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Production, growth and properties of ultrafine atmospheric aerosol particles in an urban environment

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Number concentrations of atmospheric aerosol particles were measured by a flow-switching type differential mobility particle sizer in an electrical mobility diameter range of 6–1000 nm in 30 channels near central Budapest with a time resolution of 10 min continuously from 3 November 2008 to 2 November 2009. Daily median number concentrations of particles varied from 3.8×10^3 to $29 \times 10^3 \text{ cm}^{-3}$ with a yearly median of $11.8 \times 10^3 \text{ cm}^{-3}$. Contribution of ultrafine particles to the total particle number ranged from 58 to 92% with a mean ratio and standard deviation of $(79 \pm 6)\%$. Daily average number concentrations in various size fractions and contribution of ultrafine particles to the total particle number showed no seasonal dependency. Monthly mean number size distributions were similar to each other. Overall mean for the number median mobility diameter of the Aitken and accumulation modes were 26 and 93 nm, respectively, which are substantially smaller than for rural or background environments. The Aitken and accumulation modes contributed similarly to the total particle number concentrations at the actual measurement location. Median diameters of the Aitken and accumulation modes were shifted to larger values before nucleation started and over the growth process, which can be related to the presence of aged aerosol under the conditions that favour nucleation and growth. Particle concentrations were usually increased substantially after nucleations. Overall mean and standard deviation of the nucleation mode number concentrations were $(10.4 \pm 2.8) \times 10^3 \text{ cm}^{-3}$. Mean ratio and standard deviation of the nucleation mode number concentration to the total particle number concentration that was averaged for two hours just before the formation was detected was 2.3 ± 1.1 . Nucleation unambiguously occurred on 83 days, which represent 27% of all relevant days. Its frequency showed a remarkable seasonal variation with a minimum of 7.3% in winter and a maximum of 44% in spring. Formation rate of particles with a diameter of 6 nm varied between 1.65 and $12.5 \text{ cm}^{-3} \text{ s}^{-1}$ with a mean and standard deviation of $(4.2 \pm 2.5) \text{ cm}^{-3} \text{ s}^{-1}$. Seasonal dependency for the formation rate could not be identified. Growth curves of nucleated particles were usually su-

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perimposed on the characteristic diurnal pattern of road traffic direct emissions. The growth rate of the nucleation mode with a median diameter of 6 nm varied from 2.0 to 13.3 nm h⁻¹ with a mean and standard deviation of (7.7±2.4) nm h⁻¹. There was an indicative tendency for larger growth rates in summer and for smaller values in winter.

5 Several indirect evidences suggest that the nucleation events occurred at least over the whole city, and were of regional type.

1 Introduction

Ultrafine aerosol contains particles with an electrical mobility diameter smaller than 100 nm. Most of the time, their size distribution can be resolved into Aitken and accu-
10 mulation modes. Residence time for the Aitken mode particles in the planetary bound-
ary layer is relatively short. Therefore, their appearance and presence in the air in
increased concentrations (which is sometimes referred to as ultrafine particle event,
Woo et al., 2001; Park et al., 2008) can be directly related to their major sources. Ul-
trafine aerosol particles are either emitted directly from high-temperature processes or
15 they are formed in the air as secondary particles. Their major production types in urban
environments can be identified on the basis of: the size interval in which the increased
particle number concentration occurs; the correlation between the concentration of
their certain size fractions and concentrations of some atmospheric pollutants (e.g.,
CO, NO_x or SO₂) or traffic counts; and particle growth properties (Woo et al., 2001;
20 Jeong et al., 2006; Watson et al., 2006; Qian et al., 2007; Park et al., 2008). They
include 1) automotive road traffic emissions, for which the typical size interval where
the particles appear is ca. 20–100 nm, 2) residential heating emissions, for which the
appearance interval is ca. 50–100 nm, and 3) nucleation, for which the appearance
is at the lower limit of the aerosol size range of ca. 1 nm in diameter (or at the lower
25 size limit of the measurement system). Combustion sources (production types 1 and
2) emit larger (i.e., Aitken mode) particles than those formed by nucleation (production
type 3) because particles from these sources already grow inside or immediately after

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leaving the source due to condensation of semi-volatile substances when the exhaust or flue-gas are cooled and diluted (Shi and Harrison, 1999; Alam et al., 2003; Charon and Harrison, 2003). The size range for the road traffic emissions is consistent with the particle number size distributions for injection gasoline and diesel engines with the majority of particles in the diameter interval of 20–60 and 20–130 nm, respectively (Maricq et al., 1998; Morawska et al., 1998, 2008; Ristovski et al., 1998). Diurnal variation of the road traffic in cities has a typical time pattern with a maximum in the early morning and a second broader maximum in the afternoon on workdays (e.g., Salma et al., 2004). As a consequence, similar time dependency is often observed for number concentration of particles emitted by road traffic. Particles from combustion can sometimes grow in size in urban environments mainly due to condensation of vapours formed by restricted-scale photochemical processes (Jeong et al., 2006; Park et al., 2008). This growth is mainly favoured in the afternoons. Atmospheric nucleation is thought to arise from SO₂ oxidation and probably also from the oxidation of volatile organic compounds (Jeong et al., 2004; Zhang et al., 2004b; Kulmala et al., 2007; Metzger et al., 2010; Sipilä et al., 2010; Hamed et al., 2010). It can occur as a burst or persist up to several hours (Qian et al., 2007). Organic acids were proposed to enhance new particle formation possibly through stable complex formation with sulphuric acid (Zhang et al., 2004a, b). Since the conditions for nucleation include supersaturation of some low volatility components, the freshly nucleated particles almost always grow in size. It seems likely that condensation of organic vapours contribute substantially to the growth (O'Dowd et al., 2002), and participates at least in the initial steps of the growth (Kulmala et al., 2004b). Coagulation also affects the growth significantly (Stolzenburg et al., 2005). In cities, particle growth is observed together with nucleation. Primary ultrafine particles have major relevance for the air quality and emission source apportionment, and are associated with local sources of point or diffusive character. Nucleation, however, usually happens on larger spatial scales than the other two production types, and it influences the global particle budget significantly as one of the basic atmospheric aerosol processes (Kulmala et al., 2004a).

