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Evaluation and modeling of the size fractionated aerosol particle number concentration measurements nearby a major road in Helsinki – Part II: Aerosol measurements within the SAPPHIRE project

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Abstract. This study presents an evaluation and modeling exercise of the size fractionated aerosol particle number concentrations measured nearby a major road in Helsinki during 23 August–19 September 2003 and 14 January–11 February 2004. The available information also included electronic traffic counts, on-site meteorological measurements, and urban background particle number size distribution measurement. The ultrafine particle (UFP, diameter <100 nm) number concentrations at the roadside site were approximately an order of magnitude higher than those at the urban background site during daytime and downwind conditions. Both the modal structure analysis of the particle number size distributions and the statistical correlation between the traffic density and the UFP number concentrations indicate that the UFP were evidently from traffic related emissions. The modeling exercise included the evolution of the particle number size distribution nearby the road during downwind conditions. The model simulation results revealed that the evaluation of the emission factors of aerosol particles might not be valid for the same site during different time.

1 Introduction

There is a strong indication that adverse health effects of particulate matter (PM) are not related only to aerosol particle mass concentration, but instead, related to many other aerosol properties including particle number concentration and chemical composition (e.g. Penttinen et al., 2001; Osunsanya et al., 2001). In addition, the state (liquid or solid), volatility, hygroscopicity, morphology, and density of particles can be considered important.

Regionally, new particle formation contributes a major part to ultrafine particle (UFP, diameter <100 nm) number concentrations (Kulmala et al., 2004). In Nordic countries, long-range transport (LRT) of aerosol particles constitutes an important part of the total urban background $PM_{2.5}$ (e.g. Karppinen et al., 2004; Johansson et al., 1999). In addition to LRT, aerosol particles in urban areas are originated from several local sources, most frequently vehicular traffic and industrial emissions. In urban areas, the temporal and spatial variation of both the particle number and PM_{10} concentrations are commonly closely related to local meteorology and traffic flows. These dependencies are moderate for $PM_{2.5}$ as they are caused mainly by the substantial LRT background (e.g. Pohjola et al., 2000, 2002, 2003; Laakso et al., 2003).

Vehicular dispersion models are essential computational tools in modern municipal and urban planning. For model

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evaluation, air quality data have to be combined with simultaneous measurements of traffic flow and meteorological parameters as well as information about local topography and buildings (Luhar and Patil, 1989; Benson, 1992; Kukkonen et al., 2001; Oettl et al., 2001; Tiitta et al., 2002; Levitin et al., 2005). Evaluation of roadside dispersion models is seriously hindered by the scarcity of appropriate good-quality experimental datasets (e.g. Kukkonen et al., 2001). However, few studies addressed size-fractionated aerosol particles such as Wåhlin et al. (2001) and Ketzel et al. (2003) who developed inverse modeling tools to evaluate the emission factors of UFP originating from various pollution sources at various urban and rural locations in Denmark. Pohjola et al. (2003) and Vignati et al. (1999) combined vehicular plume models with particle transformation models (i.e. aerosol dynamics models) in order to assess the impacts of coagulation, condensation of water vapour, and plume dilution on the particle number size distribution. Pohjola et al. (2003) also used a simple plume model combined with a monodisperse aerosol dynamic model that included chemical composition of particles in order to study the importance of aerosol processes during dispersion near a road. Dilution ratios of aerosol particles can be also obtained from empirical CO dilution ratios when the evolution of aerosol particles is dominated by dilution, condensation, and evaporation processes and the effects of coagulation and deposition are minor (Zhang and Wexler, 2004).

