contribution of ultrafine particles to the total number concentrations was similar for the three cities. Primary ultrafine particles have major relevance for the air quality and emission source apportionment, and they are associated with local sources of point or diffusive character. Nucleation, however, usually happens on larger spatial scales than these production types, and it influences the particle budget significantly. Ultrafine particles dominated the atmospheric concentrations. Diurnal variations of the number concentration had similar pattern, showing three peaks in general: a morning, a midday and an evening peak. The morning peak was caused (i.e., created) by fresh vehicular emissions during the morning rush hours. Particle concentrations usually decreased in the early afternoon due to reduced

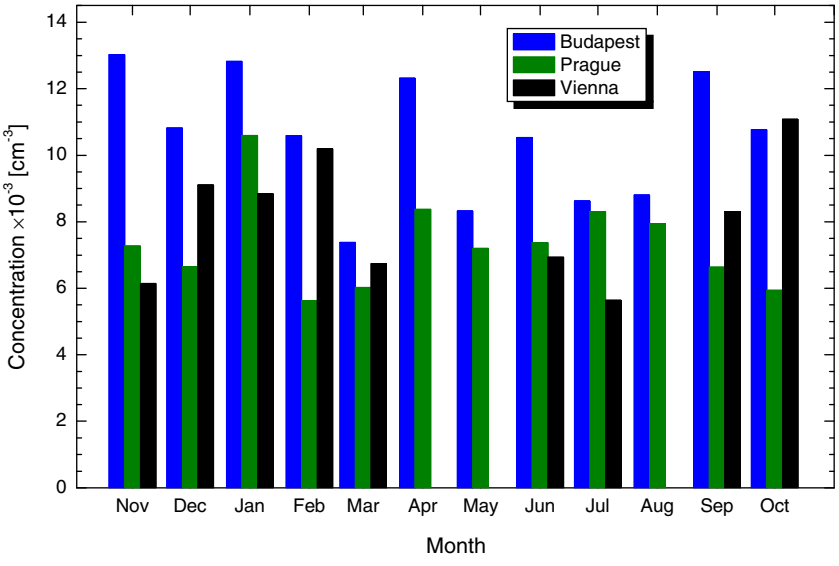

Fig. 4. Variation of monthly median particle number concentrations ($N_{10-1000}$).

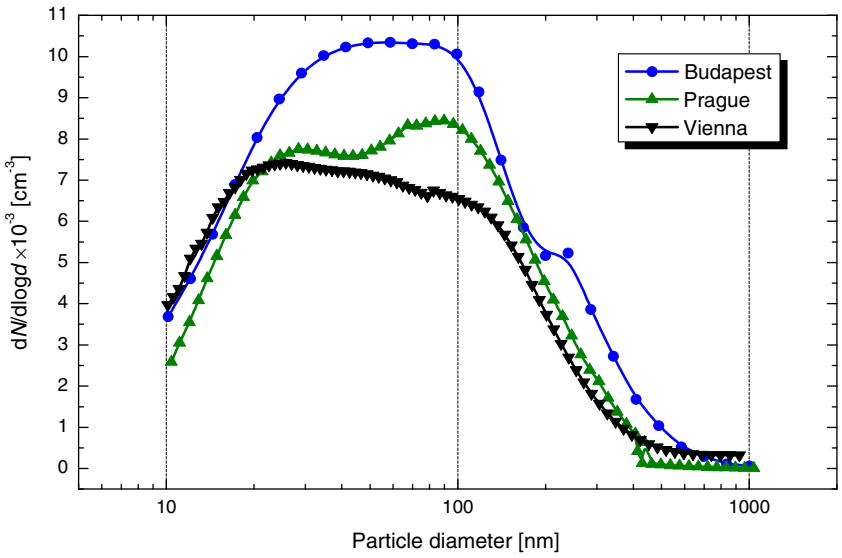

Fig. 5. Monthly mean number size distribution of aerosol particles for January.

