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Urban aerosol number size distributions

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Abstract

Aerosol number size distributions were measured continuously in Helsinki, Finland from 5 May 1997 to 28 February 2003. The daily, monthly and annual patterns were investigated. The temporal variation of the particle number concentration was seen to follow the traffic density. The highest total particle number concentrations were usually observed during workdays; especially on Fridays, and the lower concentrations occurred during weekends; especially Sundays. Seasonally, the highest total number concentrations were usually observed during winter and spring and the lowest during June and July. More than 80% of the particle number size distributions were tri-modal: nucleation mode (Dp<30 nm), Aitken mode (20–100 nm) and accumulation mode (Dp>90 nm). Less than 20% of the particle number size distributions have either two modes or consisted of more than three modes. Two different measurement sites are used; in the first place (Siltavuori, 5 May 1997–5 March 2001), the overall means of the integrated particle number concentrations were 7100 cm^{-3}, 6320 cm^{-3}, and 960 cm^{-3}, respectively, for nucleation, Aitken, and accumulation modes. In the second site (Kumpula, 6 March 2001–28 February 2003) they were 5670 cm^{-3}, 4050 cm^{-3}, and 900 cm^{-3}. The total number concentration in nucleation and Aitken modes were usually significantly higher during weekdays than during weekends. The variations in accumulation mode were less pronounced. The smaller concentrations in Kumpula were mainly due to building construction and also slight overall decreasing trend during these years. During the site changing a period of simultaneous measurements over two weeks were performed showing nice correlation in both sites.

1. Introduction

Atmospheric aerosol particles in urban areas cause the loss of visibility (e.g. Finlayson-Pitts and Pitts, 2000) and health effects (Dockery and Pope, 1994). Heavily industrialized areas suffer from pollution fogs (smogs) that are often related to coal burning and

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nowadays also to traffic. The most well-known example of such smogs is the London "pea-souper" smog, which occurred every once in a while until the 50's, when coal burning was forbidden. Besides visibility degradation, the London smog episodes caused serious health effects and "excess deaths". One significant part of health problems is related to atmospheric aerosols and fog droplets, since particles having diameters less than 10 μm can penetrate deep enough into the respiratory system (Dockery and Pope, 1994). Recently, the role of ultrafine particles (Dp<100 nm) have been discussed (e.g. Katsouyanni et al., 2001; Samet et al., 2000; Pope and Dockery, 1999). Peters et al. (1997) have indicated that many of the pollution-related adverse health effects may be closely related to the presence of ultrafine airborne particles.

Routine monitoring of airborne particulate matter has been generally performed using PM_{10} and $PM_{2.5}$ measurements (mass of particulate matter smaller than 10 and 2.5μm in aerodynamic diameter, respectively). The number concentration and size distribution measurements are very rare. However, in rural areas there are several ongoing measurements (see e.g. Mäkelä et al., 1997). In urban areas the results based on measurement campaigns are typically reported. E.g. Wehner et al. (2002) performed particle size distribution measurement in street canyon and Väkevä et al. (1999) measured the evolution of number concentration in street canyon. Based on size distribution measurements Shi et al. (2001) found significant amounts of nanoparticles (<10 nm diameter). Buzorius et al. (1999) studied the local and temporal variations of aerosol number concentrations in urban area finding clear temporal variation with different frequencies.

In rural areas the typical aerosol particle concentrations are between 1000 and 10 000 cm^{-3}. In urban environments air quality is typically strongly influenced by motor vehicle emissions, and number concentrations of particles exceed 104 cm^{-3}. Harrison et al. (1999) found that particle number concentration was 7.5 times higher than the background level near a busy road in Bristol, Rd Birmingham. Most of the particles emitted by engines are in the ultrafine range (Kittelson 1998), and therefore not directly influenced by industrial emissions in urban environments. Over 80% of the airborne

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particles are in the ultrafine size range (Morawska et al., 1998b; Shi et al., 2001). Ruuskanen et al. (2001) showed that the overall median (30 November 1996–13 March 1997) of the hourly mean total number concentration at an urban site in Helsinki was 16 300 cm^{-3} (more than 80% were ultrafine particles), with a maximum value of about 145 000 cm^{-3} and a mean about 20 500 cm^{-3}. In the same study (Ruuskanen et al., 2001), similar concentration levels were observed in two other European cities (Alkmaar in Netherlands and Erfurt in Germany). In Brisbane, Australia, the concentration of submicrometer particles correlated strongly with the concentrations of NO_x and CO (Thomas and Morawska, 2002; Morawska et al., 1998b), suggesting that motor vehicle emissions constituted the main source of ultrafine particles. Under varying conditions, the number of emitted particles increased with increasing engine load, whereas the mean particle diameter decreased (Franz et al., 2000; Morawska et al., 1998a). In cities, cars are usually not driven at a constant speed; there is continuous acceleration and deceleration. In acceleration a higher number of smaller particles are probably emitted (Wehner et al., 2002).

Besides vehicles fuel combustions the industry pollutants and long range transport (LRT) affect aerosol particle number and mass concentrations in urban areas. E.g. about 40–70% of $PM_{2.5}$ is assumed to be LRT in Finland (Tiitta et al., 2002; Vallius et al., 2002). Also accumulation mode particles transport well whereas the lifetime of Aitken and nucleation mode particles is much smaller. Due to coagulation and growth, nucleation mode particles may survive only few hours which correspond to the spatial scale of maximum a few hundred kilometers. Even if nucleation mode particles do not transport, the spatial scale of nucleation can still be thousands of kilometers (Mäkelä et al., 1998). Coarse fraction of PM_{10} do not normally travel more than a few tens of kilometers except some special cases like Sahara dust episodes (Arimoto et al., 1995). In practice, LRT is affected by mesoscale meteorology. If the air masses come from areas with high emissions, air parcels contain more particles particularly in accumulation mode (Kulmala et al., 2000). In Finnish conditions the position of polar front is an important factor making changes to aerosol particle concentrations (Nilsson et al.,

2001). The amount of particles arrive to a certain location depends also on wet and dry deposition and cloud processes.

Meteorological conditions and season have their influence on the emissions. In cities many people have their holidays in July which changes traffic emissions. On the other hand, in spring and summer higher concentration of aerosol precursor gases may allow photochemical reactions to produce condensable gases and subsequent nucleation and growth in urban air while it may be impossible in other conditions (Väkevä et al., 1999; Shi et al., 2001). Another example on the complexity is the background (regional) nucleation "upwind" from urban areas. It increases particle concentration in cities even if actual nucleation occurs in cleaner background areas. In this case particles grow due to higher urban pollutant concentrations, and the particle composition and mass will change. The seasonal variation is well-known e.g. for re-suspended PM_{10} which is related to springtime snow smelting governing the release of sand from wintertime gravelling. Natural primary emissions like sea-spraying have a strong annual cycle related to wind speed and sea-ice cover. The effect of season is also related to house heating. Small scale biomass burning is a significant emission source during the winter especially in the countryside. Particle concentrations are also affected by local mixing. If boundary layer is stable, locally emitted or re-suspended particles stay in smaller volume (Väkevä et al., 2000). In practice, mixing depend on wind speed, turbulence, and relative humidity (Pohjola et al., 2000).

In spite of its importance only some studies have concentrated on the seasonal variation of particulate mass (PM_x) or number of aerosol particles using long-term measurements spanning over several months or even few years (Havasi and Zlativ, 2002; Hussein et al., 2002; Kimmel et al., 2002; Morawska et al., 2002; Yang, 2002; Zhang et al., 2002). However, long-term studies will provide sufficient information on the aerosol characteristics. Recently, Laakso et al. (2003) investigated the number and mass concentrations as well as correlations between different particle size classes (nucleation, Aitken, and accumulation modes) and between number and mass concentrations in different regions (urban, suburban, forest, and rural) in Finland. On the other hand the

long term studies of aerosol size distributions are missing. The objective of the present study is to characterize the aerosol particle number size distributions in Helsinki urban area during a long-term period (6 years, 1997–2003) with continuous monitoring of the particle number size distributions (diameter <400 nm). This includes investigations on (1) year-by-year temporal variation of both the aerosol particle number concentration and size distributions, (2) the seasonal variation within each year, (3) the difference between daily patterns of workdays and weekends of aerosol number concentration, and (4) the modal behavior of aerosol particles during different times in the year, season, and day.

2. Experimental setup

2.1. Measurement site location

The particle number size distributions were measured in Helsinki (60°10′ N and 24°57′ E); Finland. The measurements took place at the University of Helsinki Department of Physics building, which was located on the bank hill Siltavuori. Siltavuori was about 20 m high peninsula that is located by the sea, and it was surrounded with urban sites including the downtown at a distance of few hundreds meters to the south west. Siltavuori was densely populated with residential and office buildings.

On 5 March 2001, the department moved into a new building in Kumpula, which was located about 3 km northeast to Siltavuori. The measurements were continued in the forth floor of the new department building, which was located on a hilltop (~20 m high). At a distance of 100 m, there was one of the major highways providing significant source of traffic emissions. The area itself was populated with residential buildings in the northeastern side, and to the west it was full of greenswards and mainly small forest mixed with houses (see also Fig. 1). Both sites can be considered as urban background sites.

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2.2. Aerosol particle measurements

The ultrafine and fine particle number size distributions have been measured continuously (5-min resolution) with a differential mobility particle sizer (DMPS) since 5 May 1997. There have been several time periods when the measurement was not operating due to instrument maintenance. In the early stages of the measurement, the instruments were under testing, and hence the reliable data were obtained since December 1997. In the present study we investigate measured size distributions until 28 February 2003.

The differential mobility analysis of aerosol particles (e.g. Adachi et al., 1990; Winklmayr et al., 1991) relies on bipolar charging of the particles, for which we used a 74 MBq KR-85 neutralizer (Liu and Pui, 1974), followed by classification of particles due to electrical mobility by a differential mobility analyzer TSI-3071 (Knutson and Whitby, 1975) and number counting with a condensation particle counter CPC TSI 3022 (Quant et al., 1992). Occasionally, the CPC was replaced with either CPC TSI 3025 (Kesten et al., 1991) or CPC TSI 3010 (Mertes et al., 1995). Consequently, the measured particle size range slightly varied, and thus, in this study we consider the particle size of 8–400 nm, which was conformal for all the measurements. The sampling volume flow rate was 1.5 lpm and the particle free and dried sheath air flow was set to a constant value of 8.5 lpm, arranged for a stable long time measurement using a recirculation sheath air flow loop (Jokinen and Mäkelä, 1997; Birmili et al., 1999).

3. Data analysis

3.1. Particle number size distributions

Aerosol data consisted of particle number size distributions (8–400 nm in diameter) during six years (1997–2003). This aerosol data is considered very huge to analyze. It required careful handling and great care during the analysis. The aerosol data was

first stored in separate files as daily basis; i.e. each year has 365 (366 for leap years) separate data files that contain the measured particle number size distributions.