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Atmospheric ultrafine aerosol particles can grow into the cloud condensation or ice nuclei. They influence the indirect climate effects of atmospheric aerosol, and water cycling (Andreae and Rosenfeld, 2008, and references therein). The uncertainty of their climate impacts belong to the largest single contributions in the atmospheric radiative forcing calculations. Ultrafine aerosol particles are present in large numbers in polluted urban, workplace or indoor air, and they 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, and references therein). With the advancement and applications of nano-science and -technology, the relevance of the topic is expected to increase further because they or their products potentially emit ultrafine particles into the air.

Time evolution of number size distributions makes it feasible to study the nucleation and particle growth. However, the available instruments do not allow the direct measurement of nucleated particles at present but they are limited to a certain size. The detection limit has been recently moved to diameters being nowadays smaller than 1.5 nm (Mirme et al., 2010; Sipilä et al., 2010). This means that the initial particles, which mass diameters are considered to be around 1 nm (Kulmala et al., 2000, 2005b) and electrical mobility diameters around 1.5 nm, will already be observed if proper instruments are available. However, the measurements typically yield formation rates (J_d , at a measurable diameter $d > 1.5$ nm) instead of nucleation rates ($J_{1.5}$), and $J_d < J_{1.5}$ under steady state conditions. The relationship between nucleation and formation rates is influenced by the competition between condensational growth (which also depends on the condensing vapour concentrations) and coagulation scavenging, and it strongly depends on the size distribution of the existing aerosol particles. Evaluation of the experimental data from atmospheric measurements is disturbed by large associated uncertainties of the input data, sensitivity of the theoretical model to the uncertainties, and by some unknown variables. In addition, it was revealed that the nucleation process is tied to the existence of sub-3 nm pool of neutral and charged clusters which exists almost all the

time (Kulmala et al., 2005b, 2007; Sipilä et al., 2008; Mirme et al., 2010), and which further obscures the description. The exact growth behaviour for the initial phase is unknown. Therefore, formation rates are derived and interpreted in the present paper rather than nucleation rates.

New aerosol particle formation events and growth were observed around the world in a variety of geographic locations and ambient conditions in both the boundary layer and free troposphere including remote (Arctic, Antarctic or Alpine) locations, coastal environments, boreal forests, rural, agricultural and urban sites, polluted cities, volcanic emissions and exhaust plumes. The observations were reviewed by Kulmala et al. (2004a), Holmes (2007), and Kulmala and Kerminen (2008). It was concluded that long-term continuous measurements in all types of environment are desirable for better understanding and quantifying the mechanism, the extent of production, and role of ultrafine particles in more detail. Considering this, number size distributions of atmospheric aerosol particles were determined in an electrical mobility diameter range of 6–1000 nm near central Budapest continuously for one year, as a follow up of our research on urban aerosol (e.g., Salma et al., 2005, 2006, 2007; Maenhaut et al., 2005) and with the primary purpose of studying new particle formation in an urban environment. The main objectives of this paper are: to report average atmospheric particle number concentrations for various size fractions; to present individual and mean particle number size distributions; to discuss the occurrence of nucleation events; to derive and evaluate particle formation and growth rates; to study the seasonal dependency of properties mentioned; to estimate the spatial scale of the nucleation events.

2 Experimental

2.1 Measurements

The measurements were carried out in the Lágymányos campus of the Eötvös University in Budapest (47°28'29" N, 19°03'43" E, 115 m above the m.s.l.). The location

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is near the central part of the city in downwind direction. The major pollution sources include automotive road traffic, residential and commercial heating, biomass burning and biogenic emissions within the agglomeration. Long-range transport of some pollutants also plays a considerable role (Salma et al., 2001, 2004; Salma and Maenhaut, 2006). The measurements were performed in a distance of about 80 m from the bank of the river Danube. The site is influenced by a wind channel which is formed above the river. 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. The location is displayed in Fig. 1. The map also reveals the advantageous coincidence between the prevailing wind direction and orientation of the natural topological forms. The measurements were performed continuously from 3 November 2008 to 2 November 2009, thus for one year. An overview on days with available and missing data is shown in Table 1. The measurement campaign covers 95% of the days. Relative numbers of days for which the measured data were missing for more than 4 h are 3.3, 2.2, 10.9 and 4.4% for the winter, spring, summer and autumn, respectively. No data are missing at all for 90% of the days. This means that the acquired data sets are representative for the seasons and year.

The measurement system consisted of a flow-switching type differential mobility particle sizer (DMPS) and a meteorological station. The main parts of the DMPS included a ^{241}Am neutralizer, 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 DMPS operates at two sets of flows. In the first (high flow) mode, the sample air and sheath air flows are 2 and 20 l min^{-1} , respectively, while in the second (low flow) mode, they are 0.3 and 3 l min^{-1} , respectively (Aalto et al., 2001; Vakkari, 2008). The DMPS records particle number concentrations for a unity logarithmic electrical mobility diameter ($dN/d\log d$, where N is the particle number concentration) from 6 to 1000 nm in 30 channels. Of them, 20 channels were recorded during the high flow mode, and 10 channels were acquired during the low flow mode. This means that particles with a diameter from 6 to 200 nm were measured during the

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high flow mode, and particles with a diameter from 200 to 1000 nm were measured during the low flow mode. Measuring the whole size range of interest takes approximately 10 min, which was set as the compromise between the time resolution and particle counting statistics. This choice yielded about 138 spectra a day. Operational parameters (flows and high voltage values) of the DMPS were checked and calibrated at least once in two weeks. The sampling line was made of Cu tubing with an internal diameter of 4 mm and length of approximately 2 m. There was no upper size cut-off inlet applied to the sampling line, and a weather shield was only adopted. The performance of the DMPS to measure the total number of particles accurately was tested with another CPC (model 3786, TSI, USA) with a lower measurement limit of 2.5 nm. It was operated in parallel with the DMPS system on a day when aged aerosol was dominating. The DMPS-to-CPC3786 concentration ratio was 0.93 ± 0.14 , and a correlation coefficient of $R=0.933$ ($p < 2 \times 10^{-4}$) was obtained. These indicate a satisfactory agreement between the two instruments (Morawska et al., 2008). It has to be mentioned that the DMPS operates in a dried sample flow (with typical relative humidities between 10 and 25%) and, hence, it measures particles in the dry state. This paper deals with dried sizes since no information was available on hygroscopicity of ultrafine particles in Budapest.