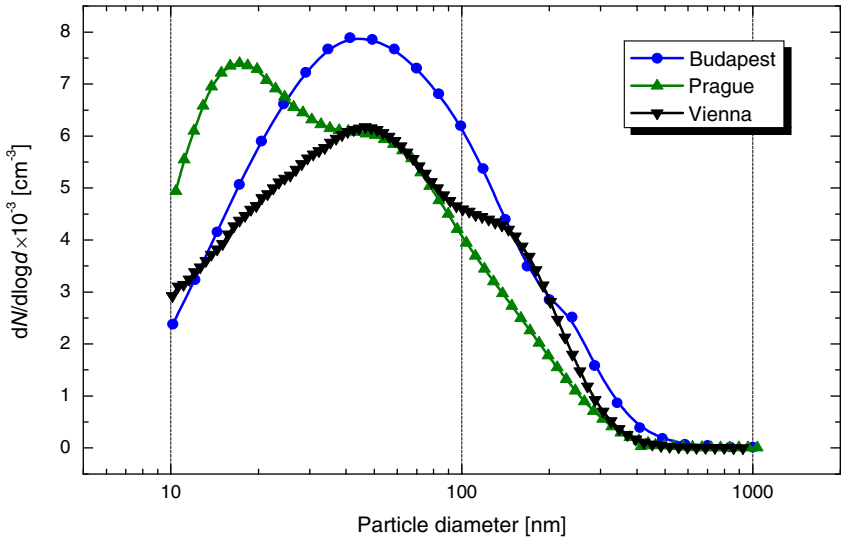

Fig. 6. Monthly mean number size distribution of aerosol particles for July.

emission strength and increased mixing height. The evening peak was also related to traffic emissions but its location was considerably shifted from afternoon rush hours to late evening hours by specific meteorological effects. The midday peak was explained by atmospheric nucleation. Nucleation frequencies exhibited seasonal variation, and had basically similar tendencies for Budapest and Prague. At the same time, no obvious seasonal dependence was observed for the cities regarding daily mean number concentrations, which is a new feature of the cities. The combined or alternating effect of direct emissions and nucleation could contribute to this independency.

Acknowledgements

Financial support by a bilateral project between Hungary and Czech Republic, by the Hungarian Scientific Research Fund (K84091), by the Czech Science Foundation (CSF P209/11/1342), and by the Austrian

Table 3
Number of all days, days with evident new particle formation event, with obviously no event, with missing data (missing data for >4 h a day), and undefined days over the four seasons in Budapest (Bp) and Prague (Pr).

	Winter		Spring		Summer		Autumn	
	Bp	Pr	Bp	Pr	Bp	Pr	Bp	Pr
Total number of days	90		92		92		91	
Number of days with new particle formation	6	3	34	29	21	34	22	7
Number of days without new particle formation	76	74	44	58	54	49	55	64
Number of undefined days	5	0	12	5	7	2	10	2
Number of days with missing data	3	13	2	0	10	7	4	18

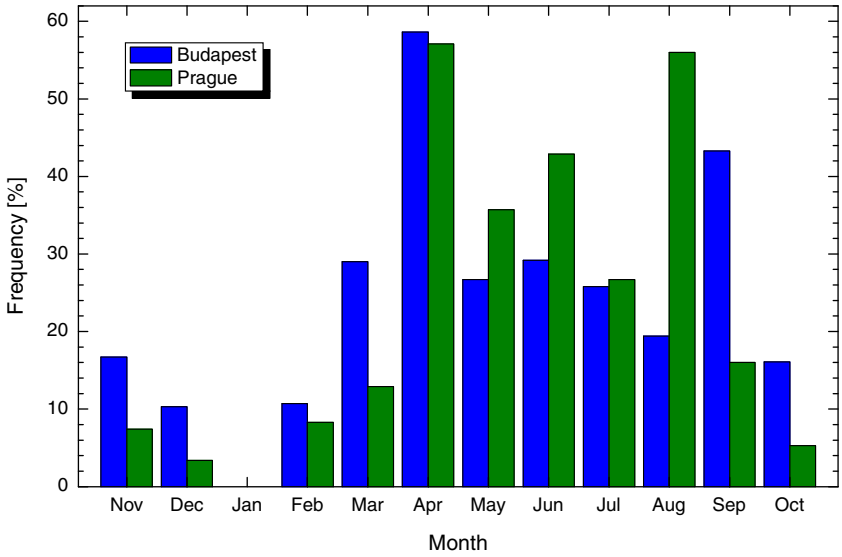

Fig. 7. Monthly mean new particle formation frequencies in Budapest and Prague.