The first step of the data analysis was performed as follows: (1) the aerosol data was checked for quality assurance according to reliability of the measurement and data inversion, (2) the data was then prepared for processing, which required relatively long time, (3) the data was then accumulated in larger files as yearly basis, and (4) hourly, half-hourly, daily, and monthly means, medians, standard deviations and several percentile were evaluated. The total number concentration of fine particles was integrated from the particle number size distributions.

From the cumulative frequency plots, the aerosol data showed lognormal distributions. Standard central measures were then either represented by the geometric mean (GM) and its geometric standard deviation (GSD), or simply the median. The arithmetic mean (AM) and arithmetic standard deviation (ASD) were not representative of the central measures. The GM and GSD were evaluated according to

$$\log_{10}(\mathrm{GM}_x) = \frac{1}{N}\sum_{1}^{N}\log_{10}(x_i) \tag{1}$$

$$\log_{10}(\mathrm{GSD}_x) = \frac{1}{N}\sum_{1}^{N}\left[\log_{10}(x_i) - \log_{10}(\mathrm{GM}_x)\right]^2. \tag{2}$$

The second step of the data analysis was performed as follows: (1) the data was investigated separately for workdays (Mondays–Fridays) and weekends (Saturdays–Sundays), (2) the diurnal patterns were evaluated, (3) seasonal analysis was performed, (4) the annual variation was investigated, and (5) multi-lognormal fitting was performed for the particle number size distributions.

Particle number size distributions were fitted to a multi-lognormal distribution function

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in the form (Seinfeld and Pandis, 1998)

$$\frac{dN}{d(\log(D_p))} = \sum_{i=1}^{n} \frac{N_i}{\sqrt{2\pi}\log(o_{g,i})} \exp\left[-\frac{\left(\log(D_p) - \log(\overline{D}_{pg,i})\right)^2}{2\log^2(o_{g,i})}\right], \quad (3)$$

where D_p [nm] is the particle diameter, and the three parameters that characterize an individual mode i are: the total particle number concentration N [cm^{-3}], the geometrical variance σ_g^2 [nm^2], and the geometrical mean diameter D_{pg} (nm). n is the maximum number of possible individual modes, thus $3n$ parameters are required to identify a particle number size distribution with n number of individual modes.

3.2. Weather conditions in Helsinki during 1997–2002

In addition to the aerosol particle measurements, the weather conditions (temperature, pressure, and wind speed and direction) were provided by the Finnish Meteorological Institute (FMI) in Helsinki. Throughout the five years (1997–2002), the daily mean pressure varied between 980 and 1040 hPa. The coldest period was during January and February. The wind was southwesterly and the daily mean wind speed was lower than 9 m/s. The daily mean temperature varied between −10 and 25°C; seldom was lower than −10°C in the wintertime. An annual periodicity was observed in the daily mean temperature, which was best fit to the following periodic function described

$$T(d) = 7.2 + 12.0 \cdot \sin\left[2\pi\frac{d}{366} + \frac{4}{3}\pi\right], \quad (4)$$

where T [°C] is the daily mean temperature and d is the day number starting by 1 for 1 January 1997.

According to the temperature variation, the seasons have been defined by the daily mean temperatures. However, with this method the length of different seasons might vary considerably from year to year. In the southern part of Finland, winter is usually

considered to begin during November and ends by late March. Spring starts in early or mid April and ends in mid May, when summer usually begins and lasts until mid September. Autumn begins around the last week of September. Detailed information about the weather changes and climatology can be found from the webpage of the Finnish Meteorological Institute http://www.fmi.fi/weather/climate_4.html.

3.3. Traffic density in Helsinki metropolitan area during 1997–2002

The general development of traffic in Helsinki was slightly increasing from 1997 to 2001. The annual increase was about 2% on average (Lilleberg and Hellman, 2003). In the year 2002 the traffic density in Helsinki decreased about 3%. The overall mean constitution of all vehicles in the city center during 2000–2002 was as follows: passenger cars 81%, vans 10%, trucks 3% and buses 6%. When moving away from the city center towards the city border the amount of passenger cars was 83%, vans 9%, trucks 6% and buses 2%.

Traffic density during workdays:

The daily mean (1997–2002) traffic density of all vehicles ranged between 463 000 and 525 000 vehicle/day during September on the outer motorways inside the Helsinki metropolitan. The traffic density decreased while moving towards the city center (between 360 000 and 260 000 vehicle/day). Among the weekdays, the traffic density was highest during Wednesday through Friday. Among the months, the traffic density was highest during May and June and lowest during July (summer holiday), right after Christmas, and February (ski holidays).

Among the day, the traffic density was lowest during the late night time (after 22:00) and early morning (before 04:30). The highest traffic activity was during the morning (05:00–09:00) and afternoon (13.00–17:00) rush hours. During the daytime (09:00–13:00), the traffic activity was approximately half of the value during the rush hours.

Traffic density during weekends:

The traffic density during weekends was lower than during workdays. The traffic density during weekends was higher in the city center than on the outside the city. So

the main differences in traffic density between workdays and weekends were: traffic density was higher than average during workdays and lower during weekends, and it decreased towards city center during weekdays while it increased towards city center on weekends. The daily pattern of the traffic density during workdays consisted of two peaks (morning and afternoon rush hours), while the daily pattern during weekend days consisted of a wide shallow peak (see Fig. 2).

4. Results and discussion

4.1. Particle number size distributions

The multi-lognormal fitting was performed to the particle number size distribution as half-hourly means. About 18% out of the fitting cases required either two or four modes to fit the particle number size distribution. The requirement of more than three modes could be due to very dynamistic feature of urban aerosols. In urban conditions, the particle size distribution varies rapidly in shape and magnitude following the instantaneous traffic density and local meteorology. Therefore, running half-hour means, which may include six instantaneous particle number size distributions, produces sometimes artificial mean particle number size distributions. Birmili et al. (2001) have investigated modal characteristics of urban aerosols, and they concluded that urban aerosols consist of three modes, and rarely consist of either two or more than three modes. There were clearly (as will be shown later in this study) three main modes in both sites: mode 1 (nucleation mode) that existed within particle diameter smaller than 30 nm, mode 2 (Aitken mode) within 20–100 nm, and mode 3 (accumulation mode) within particles larger than 90 nm. The geometric mean diameter of the accumulation mode is not expected to exist within particle diameters larger than 350 nm.

The overall frequencies of the geometric mean diameter within the three modes in both sites are presented in Figs. 3a–b, and those for individual modes are shown in Figs. 3c–h. Throughout the measurement period (1 November 1997–5 March 2001

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in Siltavuori and 6 March 2001–28 February 2003 in Kumpula), the nucleation mode (mode 1; Figs. 3c and f) was uni-modal in Siltavuori (D_{pg} was about 11.7 nm) and bi-modal in Kumpula (Dpg was about 8.9 and 17.7 nm, with overall geometric mean 13.8 nm). There was also bi-modal Aitken mode (Mode 2; Figs. 3d and g) in Siltavuori (D_{pg} was about 25.9 and 43.8 nm, with overall geometric mean 37.3 nm) and in Kumpula (D_{pg} was about 24.4 and 48.1 nm, with overall geometric mean 42.5 nm). The accumulation mode (mode 3; Figs. 3e and h) was uni-modal in both sites (D_{pg} was about 150.5 nm in Siltavuori and 151.8 nm in Kumpula). It should be noticed that the overall geometric mean diameters in Kumpula were larger than those in Siltavuori. This was probably due to the evolution of the aerosol particles while transported from its origin; i.e. the ultrafine particles ($D_p < 100$ nm) in Kumpula were a bit older than those in Siltavuori. On the other hand, the existence of two sub-modes within the Aitken mode range is a sign of several different processes that governed the existence and the evolution of aerosol particles in the two sites.

The geometric means as a function of time is shown in Fig. 4. In this figure we see clearly, how mean size of nucleation mode is increasing in Kumpula after the construction of new building in vicinity started. Particularly this is true during the intensive rock blasting. This is mainly due to higher concentration of big particles causing higher coagulation sink to eat up smaller particles in nucleation mode and also hindering formation of new particles (see e.g. Kulmala et al., 2001).

The frequencies of the total number concentration within each mode are presented in Fig. 5 for both sites separately. The total number concentration of the three modes was about 7100 cm^{-3}, 6320 cm^{-3}, and 960 cm^{-3} in nucleation, Aitken and accumulation modes, respectively in Siltavuori, and about 5670 cm^{-3}, 4050 cm^{-3}, and 900 cm^{-3} in Kumpula. The frequencies within a certain total number concentration range (ΔN_{tot}) and a certain geometric mean particle diameter range (ΔD_{pg}) are presented in Fig. 6. As seen from Figs. 6a and b, the total number concentration within the accumulation mode decreased when the geometric mean particle diameter increased. It is also important to notice that the total number concentration within a mode behaved relatively

the same in both sites (Fig. 6) having maximum concentrations well below 100 nm. However, the total number concentration within a mode was about 2000 cm^{-3} higher in Siltavuori than that in Kumpula within the geometric mean diameter (D_{pg}) ranges 10–20 nm and 30–60 nm (Fig. 6c). There was also a slight difference within the geometric particle diameter range 150–200 nm (Fig. 6c). The smaller concentrations in Kumpula were mainly due to building construction and also slight overall decreasing trend during the latest years. The building constructions produce high concentrations of dust particles, which act as a condensation sink for smaller particles.

During the site changing, a period of simultaneous measurements over two weeks were performed showing nice correlation. On the other hand the difference can be explained partly by the re-suspension of dust and soil from the roadbeds. The traffic activity in Siltavuori was somewhat higher than that in Kumpula, and this is why the number concentrations as well as the re-suspension were a little bit higher in Siltavuori than in Kumpula. The geometric mean and geometric standard deviations of the fitting parameters (D_{pg}, σ_g, and N_{tot}) throughout each month are presented in Appendix A. The overall geometric mean of the geometric standard deviation was about 1.71, 1.78, and 1.60, respectively for nucleation mode, Aitken mode, and accumulation mode in Siltavuori, and about 1.63, 1.71, and 1.54 for the same order in Kumpula. The overall geometric means of the fitting parameters are also presented in Table 1.

4.2. Total particle number concentrations

The total particle number concentrations were integrated from the measured particle number size distributions. The concentrations were analyzed for Siltavuori and Kumpula separately; considering 5 March 2001 as the boarder line, which represented the time of move of the measurement site from Siltavuori to Kumpula.

The yearly median and means (arithmetic and geometric) were evaluated for years 1998 through 2002. The years 1997 and 2003 were skipped from this part because they contained only short periods. Therefore, they will be presented later in the monthly variation. As can be seen from Table 2, the annual particle number concentration in

Siltavuori was relatively larger than 13 000 cm^{-3} as median, while in Kumpula it was smaller than 12 000 cm^{-3} as median. It should be noticed that about one-sixth of year 2001 (namely January, February, and the first five days of March) was contributed by Siltavuori data and the rest was from Kumpula. Also the intensive building construction and rock blasting started in Kumpula during late spring 2001. This explains the higher value of the annual number concentration for year 2001 (11 500 cm^{-3} as medians) compared to year 2002 (9500 cm^{-3} as medians). On the other hand Kumpula is somewhat less urbanized than Siltavuori, which is expected because Siltavuori was located in the heart of downtown Helsinki, while Kumpula was located outside the downtown area but nearby a main road.