The main objectives of this study (Part II) are to investigate, evaluate, and simulate size-fractionated aerosol particle number concentrations measured within the SAPPHIRE ("Source Apportionment of Airborne Particulate Matter and Polycyclic Aromatic Hydrocarbons in Urban Regions of Europe", 2003–2006) project in Helsinki. The size-fractionated aerosol measurements were performed simultaneously at a roadside site and a background site during 23 August–19 September 2003 and 14 January–11 February 2004. Traffic densities were also measured electronically and meteorological parameters were measured on-site. The measured data sets can be utilized to investigate the physical characteristics of fine particulate matter originating from vehicular traffic combustion emissions. We also used these data sets to perform a model exercise, which included an aerosol dynamics model (UHMA, Korhonen et al., 2004) and dispersion model, to simulate the particle number size distribution nearby a road. Part I of this study discussed the model performance and development in more details (Pohjola et al., 2007).

2 Materials and methods

2.1 Measurement sites

Helsinki Metropolitan Area and its surrounding regions are situated on a fairly flat coastal area by the Baltic Sea at the latitude of 60° N. The climate is relatively mild compared with many other regions of the same latitudes, mainly due to the prevailing global atmospheric circulation and the Gulf Stream. The Helsinki Metropolitan Area comprises four cities (Helsinki, Espoo, Vantaa and Kauniainen) with a total area of 743 km^2 and population of approximately one million.

The measurements were performed simultaneously at two locations: an urban background station (Kumpula) and a roadside site (Herttoniemi), as presented in Fig. 1. The roadside site was located about 6 km east of the centre of Helsinki. The surrounding area was suburban, with substantial local traffic. In the vicinity of the site, there was a major highway (Itäväylä) at a distance of 65 m, as measured from the center line of the road (Fig. 1). The measurement site was located in a fairly flat homogeneous terrain; there were no obstacles towards the direction of the major road (Itäväylä) except for few low buildings (including a plastic factory) and trees that shielded a part of the major road. The plastic factory occasionally provided emissions of aerosol particles during some time periods, and consequently, these time periods were excluded from the analysis when the prevailing wind was from that direction.

for comparison purposes, we utilized a part of the continuous measurements at the urban background measurement station that was located on the third floor of the Department of Physical sciences (University of Helsinki). The department building was located on a hilltop of approximately 20 m height from a nearby Major road (Lahdentie, about 200 m to the east). The area between this highway and the measurement site is forested (∼10 m high); this partly prevents the transport of the traffic emissions to this direction. The department building was located 5 km northeast the centre of Helsinki and it was surrounded by residential and university buildings situated on the northeastern side.

2.2 Measurements of particle number size distributions

The aerosol particle measurements consisted of high-time resolution of particle number size distributions (dry size between 8–320 nm at the urban background station and between 3–800 nm at the roadside site). The aerosol measurements at the background station were performed with a differential mobility particle sizer (DMPS, Aalto et al., 2001) and a twin scanning mobility particle sizer (SMPS) at the roadside site. The differential mobility analysis of aerosol particles relies on bipolar charging of aerosol particles (Liu and Pui, 1974) followed by classification of aerosol particles according to their electrical mobility by a differential
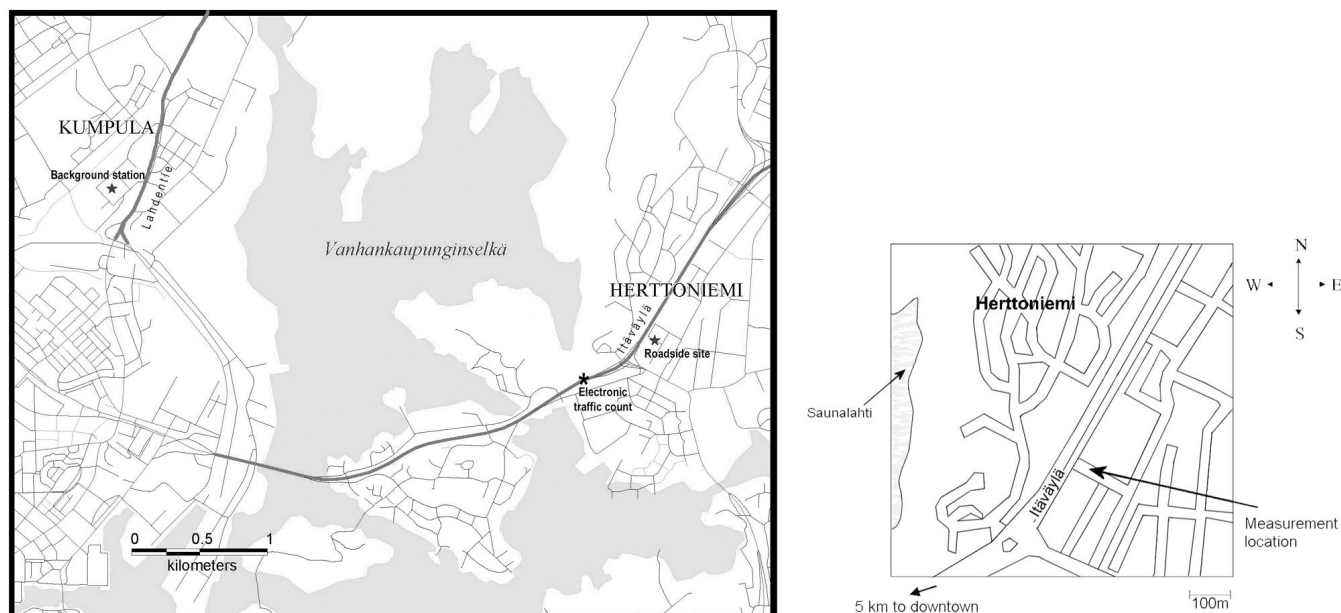
Fig. 1. Measurement sites and traffic count locations (marked with stars) with a detailed map nearby the roadside Itäväylä. The map also shows the road and street network; the nearby major roads are indicated with dark lines. Grey area is a part of the Baltic Sea. This map was provided by Ground map material (C) National Land Survey of Finland, license number 158/MYY/06.