Science Fund (FWF, P19515-N20) is appreciated. The authors thank J. Šilhavý from the Czech Hydrometeorological Institute for providing the meteorological data, the Viennese Environmental Department for providing the data from the air quality monitoring network, and the Austrian Central Institute for Meteorology and Geodynamics (ZAMG) for providing the meteorological data. We thank D. Neubauer of the University of Vienna for the help with the data analysis.

References

Aalto P, Hämeri K, Becker E, Weber R, Salm J, Mäkelä JM, et al. Physical characterization of aerosol particles during nucleation events. Tellus B 2001;53:344–58.
Aalto P, Hämeri K, Paatero P, Kulmala M, Bellander T, Berglind N, et al. Aerosol particle number concentration measurements in five European cities using TSI-3022 condensation particle counter over a three-year period during health effects of air pollution on susceptible subpopulations. J Air Waste Manag Assoc 2005;55:1064–76.
Avino P, Casciardi S, Fanizza C, Manigrasso M. Deep investigation of ultrafine particles in urban air. Aerosol Air Qual Res 2011;11:654–63.
Bukowiecki N, Dommen J, Prevot ASH, Weingartner E, Baltensperger U. Fine and ultrafine particles in the Zurich (Switzerland) area measured with a mobile laboratory: an assessment of the seasonal and regional variation throughout a year. Atmos Chem Phys 2003;3:1477–94.
Burkart JG, Steiner G Reischl, Hitzenberger R. Long-term study of cloud condensation nuclei (CCN) activation of the atmospheric aerosol in Vienna. Atmos Environ 2011;45:5751–9.
Dal Maso M, Kulmala M, Riipinen I, Wagner R, Hussein T, Aalto PP, et al. Formation and growth of fresh atmospheric aerosols: eight years of aerosol size distribution data from SMEAR II, Hyytiälä, Finland. Boreal Environ Res 2005;10:323–36.
Giebl H, Berner A, Reischl G, Puxbaum H, Kasper-Giebl A, Hitzenberger R. CCN activation of oxalic and malonic acid test aerosols with the University of Vienna cloud condensation nuclei counter. J Aerosol Sci 2002;33:1623–34.
Harrison RM, Jones AM. Multisite study of particulate number concentrations in urban air. Environ Sci Technol 2005;39:6063–70.
Hitzenberger R, Petzold A, Bauer H, Ctyroky P, Pouresmaeil P, Laskus L, et al. Intercomparison of thermal and optical measurement methods for elemental carbon and black carbon at an urban location. Environ Sci Technol 2006;40:6377–83.
Hussein T, Puustinen A, Aalto PP, Mäkelä JM, Hämeri K, Kulmala M. Urban aerosol number size distributions. Atmos Chem Phys 2004;4:391–411.
Jeong C-H, Evans GJ, Hopke PK, Chalupa D, Utell MJ. Influence of atmospheric dispersion and new particle formation events on ambient particle number concentration in Rochester, United States, and Toronto, Canada. J Air Waste Manag Assoc 2006;56:431–43.
Jokinen V, Mäkelä JM. Closed loop arrangement with critical orifice for DMA sheath/excess flow system. J Aerosol Sci 1997;28:643–8.
JRC-EASAC 24847 (European Commission, Joint Research Centre — European Academies Science Advisory Council). Impact of engineered nanoparticles on health: considerations for benefit — risk assessment, reference report 24847. Luxenbourg: Publications Office of the European Union; 2011.
Khlystov A, Stanier C, Pandis SN. An algorithm for combining electrical mobility and aerodynamic size distribution data when measuring ambient aerosol. Aerosol Sci Technol 2004;38(S1):229–38.
Kiendler-Scharr A, Wildt J, Dal Maso M, Hohaus T, Kleist E, Mentel TF, et al. New particle formation in forests inhibited by isoprene emissions. Nature 2009;461:381–4.
Kulmala M, Vehkamäki H, Petäjä T, Dal Maso M, Lauri A, Kerminen V-M, et al. Formation and growth rates of ultrafine atmospheric particles: a review of observations. J Aerosol Sci 2004;35:143–76.
Li N, Sioutas C, Cho A, Schmitz D, Misra C, Sempf J, et al. Ultrafine particulate pollutants induce oxidative stress and mitochondrial damage. Environ Health Perspect 2003;111:455–60.