Monthly means, standard deviations (std), and medians with different convenience intervals (5%, 25%, 75%, and 95%) of the total particle number concentrations were evaluated from the hourly medians. The results are presented in Appendix B. The minimum 30 minute value was around 1000 cm^{-3} and maximum around 60 000 cm^{-3}. Typical median values were around 10 000 cm^{-3}. The monthly particle number concentrations showed a decreasing trend accompanied the periodic behavior. This was observed at both Siltavuori and Kumpula (Fig. 7), and enforces the observation of the annual decrease in the number concentration. The decreasing of the number concentration can be assumed to be directly related to the traffic density and type as well as the engine performance. The annual change in the traffic density was very small (around 3%), and on the other hand, the car engine has improved noticeable during the five years. In Helsinki, the cars have been renewed continuously, and because new cars are less harmful to environment due to their engine type, this would lead to a decrease in the pollutants due to traffic.

The total particle number concentration showed a periodic behavior with high number concentrations during the wintertime and low number concentrations during the summertime (Fig. 7). A mathematical function was used to fit the logarithm of total particle number concentrations daily medians. The fitting was performed for Siltavuori

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and Kumpula separately; in Siltavuori the periodic variation was best represented by

$$\log_{10}\left[\frac{N_{tot}(d)}{1\mathrm{cm}^{-3}}\right] = 4.1369 - 0.0903 \cdot \sin\left[2\pi\frac{d}{366} + \frac{4}{3}\pi\right] \quad (5)$$

and for that in Kumpula

$$\log_{10}\left[\frac{N_{tot}(d)}{1\,\mathrm{cm}^{-3}}\right] = 4.0323 - 0.0547 \cdot \sin\left[2\pi\frac{d}{366} + \frac{4}{3}\pi\right], \quad (6)$$

where N_{tot} [cm^{-3}] is the total particle number concentration, d is the day number starting by 1 for 1 January 1997.

The weekly pattern of the number concentration was difficult to fit according to the periodic function, because the daily median number concentration variation was larger than the weekly median variation. The diurnal pattern of the number concentration cannot be seen, because the analysis was applied for the daily medians only. Clearly, the annual periodic behavior of the total particle number concentration was inversely related to that of the temperature, which was represented by a sinusoidal function of the same period of that of the total particle number concentration but they were out of phase (180°).

4.3. Seasonal variation

4.3.1. Overall mean particle number size distributions

The particle number size distributions were averaged (and also medians) separately for the different seasons in Siltavuori and Kumpula (Fig. 8). Then the overall particle number size distributions were fitted to a multi-lognormal function. Geometric mean diameters, standard deviations, and modal particle number concentrations obtained from the fitting are presented in Table 3. Different particle number size distributions were observed for different seasons in each site. The total particle number concentrations within nucleation and Aitken modes were significantly lower during weekends

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than workdays. This was due to the traffic emissions, since traffic density is significantly smaller during weekends than during weekdays (see Fig. 2). On the other hand, the particulate matter size distributions measured from diesel engine exhaust are nearly lognormal with mean diameters ranging from 60 to 120 nm, whereas that measured from gasoline engine exhaust tend to be asymmetric with smaller mean diameters ranging from 40 to 80 nm (Harris and Maricq 2001) explaining our observed size distribution.

It should be noticed that in Siltavuori the geometrical mean diameters are smaller than those obtained for Kumpula. This is mainly due to the effect of building construction and rock blasting. On the other hand, the landscape and measurement elevations were different. Siltavuori was an open area directly exposed to the traffic emissions from the downtown area from the northern side and densely office and apartment buildings with many roads and streets from the southern side. In Kumpula, the northern side of the area was mainly park fields that were partly covered with trees, while densely tree strip (about 200 m width) was separating between the physics department building in Kumpula and the main highway from the southern side. Also, the measurements in Kumpula were held at the fourth floor level (about 15 m) above the ground level. Recently formed ultrafine particles may grow by condensation and coagulation resulting in a shift of the maximum diameter of the number size distribution (Wehner et al., 2002; Wu et al., 2002; Zhang and Wexler, 2002). Therefore, during transport from the street portion to the urban background, several processes may influence the particle number size distributions, such as coagulation, condensational growth, plume dilution, and vertical mixing (Turco and Fangqun, 1999). However, the building construction nearby will produce a large amount of dust and coarse particles increasing the coagulation sink of smaller particles; especially ultrafine particles (Jung et al., 2002).

4.3.2. Total particle number concentration

The daily patterns of the total particle number concentrations were evaluated separately for workdays (Monday–Friday) and weekends (Saturday–Sunday) on monthly

basis; i.e. for each month in every year. The diurnal patterns showed a direct relationship with the traffic density with indications of elevated total particle number concentrations during the traffic rush hours; which started early morning (between 05:00 and 06:00) and continued until the afternoon (about 18:00) on workdays and between 07:00 and 20:00 on weekends. The background total particle number concentration ($<10\,000\,cm^{-3}$) was observed during the early morning time before the heavy traffic activities started (morning rush hours).

There were similarities in the diurnal patterns of total particle number concentrations during different months. This represented a seasonal variation of the aerosol particles in Helsinki. The first month series was 1 November through end of February, which was called Season 1, with lower total particle number concentrations during November and higher values during February. The second month series was 1 March–31 May (Season 2) followed by Season 3 (1 June–15 August) and then Season 4 (15 August and 31 October). The overall seasonal diurnal patterns of the total particle number concentration and the different aerosol particle modes (nucleation [<25 nm], Aitken [25–90 nm], and accumulation [>90 nm]) are presented in Fig. 9 for workdays and in Fig. 10 for weekends.

Diurnal Patterns of the total particle number concentration:

The diurnal patterns for workdays and weekends for Season 1 were different than for the other seasons. As can be clearly seen, the workdays pattern (Fig. 9a) was characterized with two peaks representing the traffic rush hours during the morning and afternoon hours. Whereas the weekend pattern (Fig. 10a) consisted of one wide-flat peak that also represents the traffic activities during the day. On workdays, the total number concentration was about $5000\,cm^{-3}$ during the early morning (01:00–04:30), and it started to elevate around 06:00 reaching to maximum ($26\,000\,cm^{-3}$) at 09:00. Then it dropped down to about $22\,000\,cm^{-3}$ between 12:00 and 14:00. Another maximum ($24\,000\,cm^{-3}$) was observed at 16:30. Then the total number concentration decayed down reaching about $10\,000\,cm^{-3}$ at midnight. On weekends, the daytime (11:00–18:00) total particle number concentration was about $12\,000\,cm^{-3}$, while during

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early morning (01:00–04:00) total particle number concentration was about 7500 cm^{-3}.

The diurnal patterns of aerosol particles during Seasons 2 (Figs. 9b and 10b), Season 3 (Figs. 9c and 10c), and Season 4 (Figs. 9d and 10d) showed similar behaviors, but number concentrations during Season 2 daytime were higher than those during Season 4. On workdays, the total number concentration was lower than 10 000 cm^{-3} before 05:00, and then suddenly rise to a maximum (~30 000 cm^{-3} in Season 2 and ~25 000 cm^{-3} in Season 4) at 07:30. At 09:00 the total number concentration dropped down to ~20 000 cm^{-3} in Season 2 and to ~15 000 cm^{-3} in Season 4. After 10:00 until midnight, the total number concentration started to drop down steadily to its background value. On weekends, during the early morning time (01:00–04:00) the total number concentration was about 10 000 cm^{-3} in Season 2 and about 8000 cm^{-3} in Season 4. Between 09:00 and 21:00, the total number concentration was about 15 000 cm^{-3} in Season 2 and about 10 000 cm^{-3} in Season 4. The diurnal patterns in Season 3 was similar to that observed in Season 4, but the maximum of morning peak on workdays was about 18 000 cm^{-3} at 07:00.

The diurnal patterns in Kumpula showed similar trend as those observed in Siltavuori. For Season 3, the workdays and weekends daily patterns were very similar in trend and values in both sites. On workdays during Seasons 1, 2, and 3 between 06:00 and midnight the total particle number concentrations were significantly lower in Kumpula than in Siltavuori, whereas the total particle number concentrations before 05:30 (early morning) were at the same values in both sites. On weekends during Seasons 1, 2, and 3 the particle number concentrations were lower in Kumpula than in Siltavuori.

The diurnal variation of nucleation, Aitken and accumulation modes (Figs. 9e–9p and 10e–10p) shows similar daily features as the total number concentration. Nucleation mode has higher concentrations typically during mornings and around noon (weekends), as well as the Aitken mode. The concentrations in Accumulation mode were much smaller than in other modes.

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5. Summary and conclusions

Long-term continuous aerosol number size distribution measurements were performed in Helsinki, Finland since 5 May 1997. The obtained data until 28 February 2003 was analyzed. The measurements were done at Siltavuori during 1997–2001. After March 2001, the measurement place was Kumpula. The temporary, diurnal, seasonal and annual variation of particle size distributions and total concentrations were investigated. Using continuous measurements the annual trends as well as seasonal variations can be clearly seen. Also investigations are not sensitive to uncommon weather or emission conditions like campaign type observations.

Over 80% of the particle number size distributions were tri-modal: nucleation mode (D_p <30 nm), Aitken mode (20–100 nm) and accumulation mode (D_p >90 nm). Less than 20% of the particle number size distributions were either bi-modal or four-modal. The geometrical mean diameters were in Siltavuori 11.7 nm for nucleation mode, 37.3 nm for Aitken mode and 150.5 nm for accumulation mode on average. The GMD's in Kumpula were 13.8 nm for nucleation, 42.5 nm for Aitken and 151.8 nm for accumulation modes on average. The overall all mean of the integrated particle number concentration in Siltavuori were 7100 cm^{-3}, 6320 cm^{-3} and 960 cm^{-3}, respectively for nucleation, Aitken and accumulation modes. In Kumpula they were 5670 cm^{-3}, 4050 cm^{-3} and 900 cm^{-3}. The difference was mainly due to the fact that in Kumpula the building constructions in vicinity producing big particles (like building dust) cause large coagulation sink to eat up significant fraction of ultrafine particles and large condensation sink to prevent new particle formation. On the other hand, the bigger overall mean GMD's in Kumpula might be also due to the evolution of the aerosol particles while transported from its origin; i.e. the ultrafine particles (Dp <100 nm) in Kumpula were a bit older than those in Siltavuori. The total number concentrations in nucleation and Aitken modes were significantly higher during workdays than during weekends. The difference in accumulation mode was smaller.