Basic meteorological data, i.e., solar radiation, wind speed and direction, ambient relative humidity, temperature, pressure were directly obtained from the Urban Climatological Station of the Hungarian Meteorological Service operated in the campus.

2.2 Data treatment

The measured data were mathematically inverted on-line after each measurement cycle. The inverted individual data were 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 size fractions represent well the total number of aerosol particles and the ultrafine particles, respectively (see Sect. 3.2). Daily and yearly average concentrations were calculated for the size fractions. The

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inverted individual concentration data for a fixed channel were also averaged for separate months to derive monthly mean number size distributions. In a few cases, daily mean number size distributions were also derived. The distributions were fitted by log-normal functions using the DoFit algorithm (Hussein et al., 2005) to obtain the modal concentrations, number median mobility diameters (NMMDs) and geometric standard deviations (GSDs) for the modes identified.

The inverted individual concentrations were utilised to generate contour plots showing jointly the time variation in particle diameter and normalised particle number concentration. The plots were classified on a day-to-day basis. The time interval considered is justified by the fact that all nucleation events appeared in the daytime and were virtually finished by the evening (see Sect. 3.3). The classification was accomplished via an algorithm proposed earlier (Dal Maso et al., 2005). The relevant days were classified visually by two experts into the following groups: nucleation event class 1, nucleation event class 2, no nucleation event, and event with undefined feature. Each day was assigned only to one of the classes. The main purposes of this classification were 1) to separate the days with evident nucleation event, 2) to select the days with unambiguously no nucleation, and 3) to create a statistical overview on them. To achieve goal 1, nucleation should be clearly distinguished from the direct emissions of local combustion sources and restricted-scale photochemical growth. Therefore, the classification scheme involves the criteria for both the appearance of a new mode at the measurement limit of the instrument, and its remarkable growth subsequently. However, we are aware that there are no discrete boundaries between the groups. The space available for the formation process can, for example, affect the contour plots. Increase in the nucleation mode NMMD is not expected to be observed if the spatial extension of the nucleation is restricted to a point or line, and if the new particles are transported by horizontal advection to a fixed measurement site that is in a close distance from the nucleation area (Kulmala et al., 2004a). The algorithm works well for relatively large open spaces such as our measurement site. Fluctuations caused by the turbulent nature of the atmosphere, and limitations inherent to every spatially fixed

single measurement setups can also hinder the classification and evaluation.

Nucleation events of class 1 represented processes without disruptions and, therefore, were further analysed. Each individual size distribution on these days were fitted using the DoFit algorithm (Hussein et al., 2005), and the modal parameters for the nucleation, Aitken and accumulation modes were derived. A subset (sometimes called nucleation event class 1a) of 31 events could be separated from the total number of 45 nucleation class 1 events for which the nucleation mode growth was so well developed in time that the evolution of its parameters made it feasible to calculate particle formation and growth rates. Time evolution of an aerosol population is described by the general dynamic equation (Seinfeld and Pandis, 1998) which can be approximated by (Dal Maso et al., 2002; Kulmala et al., 2004a):

$$J_d = \frac{dN_{\text{nuc}}}{dt} + F_{\text{coag}} + F_{\text{growth}}, \quad (1)$$

where J_d is the flux of particles into the observable size range at diameter d , N_{nuc} is the number concentration of nucleated particles, F_{coag} expresses the loss of particles due to coagulation, F_{growth} is the flux of particles out of the size range due to growth, and t is time. The effects of horizontal advection and mixing from above are not taken into consideration in this approach. For regional nucleation, horizontal inhomogeneity is thought to be negligible. For our measurement system, $J_d = J_6$. The size range for the nucleated particles was chosen to be from 6 to 25 nm, thus $N_{\text{nuc}} \approx N_{6-25}$. In this way, F_{growth} can be neglected since particles rarely grow out from this range before the formation ends (Dal Maso et al., 2005). It was presumed that the intensity of the nucleation source is constant for a certain time period, and hence dN_{6-25}/dt was determined as the slope of the function N_{6-25} versus time within a period that could be approximated by a linear fit. The computation of the coagulation loss term was adopted from the procedure of Dal Maso et al. (2005). In short, the coagulation rate was expressed as:

$$F_{\text{coag}} = \text{Coag}S_{\text{nuc}}N_{\text{nuc}} \approx \text{Coag}S_{\text{NMMD}}N_{6-25}, \quad (2)$$

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where $\text{CoagS}_{\text{nuc}}$ is the coagulation efficiency of particles in the nucleation mode. In Eq. (2), it was approximated by the coagulation efficiency for the NMMD of the nucleation mode. The coagulation coefficient $K(u, v)$ for two spherical particles with volumes between $(u, u+du)$ and $(v, v+dv)$ was calculated according to Fuchs' generalisation as (Seinfeld and Pandis, 1998):

$$K(u, v) = \frac{4\pi r_{uv} D_{uv}}{\frac{r_{uv}}{r_{uv} + \sigma_{uv}} + \frac{4D_{uv}}{c_{uv} r_{uv}}}, \quad (3)$$

where $D_{uv} = D_u + D_v$ is the binary particle diffusion coefficient, $r_{uv} = (r_u + r_v)/2$ is the interception distance of the particles, $\overline{c_{uv}}$ is the relative thermal velocity of the particles, σ_{uv} is a distance parameter resulting from the flux matching approach. The coagulation coefficient for a particle with a reference diameter of NMMD and for polydisperse aerosol was obtained by integrating Eq. (3). The mean value of F_{coag} over the observed linear formation period was derived, and it was adopted in the calculation of the formation rate. The procedure represents an effective method that handles fluctuating data well.

Growth rate is the rate at which a characteristic diameter of an aerosol particle, population or size mode grows. Median diameter for the nucleation mode was considered to be the characteristic diameter of the nucleated aerosol population. Growth rates also depend on time. To estimate its value at the actual lower measurement limit of 6 nm, a linear line was fitted to the first several NMMD data in its time evolution plot, and the growth rate value was derived from the slope of the fitted line.