mobility analyzer (DMA, Knutson and Whitby, 1975) and then counting the aerosol particle number concentrations with a condensation particle counter (CPC3025, TSI, Inc.) according to Stolzenburg and McMurry (1991) or (CPC 3010, TSI, Inc.) according to Quant et al. (1992).

The sample flow rate in the DMPS was 1.5 liters per minute (lpm) and the sheath flow was 8.5 lpm circulated back to the DMA (electrostatic Classifier TSI 3071) after drying and filtering (Jokinen and Mäkelä, 1997; Birmili et al., 1999). The sheath flow dryer acted as a pulsation damper. The sampling line was approximately 2-m long Stainless Steel tube with 4-mm inner diameter. No special inlet was used, just a rain cover. The waiting time after the DMA voltage change was 15 s after which aerosol particles were counted over a three second period. One measurement cycle with the DMPS took five minutes. The particle number size distributions were extracted from the measured mobility size distributions by data inversion. The data inversion was done by using the MatLab non negative least squares (NNLS) algorithm. We used the transfer function of the classifier according to Stolzenburg (1988), and we defined the particle charging probability according to the semi-empirical functions by Wiedensohler (1989).

The twin SMPS system consists of two differential mobility analyzers. the first unit measured particle number size distributions between 3–50 nm and it consisted of an ultrafine DMA (HAUKE-type, 10.9 cm in length) and an ultrafine CPC TSI 3025. The sample flow rate in this unit was 3 lpm and the sheath flow was 17 lpm. The second unit measured

particle number size distributions between 10–800 nm and it consisted of a DMA (HAUKE, type 28 cm in length) and a CPC TSI 3010. The sample flow in this unit was 1.0 lpm and the sheath flow was 5 lpm. The sheath flows were also arranged as a closed loop with a drier and a filter. Sampling lines were 1-m long metal tube with 4-mm inner diameter and flow rate of 4 lpm. There was no special inlet in the main sample line. One measurement cycle with the SMPS took one minute.

The CPC calibration included particle detection efficiency calibration and the concentration calibration against an aerosol electrometer. DMA calibration included transport loss calibration and the sizing accuracy calibration with PSL particles. We estimated the losses in the transport lines and inside the DMA from the normal laminar flow tube diffusion loss equations. In the weekly maintenance of both the DMPS and the SMPS, aerosol and sheath flows were measured with a bubble flow meter and the CPC zero was checked by turning the DMA voltage to zero. The yearly maintenance of the DMPS included CPC and DMA calibration and thorough cleaning.