Both in Siltavuori and in Kumpula the particle number concentration temporal vari-

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ation followed closely that of traffic density. Highest total number concentrations were observed during workdays, especially on Fridays, and the lowest concentrations during weekends, especially on Sundays. This was due to smaller traffic density during weekends. Seasonally, the lowest total number concentrations were observed during summer-holiday months (June–July) and the highest total number concentrations during March–April. This was also because of lower traffic density during summer holidays in Helsinki. Also the peak numbers concentrations occurred during rush hours.

In order to be able to study urban aerosol dynamics and possible health effect of aerosol particles long continuous data sets are needed. The present data is a good example of this kind of data sets, which can be used for different purposes. E.g. the observed 95% value of number concentration can be used when new air quality directives are considered. On the other hand the effect of traffic density and building constructions can be seen and the local air quality information can be given according to known activities. Also the urban air quality models can be tested against representative continuous data set.

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Appendix A: Overall montly geometric mean ± geometric standard deviation of the modal geometric mean diameter (D_{pg}), geometric standard deviation (σ_{pg}), and total number concentration (N_{tot})

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1997	D_{pg} [nm]			σ_g			N_{tot} [cm^{-3}]		
	Mode 1	Mode 2	Mode 3	Mode 1	Mode 2	Mode 3	Mode 1	Mode 2	Mode 3
January	--	--	--	--	--	--	--	--	--
February	--	--	--	--	--	--	--	--	--
March	--	--	--	--	--	--	--	--	--
April	--	--	--	--	--	--	--	--	--
May	--	--	--	--	--	--	--	--	--
June	--	--	--	--	--	--	--	--	--
July	--	--	--	--	--	--	--	--	--
August	--	--	--	--	--	--	--	--	--
September	--	--	--	--	--	--	--	--	--
October	--	--	--	--	--	--	--	--	--
November	10.71 ± 1.48	36.77 ± 1.35	150.95 ± 1.40	1.70 ± 1.12	1.78 ± 1.08	1.58 ± 1.12	14371. ± 4.	8317. ± 3.	1291. ± 2.
December	9.76 ± 1.58	38.27 ± 1.42	145.12 ± 1.41	1.64 ± 1.15	1.78 ± 1.08	1.60 ± 1.12	17001. ± 6.	7159. ± 3.	1393. ± 2.

1998	D_{pg} [nm]			σ_g			N_{tot} [cm^{-3}]		
	Mode 1	Mode 2	Mode 3	Mode 1	Mode 2	Mode 3	Mode 1	Mode 2	Mode 3
January	10.73 ± 1.49	35.14 ± 1.33	136.25 ± 1.45	1.72 ± 1.14	1.77 ± 1.09	1.58 ± 1.12	10991. ± 4.	6789. ± 2.	1018. ± 2.
February	11.86 ± 1.32	34.21 ± 1.30	146.13 ± 1.40	1.72 ± 1.09	1.81 ± 1.07	1.62 ± 1.14	9108. ± 3.	7065. ± 2.	782. ± 3.
March	12.27 ± 1.32	36.17 ± 1.31	140.97 ± 1.43	1.71 ± 1.09	1.79 ± 1.08	1.58 ± 1.14	10918. ± 2.	9524. ± 2.	994. ± 3.
April	10.80 ± 1.35	36.19 ± 1.42	135.35 ± 1.41	1.70 ± 1.10	1.77 ± 1.09	1.61 ± 1.13	10984. ± 2.	7871. ± 2.	1452. ± 2.
May	10.11 ± 1.29	34.40 ± 1.36	126.05 ± 1.45	1.71 ± 1.09	1.81 ± 1.08	1.61 ± 1.13	8428. ± 2.	6518. ± 2.	1072. ± 3.
June	11.73 ± 1.31	39.89 ± 1.32	138.25 ± 1.44	1.72 ± 1.10	1.78 ± 1.08	1.56 ± 1.12	5354. ± 2.	5964. ± 2.	1012. ± 3.
July	13.92 ± 1.34	44.93 ± 1.35	154.42 ± 1.42	1.68 ± 1.10	1.76 ± 1.09	1.53 ± 1.13	3890. ± 2.	4586. ± 2.	710. ± 2.
August	16.12 ± 1.24	47.19 ± 1.23	168.16 ± 1.39	1.61 ± 1.08	1.70 ± 1.07	1.47 ± 1.10	4528. ± 2.	4662. ± 2.	499. ± 2.
September	16.27 ± 1.26	47.49 ± 1.28	168.00 ± 1.34	1.63 ± 1.09	1.69 ± 1.09	1.48 ± 1.10	4723. ± 2.	5248. ± 2.	607. ± 2.
October	13.23 ± 1.39	39.46 ± 1.35	149.76 ± 1.36	1.70 ± 1.09	1.75 ± 1.08	1.54 ± 1.13	5740. ± 2.	4814. ± 2.	555. ± 2.
November	11.70 ± 1.32	35.57 ± 1.34	151.82 ± 1.42	1.72 ± 1.09	1.80 ± 1.08	1.61 ± 1.14	9544. ± 2.	7676. ± 2.	886. ± 2.
December	13.67 ± 1.25	37.92 ± 1.29	162.72 ± 1.33	1.66 ± 1.07	1.78 ± 1.08	1.58 ± 1.13	7735. ± 2.	5940. ± 2.	725. ± 2.

1999	D_{pg} [nm]			σ_g			N_{tot} [cm^{-3}]		
	Mode 1	Mode 2	Mode 3	Mode 1	Mode 2	Mode 3	Mode 1	Mode 2	Mode 3
January	13.51 ± 1.27	39.49 ± 1.27	163.90 ± 1.35	1.67 ± 1.08	1.77 ± 1.08	1.58 ± 1.13	8294. ± 2.	7630. ± 2.	855. ± 2.
February	14.38 ± 1.34	40.37 ± 1.30	156.25 ± 1.36	1.69 ± 1.09	1.74 ± 1.08	1.57 ± 1.13	9018. ± 3.	8896. ± 2.	1044. ± 2.
March	13.42 ± 1.33	41.54 ± 1.35	155.80 ± 1.41	1.66 ± 1.10	1.77 ± 1.08	1.56 ± 1.12	9372. ± 3.	8035. ± 2.	1275. ± 2.
April	12.60 ± 1.28	41.94 ± 1.40	151.13 ± 1.44	1.65 ± 1.09	1.77 ± 1.08	1.55 ± 1.12	8686. ± 2.	6973. ± 2.	1011. ± 3.
May	12.48 ± 1.30	38.96 ± 1.32	143.68 ± 1.44	1.68 ± 1.10	1.75 ± 1.09	1.54 ± 1.12	7778. ± 2.	6178. ± 2.	874. ± 2.
June	12.30 ± 1.30	43.52 ± 1.38	138.98 ± 1.47	1.68 ± 1.10	1.79 ± 1.08	1.56 ± 1.12	5076. ± 2.	6200. ± 2.	1427. ± 2.
July	12.80 ± 1.30	43.40 ± 1.33	142.27 ± 1.44	1.70 ± 1.10	1.70 ± 1.11	1.50 ± 1.12	4276. ± 2.	4793. ± 2.	842. ± 3.
August	12.52 ± 1.28	41.22 ± 1.29	143.21 ± 1.42	1.67 ± 1.10	1.78 ± 1.09	1.54 ± 1.12	6018. ± 2.	5819. ± 2.	950. ± 2.
September	12.83 ± 1.31	42.76 ± 1.32	156.01 ± 1.37	1.68 ± 1.09	1.78 ± 1.08	1.58 ± 1.13	6144. ± 2.	6867. ± 2.	1123. ± 3.
October	11.97 ± 1.31	37.05 ± 1.36	151.21 ± 1.41	1.74 ± 1.09	1.79 ± 1.09	1.58 ± 1.14	7400. ± 2.	5799. ± 2.	664. ± 2.
November	11.43 ± 1.34	36.63 ± 1.35	153.81 ± 1.41	1.75 ± 1.09	1.79 ± 1.08	1.59 ± 1.14	6362. ± 3.	5490. ± 3.	736. ± 2.
December	11.65 ± 1.33	35.27 ± 1.31	145.09 ± 1.40	1.76 ± 1.08	1.81 ± 1.08	1.61 ± 1.14	6864. ± 3.	5598. ± 2.	734. ± 2.

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2000	D_{pg} [nm]			σ_g			N_{tot} [cm^{-3}]		
	Mode 1	Mode 2	Mode 3	Mode 1	Mode 2	Mode 3	Mode 1	Mode 2	Mode 3
January	12.52 ± 1.25	35.05 ± 1.26	148.62 ± 1.25	1.68 ± 1.08	1.79 ± 1.08	1.54 ± 1.11	6611. ± 2.	5712. ± 2.	602. ± 2.
February	13.00 ± 1.26	36.82 ± 1.27	150.03 ± 1.28	1.70 ± 1.08	1.78 ± 1.07	1.54 ± 1.12	5909. ± 2.	6139. ± 2.	613. ± 2.
March	10.00 ± 1.41	29.89 ± 1.35	128.13 ± 1.38	1.75 ± 1.10	1.79 ± 1.09	1.62 ± 1.13	10796. ± 3.	6568. ± 2.	939. ± 2.
April	8.70 ± 1.31	30.85 ± 1.37	123.95 ± 1.41	1.70 ± 1.10	1.80 ± 1.09	1.66 ± 1.13	13316. ± 2.	9287. ± 2.	1640. ± 3.
May	9.90 ± 1.36	35.95 ± 1.39	126.79 ± 1.48	1.75 ± 1.10	1.73 ± 1.11	1.54 ± 1.13	9196. ± 2.	7506. ± 2.	1014. ± 3.
June	10.27 ± 1.38	38.60 ± 1.37	126.37 ± 1.44	1.75 ± 1.11	1.74 ± 1.10	1.56 ± 1.13	6964. ± 2.	7044. ± 2.	1170. ± 3.
July	10.44 ± 1.39	37.12 ± 1.39	136.18 ± 1.40	1.73 ± 1.10	1.77 ± 1.10	1.61 ± 1.13	5519. ± 2.	5009. ± 2.	1114. ± 2.
August	10.47 ± 1.41	36.49 ± 1.40	126.28 ± 1.43	1.73 ± 1.10	1.79 ± 1.09	1.61 ± 1.14	6141. ± 2.	5740. ± 2.	1325. ± 2.
September	11.28 ± 1.36	38.23 ± 1.35	135.43 ± 1.42	1.74 ± 1.09	1.80 ± 1.08	1.59 ± 1.14	7279. ± 2.	7160. ± 2.	1162. ± 3.
October	11.08 ± 1.32	39.34 ± 1.42	151.70 ± 1.34	1.73 ± 1.09	1.83 ± 1.07	1.70 ± 1.13	4272. ± 2.	4734. ± 2.	1298. ± 2.
November	10.56 ± 1.31	38.90 ± 1.39	160.11 ± 1.38	1.73 ± 1.09	1.79 ± 1.09	1.67 ± 1.14	4759. ± 3.	4939. ± 3.	1037. ± 2.
December	9.93 ± 1.29	33.58 ± 1.37	152.70 ± 1.39	1.71 ± 1.09	1.81 ± 1.08	1.67 ± 1.14	5954. ± 2.	4923. ± 2.	897. ± 2.