3 Results and discussion

3.1 Average atmospheric concentrations

Daily median N_{6-1000} concentrations varied from 3.8×10^3 (8 March) to $29 \times 10^3 \text{ cm}^{-3}$ (9 January) with a yearly median of $11.8 \times 10^3 \text{ cm}^{-3}$. The overall mean and standard

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deviation of the daily mean concentrations were $(13.0 \pm 4.5) \times 10^3 \text{ cm}^{-3}$. This yields a relative standard deviation of 35%, implying that there was not much fluctuation in the production and removal of the particles on a daily basis. The average concentrations are comparable to, but smaller than, the typical values of $15\text{--}23 \times 10^3 \text{ cm}^{-3}$ reported for other urban environments (Väkevä et al., 2000; Woo et al., 2001; Jeong et al., 2006; Watson et al., 2006; Qian et al., 2007; Park et al., 2008). Time variation of the daily median concentration for the whole year is shown in Fig. 2. It is seen that the concentration shows no seasonal tendency. Daily and monthly mean N_{6-1000} concentrations also confirm the lack of seasonal dependency. This differs from some other cities, where the largest monthly concentration was observed in February, and the smallest monthly concentration in July (Jeong et al., 2006). The two largest values for 9 and 10 January stand out from the other data. They relate to a period when a smog alert was announced for Budapest (11 and 12 January 2009). Daily median N_{6-100} (ultrafine particle) concentration ranged from 2.7×10^3 (8 March) to $20 \times 10^3 \text{ cm}^{-3}$ (9 January) with a yearly median of $9.3 \times 10^3 \text{ cm}^{-3}$. Its time variation is also shown in Fig. 2. No seasonal tendency is observed for the ultrafine particles either. It is worth noting here that daily medians of N_{25-100} (mainly representing the particles emitted directly) also did not show seasonal tendency, similarly to most criteria air pollutants in Budapest.

Strong association between N_{6-100} and N_{6-1000} is evident from their correlation coefficient of 0.972 ($p < 10^{-4}$). Contribution of ultrafine particles to the total particle number, calculated as the N_{6-100}/N_{6-1000} concentration ratio, varied from 58% (29 December) to 92% (20 July). Daily median concentration ratios did not show remarkable seasonal tendency. The individual ratios were averaged for the whole year, for the subset of days with nucleation, and for the subset of days without nucleation. Mean ratios and standard deviations of $(79 \pm 6)\%$, $(82 \pm 5)\%$ and $(78 \pm 6)\%$, respectively were obtained for the data sets listed. The yearly mean ratio is comparable to, but somewhat smaller than, the range of 88–94% reported for some other cities (Jeong et al., 2006). For the most polluted two days of 9 and 10 January, the ratios were 68% and 60%, respectively, at the lower end of the interval observed. Nevertheless, correlation between the

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daily median N_{6-100}/N_{6-1000} ratio and N_{6-100} or N_{6-1000} concentrations were insignificant. This implies that no evident (linear) relationship between the particle number concentration and ultrafine contribution could be established. Collectively this means that ultrafine particles make up the major fraction of the total number of the particles in Budapest, and that their contribution to the total particle number is fairly constant throughout the year.

3.2 Mean size distributions

The Aitken and accumulation modes were invariably present in the individual number size distributions. They merged into a broad peak which could be resolved by fitting. In general, the accumulation mode was slightly larger in the early mornings than the Aitken mode, while the opposite was experienced in the afternoons and evenings. Monthly mean number size distribution for September 2009 is shown in Fig. 3a as example. Differences in the shape and area of the monthly mean size distributions were not substantial. Overall mean NMMDs for the Aitken and accumulation modes derived from the monthly mean distributions were (26 ± 2) nm and (93 ± 10) nm, respectively with an identical overall mean GSD of 2.1. As expected, the averaging caused broadening of the modes. In ordinary situations (except for the first 2–3 h after a nucleation event is detected), the N_{6-100} and N_{6-1000} concentrations represent accurately the number of ultrafine aerosol particles and the total number of aerosol particles, respectively. The Aitken mode and, in particular the accumulation mode were generally shifted to smaller values than for rural or background environments, where their typical dry diameters are 30–60 and 150–250 nm, respectively (Raes et al., 2000; Dal Maso et al., 2005). Mean ratio and standard deviation of the modal concentrations for the Aitken and accumulation modes was 1.14 ± 0.11 . In general, the Aitken mode was somewhat larger than the accumulation mode, which is explained by the intensity of road traffic emissions near this location.

The nucleation mode showed up episodically at the lower measurement size limit of the DMPS and it converged progressively toward the Aitken mode for several hours.

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Number size distribution of particles for 10:00 local time on 8 May 2009 is shown in Fig. 3b to illustrate this. It was found that the NMMDs for the Aitken and accumulation modes are shifted to larger values (typically to 50–60 nm and above 150 nm, respectively) over the growth process with respect to the monthly mean distributions. This can be related to the fact that aged aerosol dominates under the conditions that favour nucleation. Figure 3b also demonstrates that the total particle concentrations are increased considerably during nucleation events. Overall mean and standard deviation of the nucleation mode concentrations were $(10.4 \pm 2.8) \times 10^3 \text{ cm}^{-3}$ with a maximum daily mean of $16.1 \times 10^3 \text{ cm}^{-3}$. The ratio of the typical nucleation mode number concentration to the total particle number concentration averaged for two hours just prior to the formation, was in the range 0.89 to 5.0, with a mean and standard deviation of 2.3 ± 1.1 . Ratios below unity correspond to more polluted situations when the number concentration of particles from traffic emissions decreased suddenly due to mixing of air parcels, and due to changes in traffic circulation. Nucleation occurs when the number concentration levels are relatively low, and it increases the concentrations considerably. The combined effect smoothes the variability in the daily average concentrations mentioned in Sect. 3.1. The nucleation mode could not be identified in the monthly mean number size distributions because of its shifting position and relatively large concentrations for the Aitken and accumulation modes.