We compared both instruments before and after the measurement campaign; the differences in the measured particle size distributions (within the common size range of the measurement 8–320 nm) were less than 5%. These differences were smaller than the concentration differences measured at both sites during the daytime.

Because the measurement at the roadside site (SMPS instrument) was 1-min time resolution whereas the one at the

background site (DMPS instrument) was 5-min time resolution, we calculated the 5-min averages of the particle number size distributions measured at the roadside site. We also considered the particle size range between 8–320 nm for comparison purposes.

2.3 Meteorology

The on-site meteorological measurements at the roadside station (Herttoniemi) included temperature, relative humidity, and wind speed and direction. We measured the meteorological parameters (1-min time resolution) with MILOS500 (Vaisala) automatic weather station. Temperature and relative humidity were measured at 5 m height from the ground whereas wind speed and direction along with temperature were measured at 8 m height from the ground. We used this on-site meteorological data for the wind sector analysis to define the downwind and upwind cases.

2.4 Traffic density

Traffic densities were available from the Helsinki City Planning Department. The traffic densities on the nearest major roads (Itäväylä and Lahdentie) were similar with respect to the average traffic density and the temporal variation. The daily traffic density on Itäväylä varied between 32 000–54 000 vehicles per day (Fig. 2). Based on the traffic density analysis during year 2001, about 85% were light duty vehicles (of which 11% were diesel), about 12% were vans (of which about 84% diesel), and about 4% heavy duty vehicles. The traffic rush hours were between 07:00–10:00 and 15:00–18:00 on workdays.

2.5 Modeling

2.5.1 Modal structure analysis: multi-lognormal fitting

It is often convenient and mathematically straightforward to describe the particle number size distribution with a sum of several lognormal modes each characterized by D_{pg} (geometric mean diameter), σ_g (geometric standard deviation), and N (number concentration);

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

where the left-hand-side represents the measured particle number size distribution and the right-hand-side is the multi-lognormal distribution function (e.g. Whitby, 1978; Seinfeld and Pandis, 1998).

In this study, we used a modified version of our automatic fitting algorithm (DO-FIT), which has been developed at the University of Helsinki (Hussein et al., 2005a), to mathematically fit the measured particle number size distributions to the multi-lognormal distribution function Eq. (1). The modified fitting algorithm first analyzes the measured particle number size distribution in order to decide how many modes (maximum 5 modes) are suitable to fit the measured particle number size distribution. The algorithm automatically makes an initial guess for the modal parameters after which they are re-fined to obtain the best-fit curve. The algorithm uses the least-square minimization method to control the fitting quality. The recent modifications to the DO-FIT algorithm were to optimize and enhance its performance. The original algorithm could accurately fit about 80% of the data set discussed by Hussein et al. (2004), whereas the modified algorithm is capable of fitting more than 90% of the same data set.

We applied this method to investigate the modal structure of the particle number size distribution nearby the roadside site and compare the results to the background conditions at the urban background site.

2.5.2 Modeling the evolution of particle number size distributions

Part I (Pohjola et al., 2007) of this study is devoted to the model development to describe the evolution of particle number size distribution in the vicinity of a major road. According to this modeling scheme, a dispersion model is needed along with an aerosol dynamic model to addresses the expanding air parcels while transported from the road to the background atmosphere. The original aerosol dynamics module is MONO32 (Pirjola and Kulmala, 2000; Pirjola et al., 2003). Here we utilized a modified version of the aerosol dynamic model UHMA (Korhonen et al., 2004) instead of MONO32. The UHMA model simulates the time evolution of particle size and composition distributions; it includes mathematical treatments for Brownian coagulation, and condensation of sulphuric acid, organic vapors, and water. It is possible with the UHMA model to simulate the particle number size distributions with a sectional or modal approach. Following we briefly describe the model approach to simulate the particle number size distribution nearby a road.