2001	D_{pg} [nm]			σ_g			N_{tot} [cm^{-3}]		
	Mode 1	Mode 2	Mode 3	Mode 1	Mode 2	Mode 3	Mode 1	Mode 2	Mode 3
January	9.50 ± 1.27	29.90 ± 1.34	130.19 ± 1.31	1.74 ± 1.09	1.80 ± 1.08	1.69 ± 1.11	6242. ± 2.	4919. ± 2.	1226. ± 2.
February	9.31 ± 1.28	27.83 ± 1.30	115.22 ± 1.27	1.74 ± 1.09	1.74 ± 1.08	1.63 ± 1.10	11793. ± 2.	8741. ± 2.	1340. ± 2.
March	9.46 ± 1.36	29.23 ± 1.41	123.68 ± 1.31	1.71 ± 1.10	1.74 ± 1.09	1.64 ± 1.11	12575. ± 2.	6951. ± 2.	1395. ± 2.
April	8.17 ± 1.32	31.00 ± 1.48	117.70 ± 1.37	1.69 ± 1.11	1.77 ± 1.09	1.64 ± 1.11	9455. ± 3.	6449. ± 2.	1696. ± 3.
May	7.87 ± 1.33	30.10 ± 1.43	112.40 ± 1.36	1.65 ± 1.11	1.73 ± 1.10	1.57 ± 1.11	11821. ± 2.	6342. ± 2.	1140. ± 3.
June	7.84 ± 1.29	32.29 ± 1.47	109.33 ± 1.37	1.66 ± 1.10	1.77 ± 1.10	1.58 ± 1.11	9316. ± 2.	6359. ± 2.	1887. ± 2.
July	9.17 ± 1.35	39.00 ± 1.48	128.81 ± 1.32	1.68 ± 1.11	1.75 ± 1.11	1.57 ± 1.11	5831. ± 2.	4716. ± 2.	1851. ± 2.
August	15.39 ± 1.38	53.04 ± 1.37	151.92 ± 1.45	1.62 ± 1.11	1.69 ± 1.10	1.49 ± 1.11	3272. ± 2.	3368. ± 2.	707. ± 3.
September	20.46 ± 1.28	54.14 ± 1.32	162.06 ± 1.41	1.59 ± 1.09	1.70 ± 1.09	1.52 ± 1.10	2650. ± 2.	2772. ± 2.	598. ± 2.
October	15.20 ± 1.23	45.56 ± 1.30	173.05 ± 1.40	1.58 ± 1.09	1.71 ± 1.08	1.52 ± 1.12	3940. ± 2.	4050. ± 2.	568. ± 2.
November	14.61 ± 1.24	37.81 ± 1.37	145.80 ± 1.30	1.61 ± 1.08	1.65 ± 1.09	1.57 ± 1.10	6590. ± 3.	3197. ± 3.	714. ± 2.
December	16.40 ± 1.21	42.99 ± 1.30	155.47 ± 1.28	1.59 ± 1.07	1.66 ± 1.09	1.57 ± 1.10	6853. ± 3.	4003. ± 2.	991. ± 2.

2002	D_{pg} [nm]			σ_g			N_{tot} [cm^{-3}]		
	Mode 1	Mode 2	Mode 3	Mode 1	Mode 2	Mode 3	Mode 1	Mode 2	Mode 3
January	15.98 ± 1.27	42.93 ± 1.42	156.36 ± 1.45	1.67 ± 1.08	1.74 ± 1.10	1.59 ± 1.13	5485. ± 4.	3226. ± 3.	838. ± 2.
February	16.03 ± 1.29	41.60 ± 1.34	142.22 ± 1.26	1.64 ± 1.09	1.66 ± 1.09	1.47 ± 1.09	5326. ± 3.	3445. ± 2.	590. ± 2.
March	16.07 ± 1.29	45.23 ± 1.41	142.92 ± 1.29	1.61 ± 1.09	1.69 ± 1.10	1.48 ± 1.09	5099. ± 3.	3340. ± 2.	768. ± 3.
April	16.15 ± 1.33	47.93 ± 1.40	144.72 ± 1.33	1.64 ± 1.10	1.71 ± 1.10	1.52 ± 1.10	6237. ± 2.	4668. ± 2.	1272. ± 3.
May	14.98 ± 1.32	45.95 ± 1.39	127.91 ± 1.33	1.64 ± 1.11	1.69 ± 1.10	1.47 ± 1.08	5464. ± 3.	4448. ± 3.	963. ± 3.
June	13.50 ± 1.33	47.76 ± 1.36	138.70 ± 1.37	1.67 ± 1.11	1.73 ± 1.10	1.49 ± 1.10	3741. ± 2.	3766. ± 2.	948. ± 2.
July	13.69 ± 1.35	52.67 ± 1.40	162.70 ± 1.42	1.67 ± 1.11	1.72 ± 1.09	1.50 ± 1.11	4440. ± 3.	4476. ± 2.	987. ± 3.
August	12.70 ± 1.36	47.89 ± 1.41	152.62 ± 1.42	1.68 ± 1.10	1.76 ± 1.09	1.54 ± 1.12	5075. ± 2.	4329. ± 2.	1258. ± 2.
September	16.05 ± 1.27	49.58 ± 1.37	154.10 ± 1.36	1.57 ± 1.10	1.66 ± 1.10	1.46 ± 1.08	4823. ± 2.	2988. ± 2.	577. ± 3.
October	18.34 ± 1.27	50.24 ± 1.26	177.03 ± 1.23	1.53 ± 1.07	1.64 ± 1.08	1.45 ± 1.08	4577. ± 2.	2478. ± 2.	427. ± 2.
November	20.08 ± 1.16	54.44 ± 1.24	184.44 ± 1.19	1.48 ± 1.07	1.63 ± 1.08	1.44 ± 1.06	3483. ± 3.	2038. ± 3.	466. ± 2.
December	20.26 ± 1.17	54.38 ± 1.24	174.77 ± 1.22	1.51 ± 1.07	1.63 ± 1.07	1.45 ± 1.07	4045. ± 3.	2625. ± 3.	508. ± 2.

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2003	D_{pg} [nm]			σ_g			N_{tot} [cm^{-3}]		
	Mode 1	Mode 2	Mode 3	Mode 1	Mode 2	Mode 3	Mode 1	Mode 2	Mode 3
January	20.54 ± 1.22	52.53 ± 1.25	173.94 ± 1.21	1.54 ± 1.08	1.63 ± 1.07	1.45 ± 1.07	5010. ± 3.	3096. ± 3.	508. ± 2.
February	19.81 ± 1.19	56.91 ± 1.27	174.49 ± 1.29	1.55 ± 1.07	1.66 ± 1.08	1.45 ± 1.08	4086. ± 3.	2778. ± 3.	588. ± 2.
March	--	--	--	--	--	--	--	--	--
April	--	--	--	--	--	--	--	--	--
May	--	--	--	--	--	--	--	--	--
June	--	--	--	--	--	--	--	--	--
July	--	--	--	--	--	--	--	--	--
August	--	--	--	--	--	--	--	--	--
September	--	--	--	--	--	--	--	--	--
October	--	--	--	--	--	--	--	--	--
November	--	--	--	--	--	--	--	--	--
December	--	--	--	--	--	--	--	--	--

Appendix B: Monthly statistical values of the integrated total particle number concentrations ($8 < D_p < 400$ nm) evaluated from the hourly medians

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1997	Min.	5%	25%	**Median**	75%	95%	Max.	**Mean ± Std**	**N**
January	--	--	--	--	--	--	--	--	--
February	--	--	--	--	--	--	--	--	--
March	--	--	--	--	--	--	--	--	--
April	--	--	--	--	--	--	--	--	--
May	3940	7950	12400	16800	23500	38200	57600	19500 ± 9730	593
June	1030	6150	12200	17900	27300	42100	59500	20500 ± 11100	649
July	--	--	--	--	--	--	--	--	--
August	4710	6750	10300	15400	22900	33300	53000	17600 ± 8890	247
September	1130	5550	10600	14700	20700	32200	59400	16500 ± 8630	711
October	1830	6350	13100	21500	32800	47500	59600	23700 ± 12800	711
November	1890	4250	9950	18100	28500	48100	59000	20800 ± 13300	663
December	1700	2850	7250	15300	26500	47400	59700	18800 ± 13900	670

1998	Min.	5%	25%	**Median**	75%	95%	Max.	**Mean ± Std**	**N**
January	1250	3750	8050	13500	25200	44400	59800	18100 ± 13200	650
February	1200	4050	8950	14500	23500	44500	59300	18100 ± 12400	587
March	1870	6450	13300	19500	28900	44100	59200	22100 ± 11800	613
April	3610	6350	11800	17900	26400	39900	59500	20200 ± 10700	687
May	2270	5950	9450	13700	19300	28300	52000	15300 ± 7650	743
June	2290	3750	7450	11100	18100	31000	54100	13800 ± 8920	684
July	2910	4250	6250	9250	12500	19600	40100	10200 ± 5150	579
August	1920	4150	6550	9350	13800	23800	48700	11100 ± 6360	742
September	2330	4250	7250	11100	15500	24900	55300	12400 ± 7130	716
October	1170	3250	6450	10900	16000	27800	50600	12500 ± 7840	697
November	2240	4250	9350	16900	27000	45100	59900	19800 ± 13100	702
December	1850	3750	8250	14200	22500	38200	58600	16900 ± 11200	738

1999	Min.	5%	25%	**Median**	75%	95%	Max.	**Mean ± Std**	**N**
January	1190	4350	9250	15000	25800	47800	59700	18900 ± 12900	703
February	1620	4350	10700	18400	29500	46800	59800	21100 ± 13200	625
March	2590	4950	11100	18800	28400	44100	60000	20900 ± 12200	732
April	2980	6350	10900	15600	23100	36800	58900	18200 ± 9780	714
May	1670	5550	9950	14600	20100	32000	55500	16000 ± 8130	742
June	3040	4950	8650	12300	18300	29200	48700	14200 ± 7510	720
July	2630	4350	6850	9750	13300	21100	39400	10900 ± 5530	744
August	2440	4750	8350	11800	17300	29000	59400	13900 ± 8070	741
September	2290	5050	9250	13300	18700	32700	57400	15300 ± 8870	676
October	1610	3750	8050	12200	19200	34000	53000	14900 ± 9400	724
November	1300	3350	6550	11100	18100	32900	57000	14000 ± 9800	652
December	1150	3250	6650	11700	21600	38500	58000	15400 ± 11200	726