3.3 Time evolution of the size distributions

Typical contour plots for traffic emissions and/or nucleation events on four separate days are shown in Figs. 4a, 4b, 5a and 5b. It is seen in Fig. 4a that the number concentration of aerosol particles in the diameter range of 20–100 nm started to increase substantially around 06:00, due mainly to higher intensity of road traffic (Salma et al., 2004). It peaked around 07:00 and then it decreased monotonically as different air parcels such as those containing hotter exhaust gases and cool ambient air (possibly from the boundary layer of the previous day) were mixed. Decreasing traffic intensity was also a factor. The concentration dropped substantially by 11:00 when the morning

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rush hours were over. Increased N_{25-100} concentration was observed again from about 14:30 for the afternoon rush hours or over the whole remaining day as a consequence of a more stable boundary layer in the late afternoon and evening. These (Aitken mode) particles occasionally exhibited demonstrable growth due to restricted-scale (local) photochemical processes on sunny afternoons. Such situation is shown in Fig. 4b. Typical growth rates observed for the Aitken mode are close to the growth rates derived for the nucleation mode (see Sect. 3.5). Similar phenomena were interpreted earlier as morning particle formation events or local SO_2 -related particle formation events (Jeong et al., 2006). We do not believe they relate to nucleation because the size mode does not appear below 10 nm. These particles are just emitted directly. It is important to distinguish these events from the nucleation and consecutive growth because they involve already existing particles. It should be mentioned that the interval <10 nm, crucial for making the distinction, was not accessible for those authors.

Figure 5a demonstrates that both road traffic emissions and nucleation can contribute substantially to atmospheric concentrations on the same day. It is seen that the mixing of the air parcels mentioned above contributed substantially to the drop in the N_{25-100} concentration at about 09:30. It diluted the pre-existing aerosol, possibly increased the SO_2 and other relevant concentrations near the surface, and decreased the air temperature. These changes together with the increased solar radiation created favourable chemical and meteorological conditions for nucleation. It occurred, and the freshly nucleated particles were growing in size. They reached the measurement size limit of the instrument at 10:34. Their growth was traced from this instant for several hours. The wide onset of the growth curve probably indicates a sustained nucleation for a longer time (up to 6 h). By the beginning of the afternoon rush hours, at around 14:30, the nucleation mode had grown to a mode characterised by an NMMD of approximately 18 nm. The mode overlapped with the distribution of particles from traffic emissions that emerged again due to the increased road traffic. Their superposition led to a modest but sudden shift in the fitted nucleation mode NMMD at 14:34. The secondary (nucleated) particles were mixed with the primary particles before they reached

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their ultimate size. This type of the contour plot represents the ordinary time evolution of the size distributions for days with nucleation. The coincidence of the beginning of the nucleation events with the sudden decrease in particle number concentrations from traffic emissions, and with an increased Aitken mode NMMD, probably reflects their joint dependency on additional properties. Only a few particle growth processes occurred without disturbances. One of them is shown in Fig. 5b. A cold front passed the city in the afternoon and evening on Friday, 7 November, and it removed the air pollutants from the region, creating clean atmospheric conditions. Lower traffic densities and emissions on the next day (Saturday) also influenced the actual time development advantageously. The narrow onset of the growth curve suggests that the nucleation occurred in a short time interval as a burst. The nucleated particles were detected from 08:32. Evolution of the NMMD for the nucleation mode was well developed and it reached its ultimate value at approximately 60 nm. The contribution of the traffic emissions in the morning and afternoon was weak on Saturday though emissions are still visible on the contour plot.

3.4 Nucleation event statistics

Basic statistics on available days, days with missing data, days with evident nucleation event, days with unambiguously no nucleation, and days with undefined feature for the four seasons and whole year are shown in Table 1. It is seen that nucleation events are not rare in Budapest. They occurred on 83 days, which represents 27% of all relevant days. Nucleation frequency exhibited a remarkable seasonal variation with a minimum of 7.3% in winter and a maximum of 44% in spring. In summer and autumn, the frequencies were similar to each other with a value of 28–29%. The frequencies of the days without nucleation complement these values to unity. The numbers of days that could not be classified into the two main categories were 5, 12, 7 and 10 for winter, spring, summer and autumn, respectively. The undefined feature could be caused by disturbances from local pollution sources and, more importantly, by limitations of all measurements at a fixed site which can be exposed to anisotropic air masses causing

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repeatedly or completely discontinued modal growth. These days were removed from the statistics but their maximal contribution is smaller than the seasonal variation in the frequency. Therefore, they do not alter the tendency observed. The nucleation frequencies are similar to those at urban St. Louis, USA with a minimum of 8% in winter and a maximum of 36% in April–September (Qian et al., 2007). Figure 6 shows monthly mean nucleation frequencies throughout the year. It is seen that the largest frequency occurred in April, and a local maximum appeared in September. The distribution has no clear time dependency during the rest of the year. The largest nucleation activity was observed during summer (July) in urban St. Louis, USA (Qian et al., 2007), in spring (April) in the Hyytiälä forest, Finland (Dal Maso et al., 2005) and at Rochester, NY (Jeong et al., 2006), and in winter (January) and spring (April) in the Hohenpeisenberg mountain, Germany (Birmili et al., 2003). Annual frequency and seasonal variation of the nucleation are known to differ with location (Kulmala et al., 2004a). Nucleation activity can probably be related to local features, e.g., to seasonal variation of solar radiation which changes the photochemical oxidation of precursors of nucleating compounds, of ambient temperature because the kinetics depends on it sensitively, of relative humidity since its large values decrease the nucleation rates (Kiendler-Scharr et al., 2009), and of biogenic emission of precursors in the region which changes the nucleation in a complex way. It was also shown that local wind speed, boundary layer height, gas phase chemistry, long-range transport of pollutants, and concentration and size distribution of atmospheric aerosol particles also play a role (Väkevä et al., 2000). The nucleation activity may be also associated with some limiting atmospheric process in the actual region. Reasons for the nucleation events in Budapest, relationships between the nucleation properties, pollutant gases and meteorological variables are to be dealt with in a separate paper.

3.5 Formation and growth rates

Formation rates of particles with a diameter of 6 nm are shown in Fig. 7a. They vary between 1.65 and 12.5 cm⁻³ s⁻¹ with a mean and standard deviation of

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$(4.2 \pm 2.5) \text{ cm}^{-3} \text{ s}^{-1}$. Coagulation losses represented approximately 20–130% of the particle flux dN_{6-25}/dt with a mean of 77%. It indicates the importance of coagulation for nucleated particles in cities since the directly observable particle flux was on average ca. 60% of the real formation rate. The rate values derived are comparable to other urban data and larger than the values for background environments, though they were often reported for different diameters of 3 or 10 nm (Kulmala et al., 2004a, and references therein). Monthly mean formation rates are also included in Fig. 7a. Seasonal dependency could not be identified on the basis of the individual data or of the mean values because of their spread and limited number.