The required input parameters for this modeling scheme to simulate the evolution of the particle number size distribution nearby the road side are:

- initial height of the mixing volume according to Pirjola et al. (2006),

- particle number emission factors according to Pohjola et al. (2004, 2007),

- background particle number size distribution (measured in this study),

- rate of pollutants dilution, i.e. dispersion equation obtained from the CAR-FMI model, and
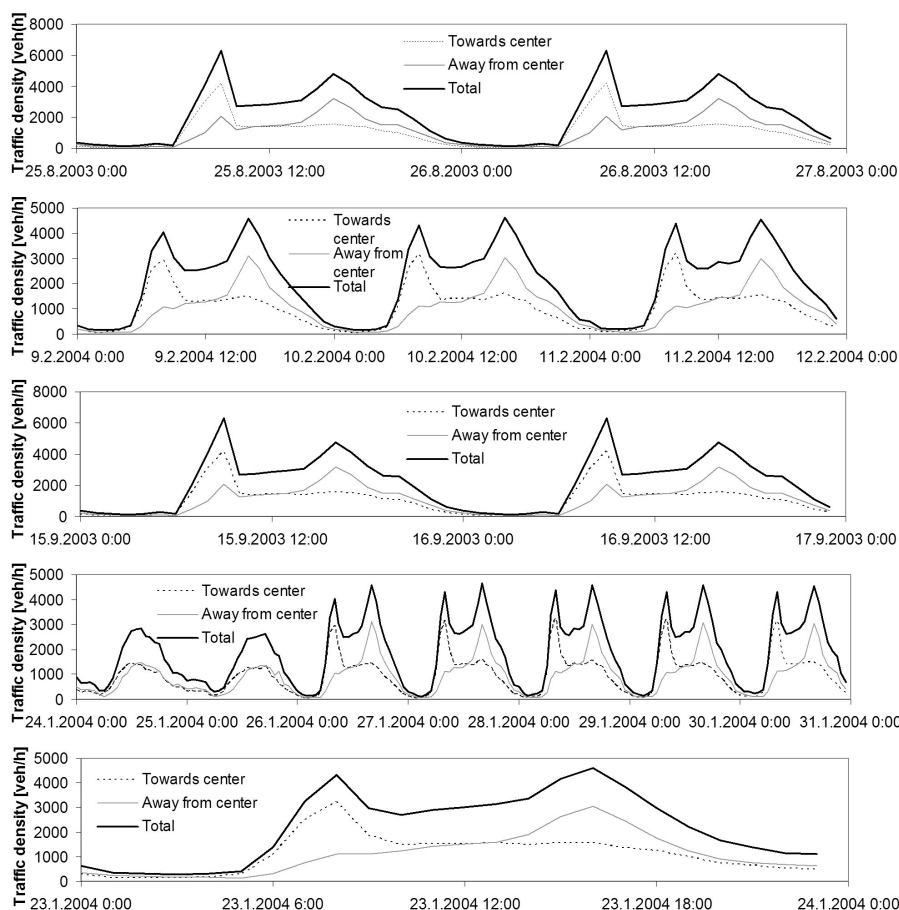
- measured traffic densities.

Fig. 2. Traffic densities on the major road in the vicinity of the roadside site at Herttoniemi. The time periods correspond to the selected cases I–V.

Here, the initial height of the mixing volume corresponds to the situation where vehicular exhaust gases and particles are initially diluted in a time scale of approximately 0.5 s after their discharge from the tailpipe (Pirjola et al., 2006). The initial mixing of aerosol particles is assumed to be dependent on traffic-induced turbulence. Traffic-induced turbulence can be obtained from the CAR-FMI model that utilizes a semi-empirical treatment based on the General Motors experiments (Petersen, 1980).