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2000	Min.	5%	25%	**Median**	75%	95%	Max.	**Mean ± Std**	**N**
January	1740	3650	7550	12700	19700	30500	48500	14500 ± 8720	712
February	2220	4450	7850	12300	18800	29400	46900	14200 ± 7990	445
March	2150	4550	9450	15900	24200	38600	59800	17800 ± 10600	712
April	2830	6550	12800	18800	29600	45900	58800	21900 ± 12200	681
May	4770	7050	11400	15400	22600	34200	53700	17700 ± 8580	708
June	3300	6050	9650	13800	20000	32900	58500	16000 ± 8660	690
July	1410	3950	6550	10200	16400	28300	46100	12600 ± 8020	665
August	2480	4650	8350	12200	17100	29100	49500	13800 ± 7400	736
September	3940	6450	9650	14400	19700	33600	56400	16100 ± 8670	652
October	1450	3050	6550	9550	14300	23200	55400	11200 ± 7070	594
November	1020	2650	5050	8950	16700	33400	59900	12400 ± 10300	712
December	1340	2850	5850	9950	17500	30500	55400	12700 ± 9170	709

2001	Min.	5%	25%	**Median**	75%	95%	Max.	**Mean ± Std**	**N**
January	1280	2250	6050	10600	17800	32000	55600	13200 ± 9570	689
February	2610	5250	10900	17600	27100	43600	58700	20200 ± 11900	624
March	2990	5550	10400	15700	25900	43700	58300	19300 ± 11800	718
April	2360	4350	7950	12400	21800	41300	57500	16400 ± 11400	669
May	2880	5650	9450	13500	19300	35500	58100	15900 ± 9400	740
June	3820	5150	8350	13300	20200	39100	59500	16200 ± 10600	712
July	2580	4450	7250	10600	15400	27600	54600	12700 ± 7640	703
August	1320	3450	5250	7350	10900	19000	50600	8980 ± 5500	506
September	1180	2250	4250	6850	9350	15500	39100	7540 ± 4430	576
October	1460	3050	5050	8450	13000	25300	57200	10400 ± 7480	698
November	1050	2450	5250	10500	18800	39800	58800	14000 ± 11600	665
December	1030	2850	6150	12100	21200	41800	59600	15600 ± 12100	737

2002	Min.	5%	25%	**Median**	75%	95%	Max.	**Mean ± Std**	**N**
January	1190	2050	4250	8650	19800	37700	59400	13500 ± 12000	722
February	1300	2650	5050	9150	17400	38100	59100	13100 ± 11000	640
March	1410	3150	6150	9750	15800	32400	59900	12600 ± 9620	737
April	2060	5350	8550	12700	17800	34400	58900	15000 ± 9360	704
May	1690	4250	8050	11800	15400	29300	53100	13200 ± 7760	728
June	2100	3550	5950	8750	12000	20200	36900	9860 ± 5330	688
July	2380	3550	6150	9650	14400	30300	50900	11900 ± 8210	741
August	2650	4050	6750	10500	14800	29800	57000	12200 ± 7910	643
September	1130	3250	5450	8850	13300	26100	55300	10800 ± 7550	705
October	1210	2450	5050	8050	12200	22800	48100	9680 ± 6610	738
November	1040	1550	3650	6750	12200	23400	38000	9000 ± 7050	709
December	1050	1450	3750	8050	15900	32500	57400	11400 ± 9980	694

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2003	Min.	5%	25%	**Median**	75%	95%	Max.	**Mean ± Std**	**N**
January	1080	2150	5150	10700	18200	30700	58500	13000 ± 9930	742
February	1010	1850	4150	8350	16000	30800	52400	11400 ± 9290	548
March	--	--	--	--	--	--	--	--	--
April	--	--	--	--	--	--	--	--	--
May	--	--	--	--	--	--	--	--	--
June	--	--	--	--	--	--	--	--	--
July	--	--	--	--	--	--	--	--	--
August	--	--	--	--	--	--	--	--	--
September	--	--	--	--	--	--	--	--	--
October	--	--	--	--	--	--	--	--	--
November	--	--	--	--	--	--	--	--	--
December	--	--	--	--	--	--	--	--	--

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Table 1. Overall geometric means of the fitting parameters of each mode

	Mode 1 (Nucleation)			Mode 2 (Aitken)			Mode 3 (Accumulation)		
	σ_g	D_{pg} [nm]	N_{tot} [cm^{-3}]	σ_g	D_{pg} [nm]	N_{tot} [cm^{-3}]	σ_g	D_{pg} [nm]	N_{tot} [cm^{-3}]
- Siltavuori									
Sub-mode 1	--	--	--	--	25.9	--	--	--	--
Sub-mode 2	--	--	--	--	43.8	--	--	--	--
Overall	1.71	11.7	7100	1.78	37.3	6320	1.60	150.5	960
- Kumpula									
Sub-mode 1	--	8.9	--	--	24.4	--	--	--	--
Sub-mode 2	--	17.7	--	--	48.1	--	--	--	--
Overall	1.63	13.8	5670	1.71	42.5	4050	1.54	151.8	900

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Table 2. Annual particle number concentrations shown in units of (1000 cm^{-3}). From 1 January 1998 to 5 March 2001 the measurement was in Siltavuori, and after that it has been in Kumpula

Year	Median	Geometric mean	Arethmatic mean
1998	12 993	13 819	17 590
1999	13 587	14 265	17 812
2000	13 042	13 313	16 450
2001	11 418	12 230	16 093
2002	9 553	10 077	13 588

Table 3. Multi-lognormal fitting parameters of overall mean particle number size distributions during different seasons

	Mode 1 (Nucleation)			Mode 2 (Aitken)			Mode 3 (Accumulation)		
	σ_g	D_{pg} [nm]	N_{tot} [cm^{-3}]	σ_g	D_{pg} [nm]	N_{tot} [cm^{-3}]	σ_g	D_{pg} [nm]	N_{tot} [cm^{-3}]
- Siltavuori									
Working-days									
Season 1 (Winter)	1.90	11.0	11126	1.90	30.8	11690	1.79	135.0	1246
Season 2 (Spring)	1.90	11.4	14360	1.90	35.2	10358	1.90	115.0	1717
Season 3 (Summer)	1.69	11.4	6059	1.90	37.6	8584	1.83	118.0	1486
Season 4 (Autumn)	1.62	11.7	5391	1.90	31.6	10339	1.90	109.0	1812
Weekends									
Season 1 (Winter)	1.90	10.9	7255	1.90	35.7	6545	1.72	155.0	878
Season 2 (Spring)	1.90	9.7	9750	1.90	33.8	7642	1.90	104.0	2189
Season 3 (Summer)	1.77	11.1	4196	1.90	42.0	6158	1.84	110.0	1173
Season 4 (Autumn)	1.87	12.5	5588	1.90	45.2	6098	1.65	158.0	749
- Kumpula									
Working-days									
Season 1 (Winter)	1.80	19.5	10128	1.80	50.2	2124	1.80	150.0	801
Season 2 (Spring)	1.80	17.0	8999	1.80	58.5	3554	1.80	101.0	900
Season 3 (Summer)	1.80	14.5	5847	1.80	55.4	4000	1.80	126.0	1300
Season 4 (Autumn)	1.65	17.0	7100	1.65	50.0	2701	1.80	130.0	901
Weekends									
Season 1 (Winter)	1.65	17.5	3581	1.65	55.0	1705	1.53	191.0	457
Season 2 (Spring)	1.75	17.5	3900	1.75	55.5	3950	1.57	172.0	543
Season 3 (Summer)	1.80	15.5	2600	1.70	55.0	3000	1.75	125.0	1200
Season 4 (Autumn)	1.80	17.0	3700	1.65	55.0	1801	1.65	140.0	600

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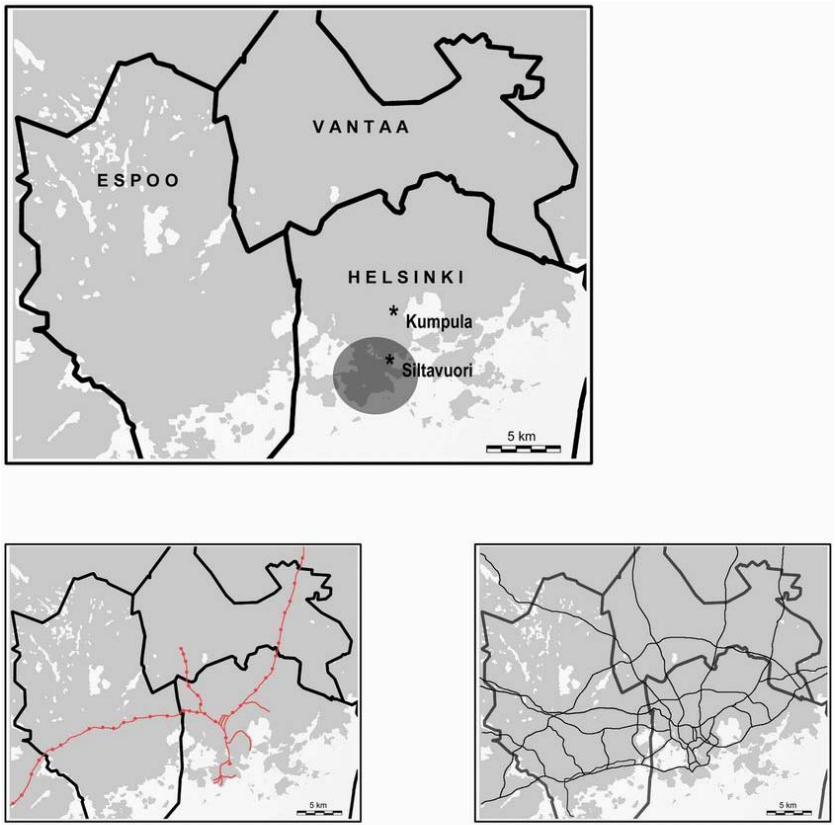

Fig. 1. A map of the Helsinki metropolitan area. The measurement site locations are indicated with a star in the (upper), and the dark circle is the downtown area. (Lower left) shows the railway routes, and (lower right) shows the main roads and highways. A more detailed map can be found from the Helsinki Area Council (YTV) webpage (http://ilma.ytv.kaapeli.fi/english/research/area_map.php).