Individual growth rate values and monthly means are shown in Fig. 7b. The growth rates range from 2.0 to 13.3 nm h^{-1} with a mean and standard deviation of $(7.7 \pm 2.4) \text{ nm h}^{-1}$. They essentially fit into the ranges of 3–11 nm h^{-1} (Alam et al., 2003), 2–6 nm h^{-1} (Kulmala et al., 2004a), 1.1–16.0 nm h^{-1} (Kulmala et al., 2005a), 3–22 nm h^{-1} (Stolzenburg et al., 2005), 5–13 nm h^{-1} (Jeong et al., 2006), $(5.9 \pm 4.7) \text{ nm h}^{-1}$ (Qian et al., 2007), and 2–5 nm h^{-1} (Park et al., 2008) reported for other urban environments. It is realised that the growth rates for various locations were not given for exactly the same particle diameters. There is a tendency for larger growth rates in summer, and for smaller values in winter in our data sets but the number of data available (in particular for winter) does not make it feasible to quantify the tendency. This indicative dependency becomes more evident when the monthly mean growth rates are considered. It is consistent with the tendency observed at another urban location (Qian et al., 2007) and other background sites (Kulmala et al., 2004a). The larger summer time growth rates seem to reflect the higher temperatures and larger concentrations of condensing species due to increased photochemical activity.

3.6 Spatial scale of nucleation

Information about the spatial scale of nucleation can be important with regard to sources of possible precursors or to nucleating species. Time evolution of the

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nucleation mode observed in Budapest (see Fig. 5a and 5b) is usually similar to the characteristic banana pattern for rural and background locations (Kulmala et al., 2004a, and references therein). This shape was preserved under various horizontal advection conditions. In addition, the nucleation events happened around the midday and not in the early morning or late afternoon (i.e., before 08:30 or after 14:15) which is typical for large scale nucleation. Its horizontal scale was estimated roughly utilising the lower measurement limit of the DMPS of 6 nm, the mean growth rate of 7.7 nm h^{-1} and mean wind speed of 3.7 m s^{-1} calculated for the mornings on the days with nucleation, and assuming that the air parcel containing the nucleated particles travelled with the mean wind speed. It is also realised that the particle growth rates in the initial phase can be much smaller than for the nucleation mode at 6 nm. An orderly estimate of 9 km was obtained in this way, while the smallest growth rate of 2.0 nm h^{-1} resulted in a spatial scale of 33 km. The estimated extensions are comparable to the linear dimensions of the city. Some nucleation events occurred when the local wind blew from the south or east. Horizontal extension of the nucleating air parcel is to be further expanded when considering the sustained types of nucleation events. This suggests indirectly that the processes involved took place at least over the whole city or its larger area. The full extent of the relevant air masses within the Carpathian basin could not be assessed because of the single fixed measurement site. Further expedient studies are required to determine the spatial scale of the nucleation more accurately. The open character of the selected location also influenced favourably the observation of regional nucleation events. It is worth noting in this respect that no nucleation followed by particle growth was recorded at all in Gwangju, Korea during several months in a 1-year measurement campaign, and the missing phenomenon was explained by the limitations of the measurement location (Park et al., 2008). Selection of an appropriate urban measurement site seems to be a more sensitive issue that in rural or background environments.

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Primary emissions generally dominate the total number of particles in Budapest. Typical median diameters for the Aitken and accumulation modes in number size distributions are substantially smaller than for rural and background environments. Although they are intermittently shifted to somewhat larger values under conditions that favour nucleation and consequent particle growth, the available diameter interval in which the time evolution of the nucleation mode can be followed without severe disturbances is still smaller. At the same time, the number concentrations for the Aitken and accumulation modes are larger than for rural and background environments. In addition, some criteria pollutants exhibit larger spatial and temporal heterogeneity in cities. On the one hand, polluted urban air may prevent nucleation due to larger particle surface concentrations (e.g., Mönkkönen et al., 2004), and, on the other hand, it can promote nucleation due to higher concentrations of some precursor gases and to enhanced photochemical production of the nucleating compounds. This complicates the urban studies on new particle formation and growth. It is desirable to apply experimental systems with as small measurement limits as possible, definitely below 10 nm. This is required to distinguish the particle production by nucleation from direct emissions by combustion sources, and to avoid misinterpretation of some ultrafine aerosol events as nucleation. The flow-switching type DMPS and the advanced evaluation methods utilised in the present paper were shown to be valuable tools in urban environments as well. The considerable number of regional-type nucleation events and subsequent particle growth identified and characterised in Budapest in cleaner air periods show that this formation type definitely belongs to the basic and intensive atmospheric processes in cities as well, and, more importantly, that the nucleation frequency derived is comparable to that for rural and background locations. Further studies on urban ultrafine aerosol, including nucleation and growth in various microenvironments, on chemical composition and relevant physicochemical properties of ultrafine particles are undoubtedly of scientific interest with marked public-health-related implications.

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Table 1. Total number of days, days when measurement data were missing for more than 4 h, days with undefined nucleation feature, days with evident nucleation, and days obviously without nucleation for the four seasons and whole year near central Budapest from 3 November 2008 to 2 November 2009.

	Winter	Spring	Summer	Autumn	Year
Total number of days	90	92	92	91	365
Number of days with missing data	3	2	10	4	19
Number of undefined days	5	12	7	10	34
Number of days with nucleation	6	34	21	22	83
Number of days with no nucleation	76	44	54	55	229

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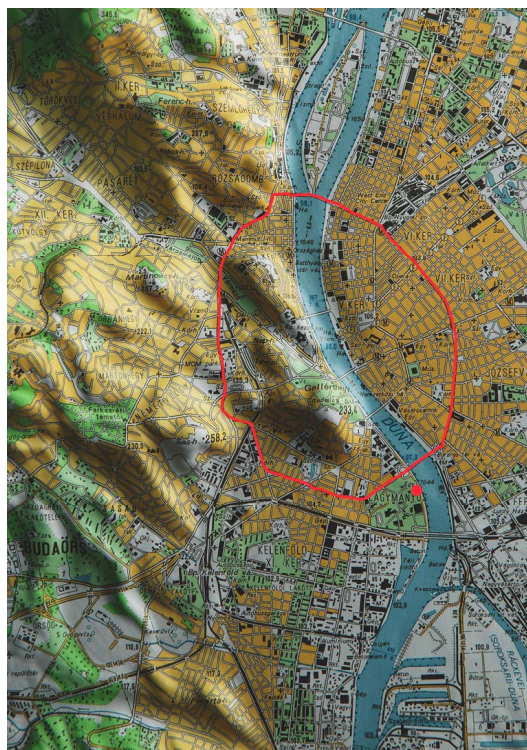


Fig. 1. Map of Budapest showing the central part of the city (indicated by red line) and the measurement location (marked by red dot) at the campus of the Eötvös University downwind from the central urban part, on the bank of the river Danube. The picture was prepared from the Topographic map of Budapest (HM Térképészeti Kht., 2004).