No emission measurements or modeling were performed within the SAPPHIRE campaign, which is presented and discussed in this article. However, the emissions factor of the particle number concentration from vehicular traffic was previously evaluated, but during different time periods, in Part I (Pohjola et al., 2007) within the LIPIKA campaign that was conducted on the same site (e.g. Pirjola et al., 2004). These evaluations were performed by using measured concentrations and inverse dispersion modeling. Due to the availability of the particle number concentrations from the LIPIKA studies for this site, the model simulations in this study were limited to the winter cases. Although the LIPIKA campaign

was not conducted during the same period as the cases considered in this study, the emission factors of aerosol particles were used here after scaling them with respect to the measured traffic densities.

The nocturnal time period between 01:00 and 03:30 was assumed to be representative for the urban background particle number size distribution because the local traffic density was lowest during that time (Hussein et al., 2005b).

The dispersion equation of aerosol particles from traffic exhaust was first extracted from the CAR-FMI model. The CAR-FMI model includes an emission model, a dispersion model, and statistical analysis of the computed time series of concentrations. The CAR-FMI model utilizes the meteorological input data evaluated with the meteorological pre-processing model MPP-FMI. The dispersion equation is based on a semi-analytical solution of the Gaussian diffusion equation for a finite line source (Luhar and Patil, 1989). For a more detailed description of these models, the reader is referred to, e.g., Härkönen et al. (2002) and Karppinen et al. (2000a, b).
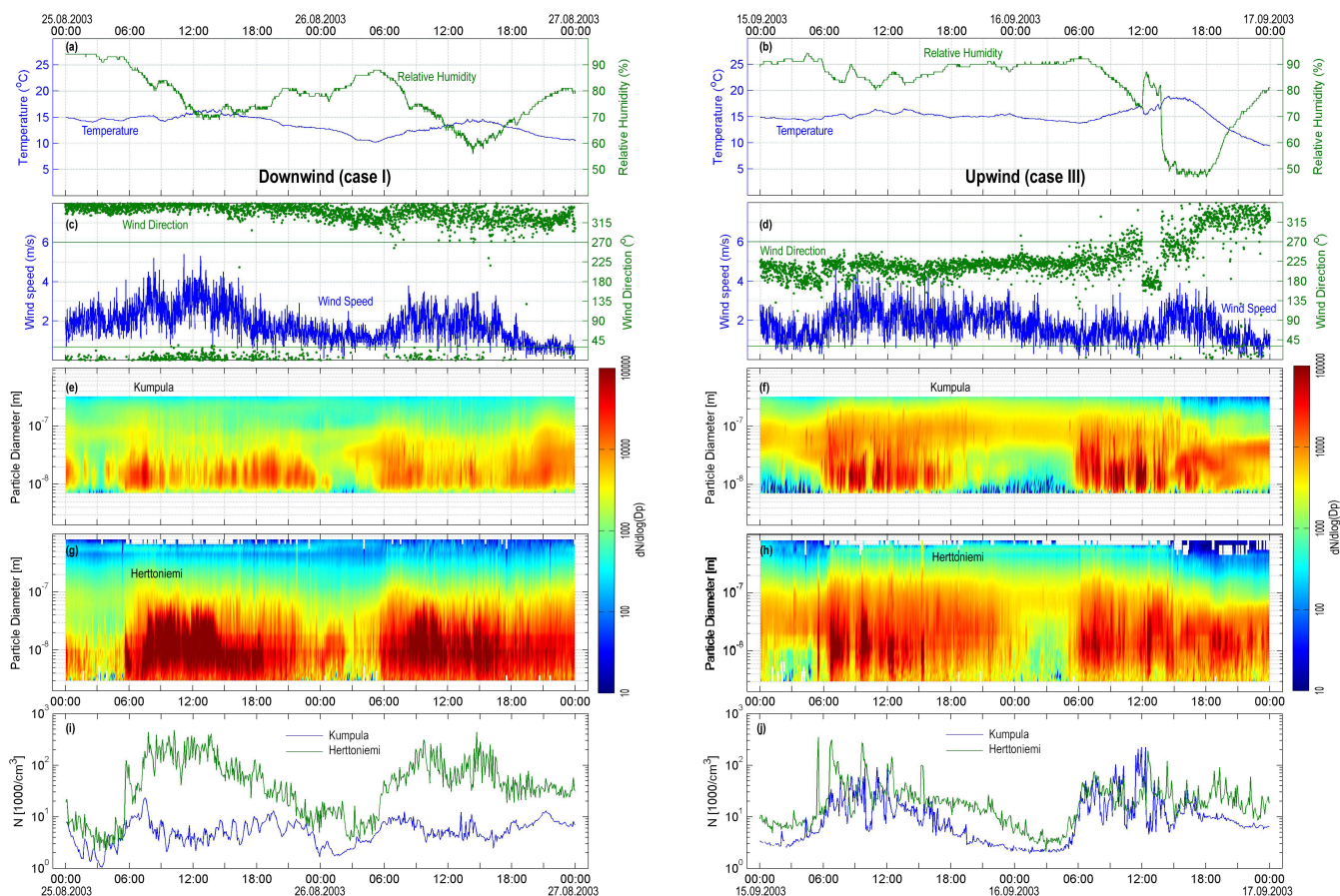
Fig. 3. Cases I and III during the autumn season: downwind condition (left panel) and upwind condition (right panel) at the roadside site (Herttoniemi). **(a–d)** Relevant weather conditions, **(e–f)** particle number size distributions at the urban background site (Kumpula), **(g–h)** particle number size distributions at the roadside site (Herttoniemi), and **(i–j)** particle number concentrations between 8–320 nm.