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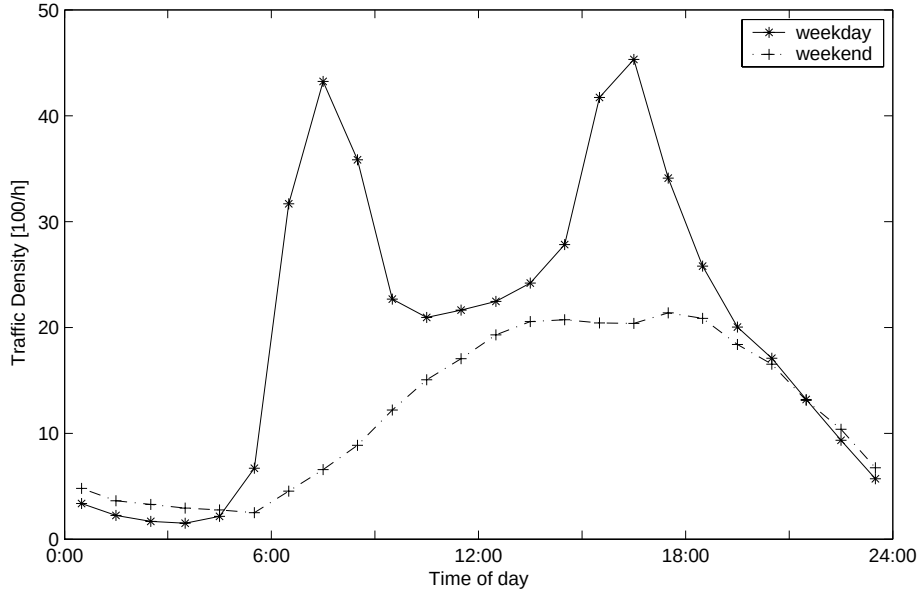

Fig. 2. Typical traffic density on a high way in Helsinki metropolitan area.

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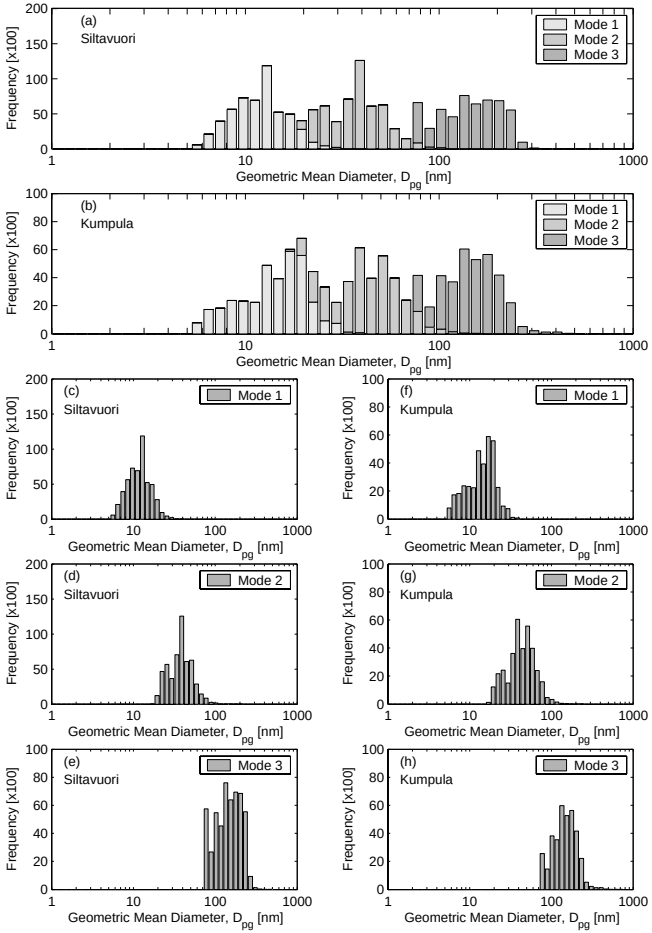

Fig. 3. Overall cumulative frequency plots of the geometric mean diameter (D_{pg}) of the three fitted modes **(a)** in Siltavuori, **(b)** in Kumpula. The cumulative frequency plots for each individual mode are presented in **(c)–(e)** in Siltavuori and **(d)–(f)** in Kumpula.

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Urban aerosol number size distributions

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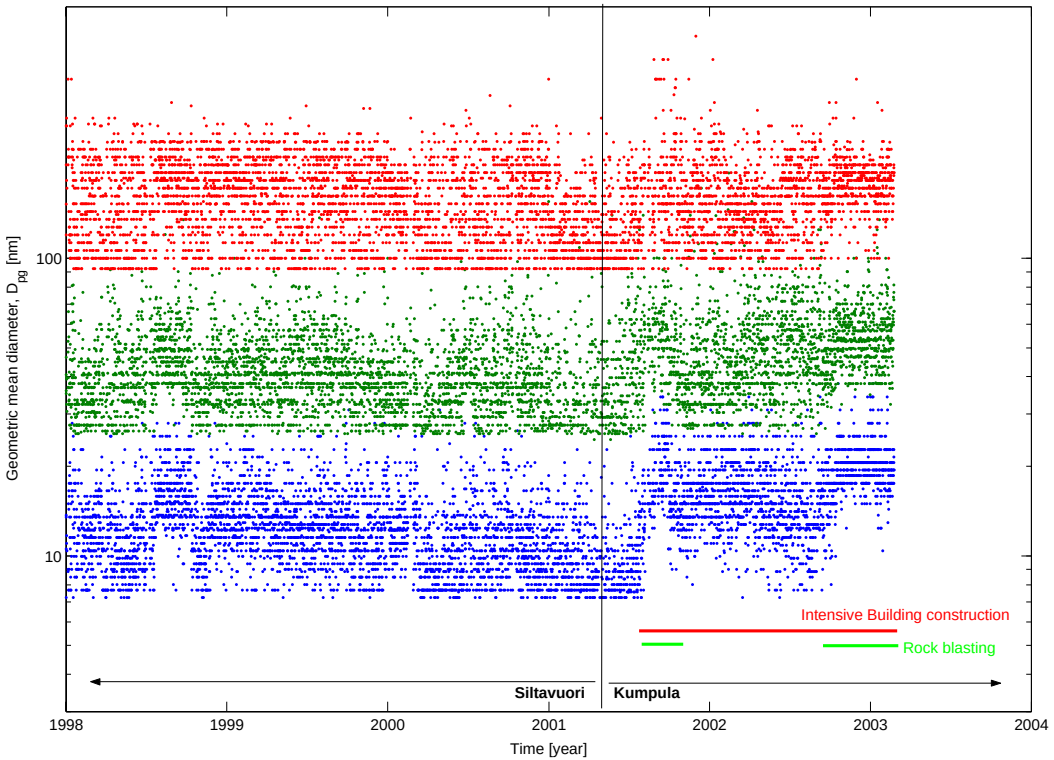

Fig. 4. The geometric mean diameters of three modes as a function of time in Siltavuori and Kumpula. The start of building construction is also indicated as well as the intensive rock blasting periods.

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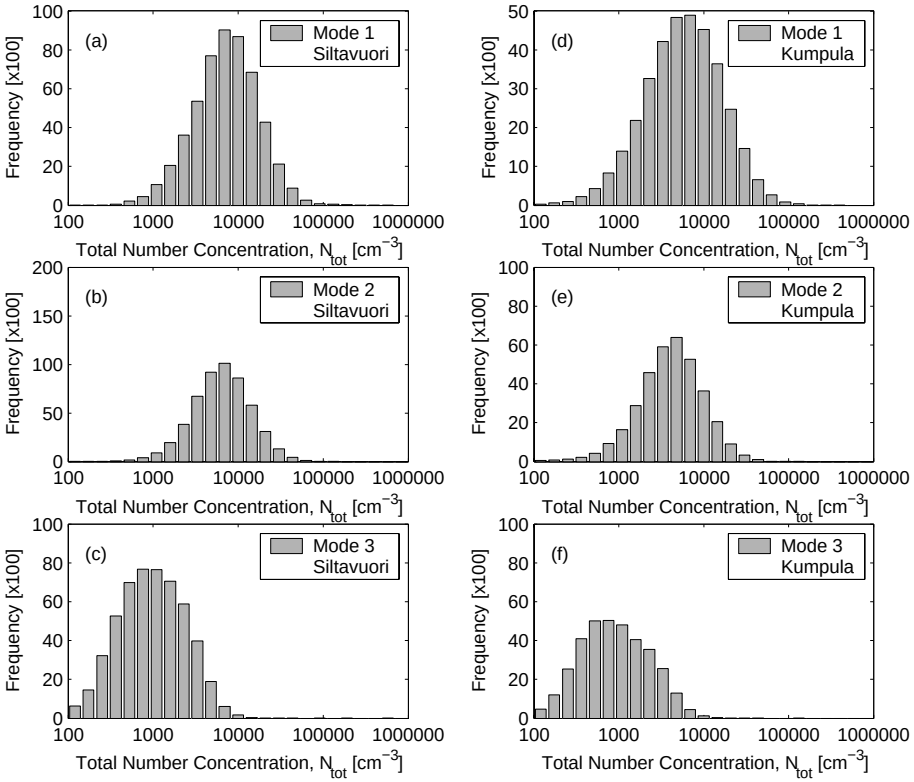

Fig. 5. Overall cumulative frequency plots of the total number concentration (N_{tot}) of each individual mode in Siltavuori **(a)–(c)** and in Kumpula **(d)–(f)**.

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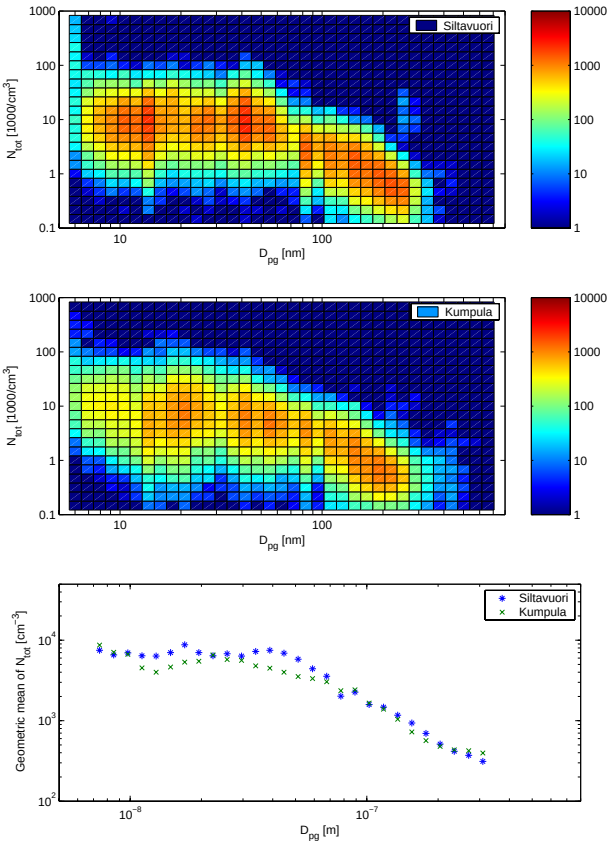

Fig. 6. Overall cumulative frequency plots within a certain total number concentration range (ΔN_{tot}) and a geometric diameter range (ΔD_{pg}) of the fitted modes **(a)** in Siltavuori and **(b)** Kumpula. **(c)** is the geometric mean of the total particle number concentration (N_{tot}) as a function of the geometric mean diameter (D_{pg}).