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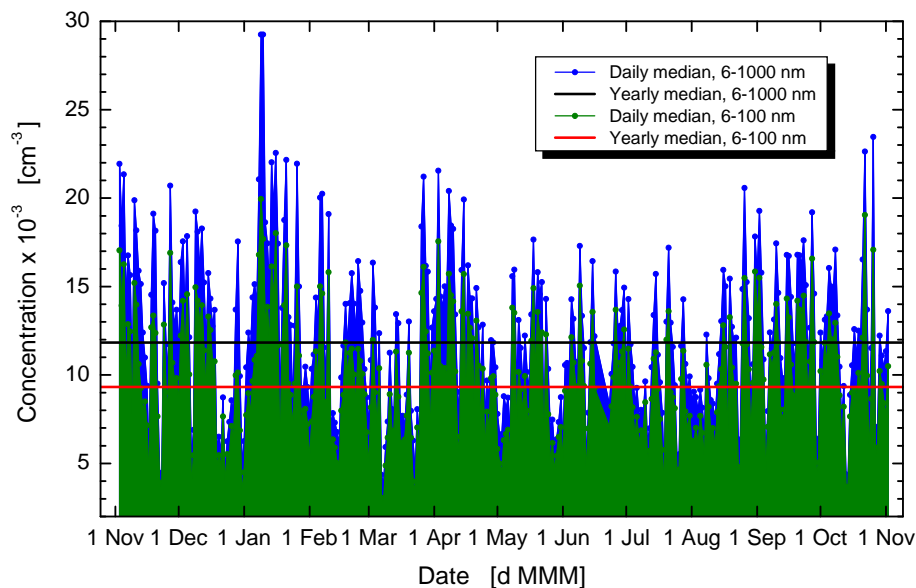


Fig. 2. Median particle number concentrations in the diameter ranges from 6 to 1000 nm and from 6 to 100 nm near central Budapest from 3 November 2008 to 2 November 2009.

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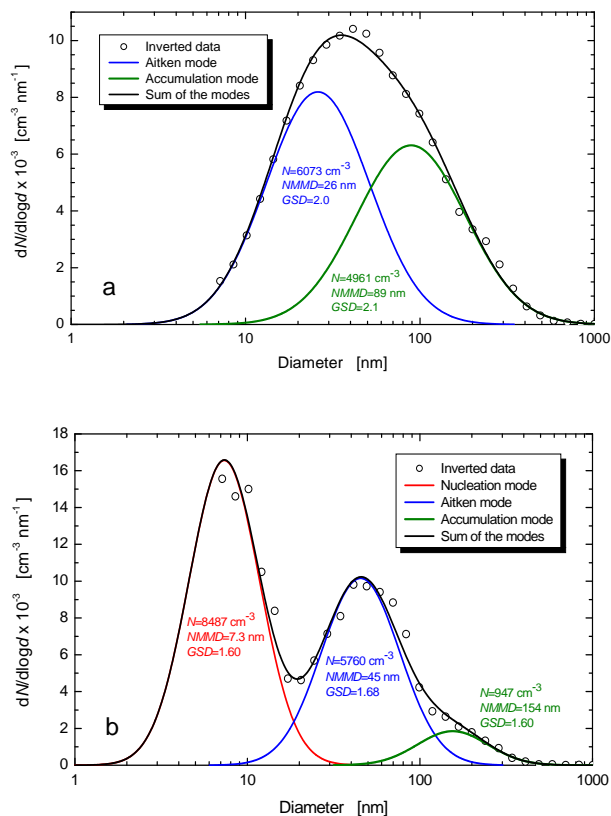


Fig. 3. Monthly mean number size distribution of atmospheric aerosol particles for September 2009 (a), and number size distribution of atmospheric aerosol particles after a nucleation event for 10:00 on 8 May 2009 (b). Modal concentrations (N), number median mobility diameters (NMMD) and geometric standard deviations (GSD) for the nucleation, Aitken and accumulation modes are also shown.

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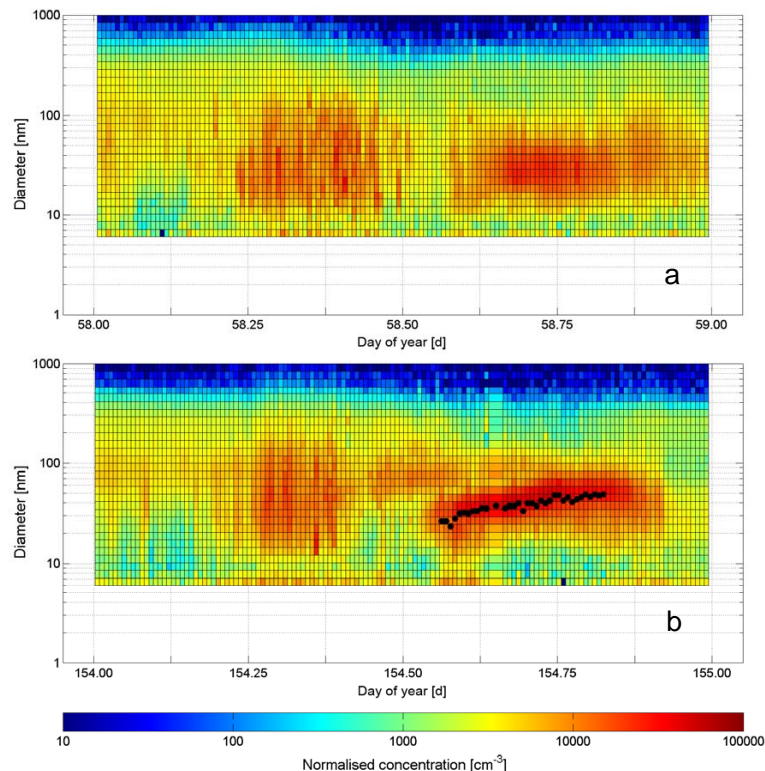


Fig. 4. Contour plots of normalised particle number concentrations showing two major automotive road traffic emission events corresponding to morning and afternoon rush hours on Friday, 27 February 2009 **(a)** and Wednesday, 3 June 2009 **(b)**. Significant growth of primary Aitken mode particles on panel b is indicated as time series for the fitted number median mobility diameter of the Aitken mode by black dots.