Table 1. Selected cases of downwind and upwind conditions at the roadside site. WD = wind direction, WS = wind speed, T = ambient temperature, and RH = relative humidity.

	Time period	WD	WS [m/s]	T [°C]	RH%	
Case I	23–28 Aug 2003	−90–30	<4	10–15	60–95	Downwind
Case II	9–11 Feb 2004	−90–0	<5	−15– −4	55–92	Downwind
Case III	14–16 Sep 2003	170–235	<4	14–17	70–95	Upwind
Case IV	24–30 Jan 2004	45–270	<5	−7.5– −2.5	75–95	Upwind

3   Results and discussion

3.1   Downwind versus upwind nearby a roadside

Based on the wind sector analysis (not shown in this article), we considered time periods when the prevailing wind was either downwind or upwind at the roadside site. Furthermore, in order to eliminate the wind direction parameter, we constrained these time periods for rather constant wind direction. As a result, we obtained four time periods: two upwind

cases and two downwind cases. Table 1 presents the average weather conditions as measured at the roadside site. Cases I and III were in the autumn season (Fig. 3) whereas cases II and IV were in the winter season (Fig. 4).

As expected, the daily pattern of the total particle number concentrations at the roadside site was closely similar to that of the traffic density and the relationship was more pronounced during downwind conditions. Furthermore, the total particle number concentrations at the roadside site were, in general, comparable to those at the background site during
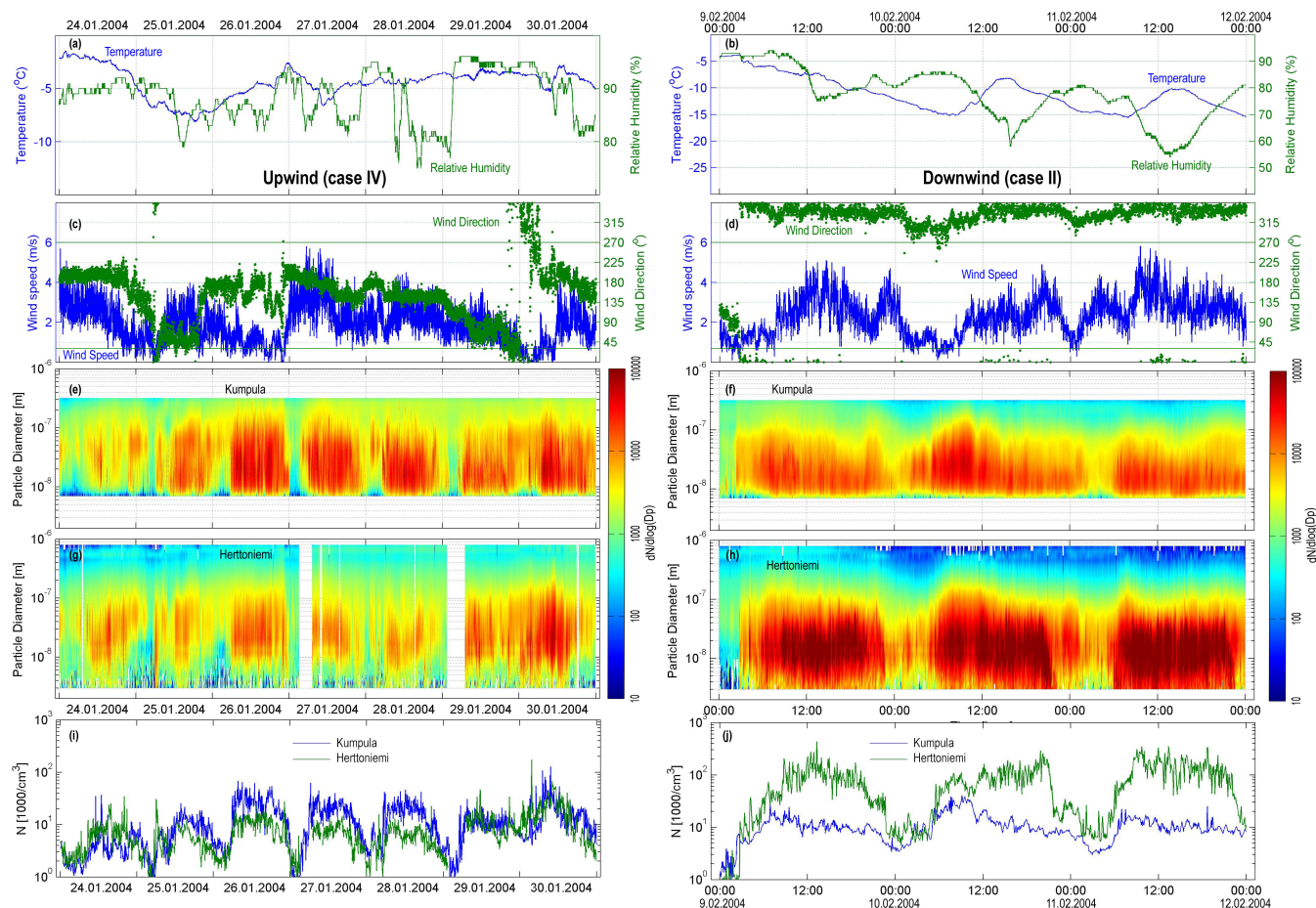