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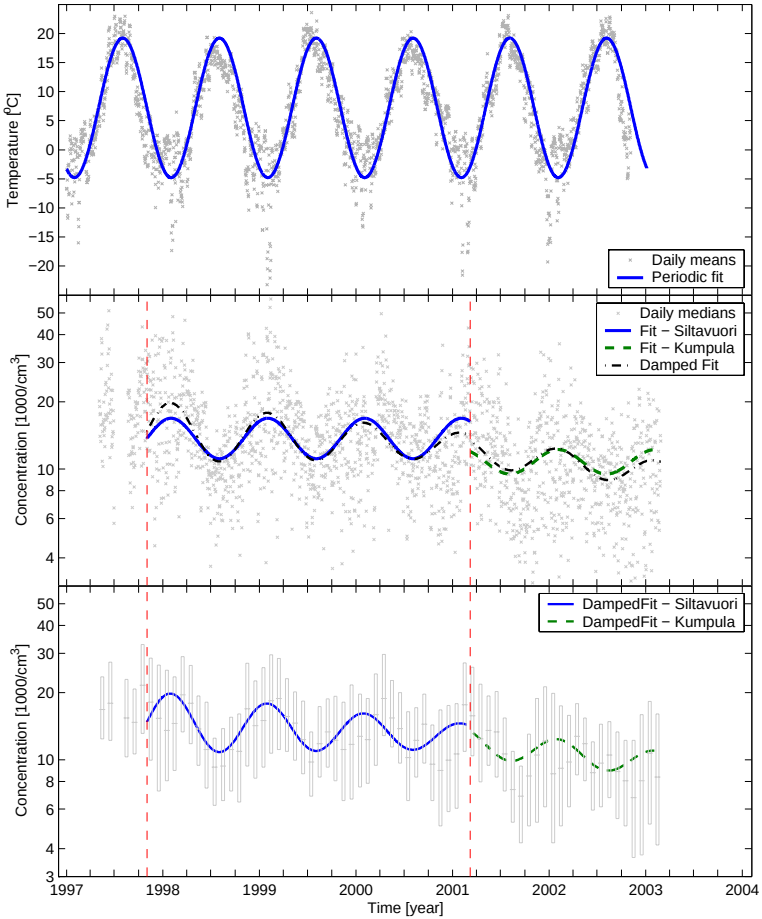

Fig. 7. (a) Annual variation of the temperature. Annual variation of the Integrated total particle number concentration (8 nm< D_p < 400 nm) presented as **(b)** daily medians and **(c)** monthly medians (with 25% as the lower limit of the box plot and 75% as the upper limit).

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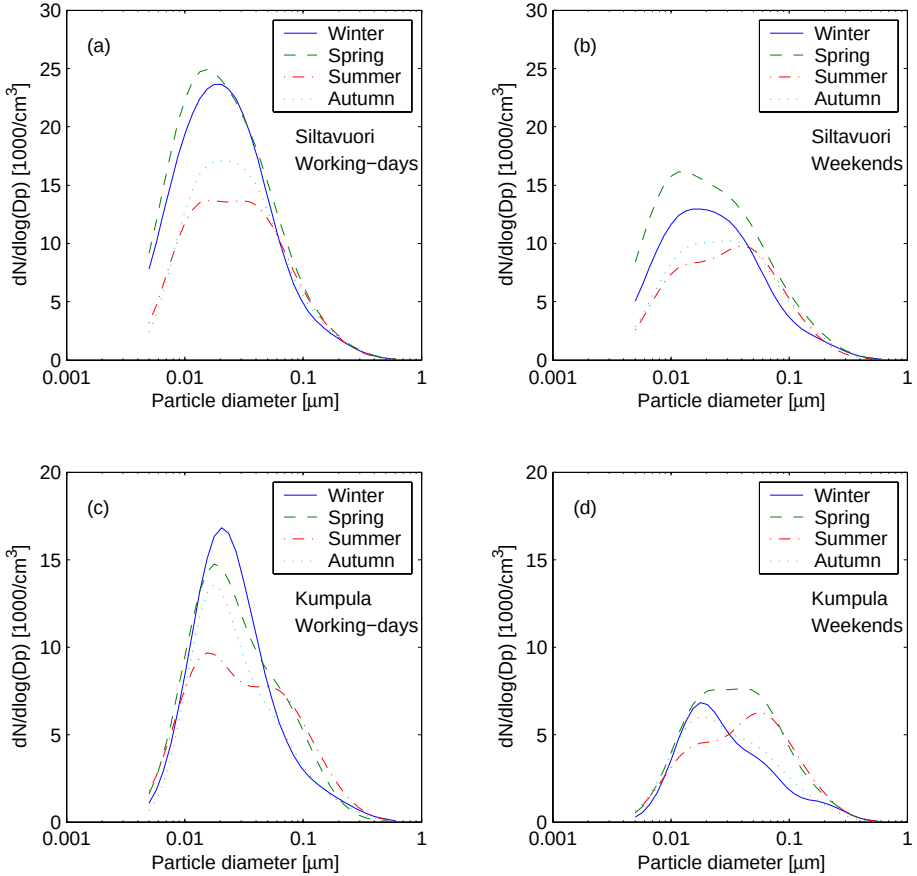

Fig. 8. Overall seasonal particle number size distributions presented mutli-lognormal fitting of the medians in Siltavuori for **(a)** workdays and **(b)** weekends. **(c)** and **(d)** are the same but for Kumpula.

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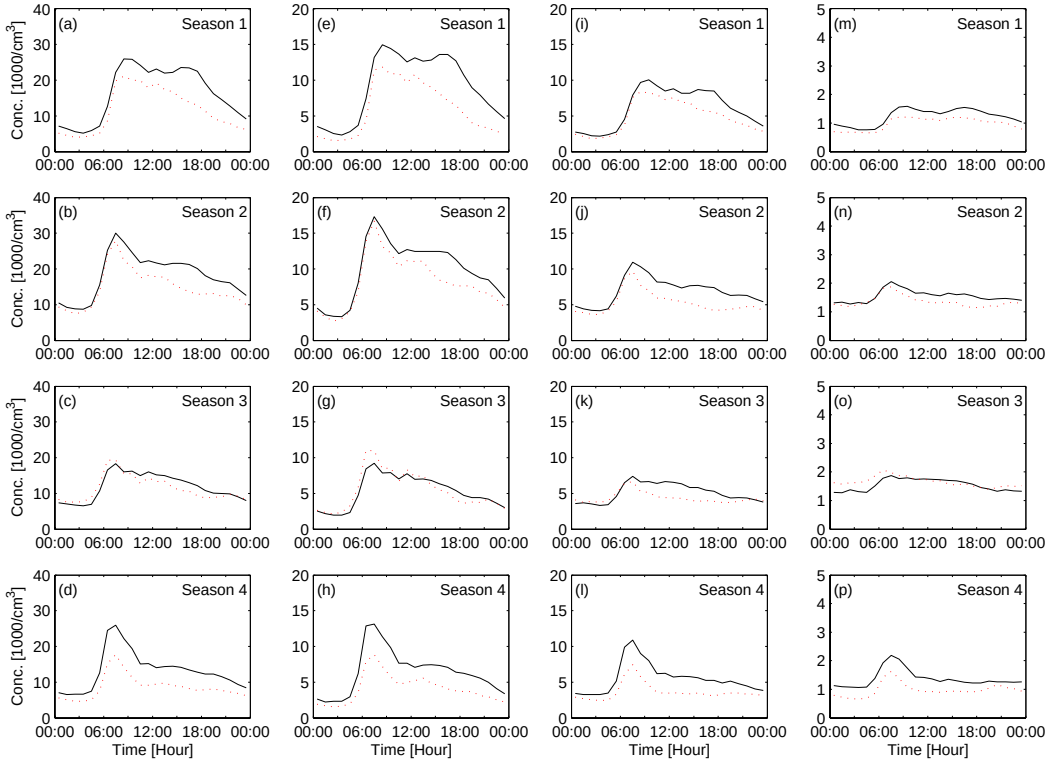

Fig. 9. Overall diurnal patterns of the integrated total particle number concentration for workdays during the different seasons. Legend: solid lines are in Siltavuori and dotted lines are in Kumpula. First column is the integrated total particle number concentration, second column is the nucleation mode, third column is the Aitken mode, and fourth column is the accumulation mode.

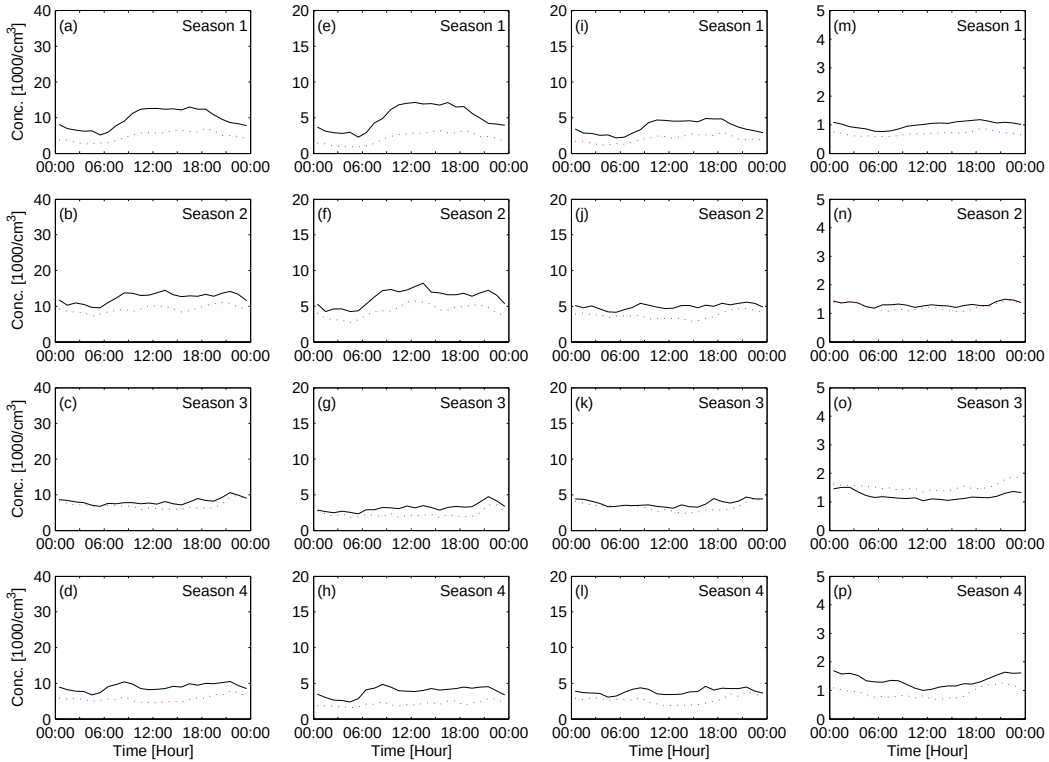

Fig. 10. Overall diurnal patterns of the integrated total particle number concentration for weekends during the different seasons. Legend: solid lines are in Siltavuori and dotted lines are in Kumpula. First column is the integrated total particle number concentration, second column is the nucleation mode, third column is the Aitken mode, and fourth column is the accumulation mode.