Mönkkönen JP. Observation of urban aerosol in India. Report series in aerosol science University of Helsinki; 2005.
Moore KF, Ning Z, Ntziachristos L, Schauer JJ, Sioutas C. Daily variation in the properties of urban ultrafine aerosol — part I: physical characterization and volatility. Atmos Environ 2007;41:8633–46.
Morawska L. Airborne particles and health. Air Qual Clim Change 2010;44:13–5.
Morawska L, Ristovski Z, Jayaratne ER, Keogh DU, Ling X. Ambient nano and ultrafine particles from motor vehicle emissions: characteristics, ambient processing and implications on human exposure. Atmos Environ 2008;42:8113–38.
Nel A, Xia T, Madler L, Li N. Toxic potential of materials at the nanolevel. Science 2006;311:622–7.
Oberdörster G, Oberdörster E, Oberdörster J. Nanotoxicology: an emerging discipline evolving from studies of ultrafine particles. Environ Health Perspect 2005;113:823–39.
Putaud J-P, Van Dingenen R, Alastuey A, Bauer H, Birmili W, Cyrys J, et al. A European aerosol phenomenology — 3: physical and chemical characteristics of particulate matter from 60 rural, urban, and kerbside sites across Europe. Atmos Environ 2010;44:1308–20.
Qian S, Sakurai H, McMurry PH. Characteristics of regional nucleation events in urban East St. Louis. Atmos Environ 2007;41:4119–27.
Řimnáčová D, Ždímal V, Schwarz J, Smolík J, Řimnáč M. Atmospheric aerosols in suburb of Prague: the dynamics of particle size distributions. Atmos Res 2011;20:539–52.
Ruuskanen J, Tuch Th, Ten Brink H, Peters A, Khlystov A, Mirme A, et al. Concentrations of ultrafine, fine and PM2.5 particles in three European cities. Atmos Environ 2001;35:3729–38.
Salma I, Maenhaut W. Changes in chemical composition and mass of atmospheric aerosol pollution between 1996 and 2002 in a Central European city. Environ Pollut 2006;143:479–88.
Salma I, Maenhaut W, Zemplén-Papp É, Záray Gy. Comprehensive characterisation of atmospheric aerosols in Budapest: physicochemical properties of inorganic species. Atmos Environ 2001;35:4367–78.
Salma I, Chi X, Maenhaut W. Elemental and organic carbon in urban canyon and background environments in Budapest, Hungary. Atmos Environ 2004;38:27–36.
Salma I, Borsós T, Weidinger T, Aalto P, Hussein T, Dal Maso M, et al. Production, growth and properties of ultrafine atmospheric aerosol particles in an urban environment. Atmos Chem Phys 2011a;11:1339–53.
Salma I, Borsós T, Aalto PP, Kulmala M. Time-resolved number concentration and size distribution of aerosol particles in an urban road tunnel. Boreal Environ Res 2011b;16:262–72.
Shi JP, Khan AA, Harrison RM. Measurements of ultrafine particle concentration and size distribution in the urban atmosphere. Sci Total Environ 1999;235:51–64.
Statistik Austria, URL: www.statistik.at, last download in January 2012.
Stolzenburg MR, McMurry PH, Sakurai H, Smith JN, Mauldin III RL, Eisele FL, et al. Growth rates of freshly nucleated atmospheric particles in Atlanta. J Geophys Res 2005;110:D22S05. http://dx.doi.org/10.1029/2005JD005935.

Väkevä M, Hämeri K, Puhakka T, Nilsson E, Hohti K, Mäkelä J. Effects of meteorological processes on aerosol particle size distribution in an urban background area. J Geophys Res 2000;105:9807–21.
Van Dingenen R, Raes F, Putaud J-P, Baltensperger U, Charron A, Facchini M-C, et al. A European aerosol phenomenology—1: physical characteristics of particulate matter at kerbside, urban, rural and background sites in Europe. Atmos Environ 2004;38:2561–77.
Wehner B, Wiedensohler A. Long term measurements of submicrometer urban aerosols: statistical analysis for correlations with meteorological conditions and trace gases. Atmos Chem Phys 2003;3:867–79.
Wichmann HE, Peters A. Epidemiological evidence of the effects of ultrafine particle exposure. Philos Trans R Soc Lond A 2000;358:2751–69.