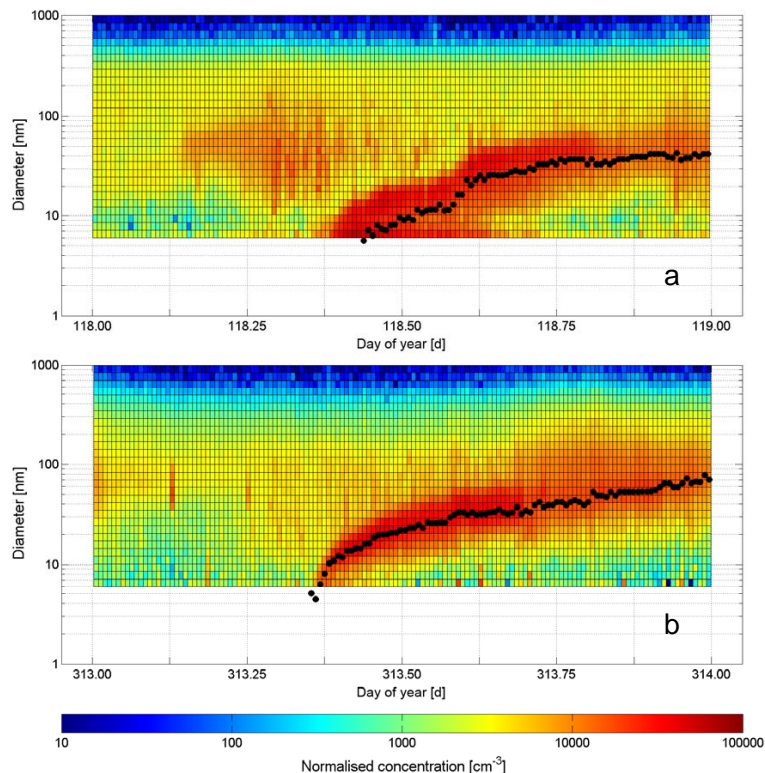


Fig. 5. Contour plots of normalised particle number concentrations measured on Tuesday, 28 April 2009 displaying road traffic emission events and a nucleation in the midday together with a consecutive particle growth **(a)**, and on Saturday, 8 November 2008 showing a nucleation and consecutive particle growth **(b)**. Time series for the fitted number median mobility diameters of the nucleation mode are indicated by black dots on both panels.

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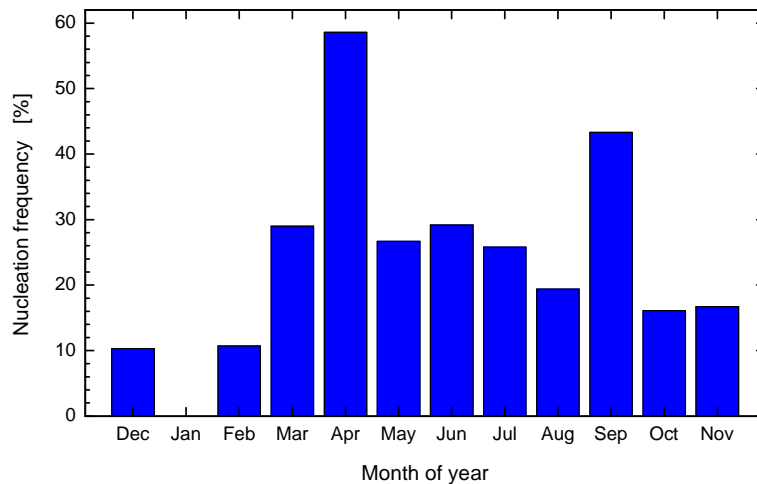


Fig. 6. Monthly mean nucleation frequency in Budapest from 3 November 2008 to 2 November 2009.

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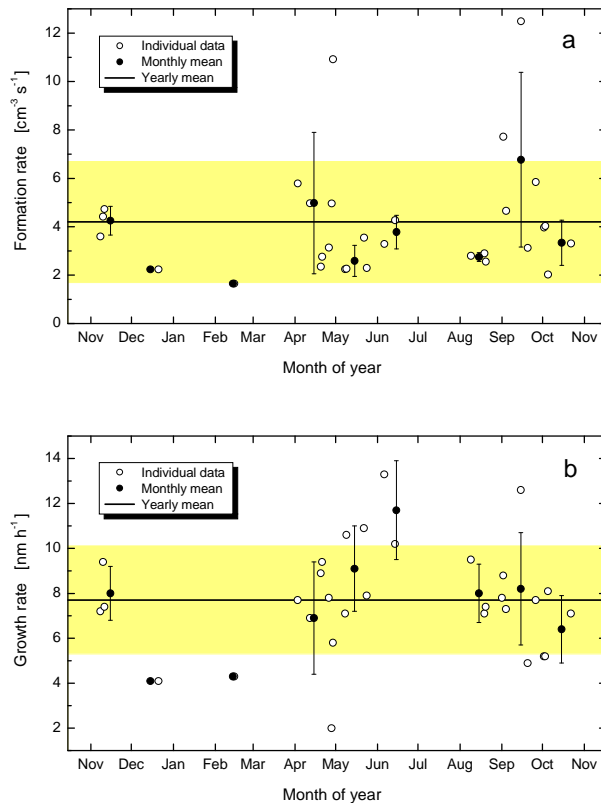


Fig. 7. Formation rates of aerosol particles with a diameter of 6 nm **(a)**, and growth rate of the nucleation mode with a median diameter of 6 nm **(b)** in Budapest from 3 November 2008 to 2 November 2009. Error bars represent standard deviations. Standard deviation of the yearly mean is indicated by yellow area.

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