Fig. 4. Cases II and IV during the winter season: upwind condition (left panel) and downwind condition (right panel) at the roadside site (Herttoniemi). **(a–d)** Relevant weather conditions, **(e–f)** particle number size distributions at the urban background site (Kumpula), **(g–h)** particle number size distributions at the roadside site (Herttoniemi), and **(i–j)** particle number concentrations between 8–320 nm.

nighttime and upwind conditions. Otherwise, the total particle number concentrations during downwind and daytime (i.e. traffic rush hours) were approximately an order of magnitude higher at the roadside site in comparison with the background site.

The median particle number size distributions at the background site clearly showed that the number concentrations of UFP were higher during the winter than during the autumn (Fig. 5). This is expected from our previous findings in Helsinki (e.g. Hussein et al., 2004). This was also observed at the roadside site for cases I and II but not for cases III and IV. We, however, believe that case IV is not an ideal situation because the particle number concentrations at the road site were either similar or lower than those observed at the background site during the same January 2004 (Figs. 4 and 5). This means that our background site is not always representative for the urban background conditions during some periods when the site is directly influenced by traffic combustion emissions on the nearby major road (Lahdentie). In fact, the traffic influence was not clearly observed dur-

ing early autumn period only because of the trees located between the site and the major road shielding the site from the traffic emissions. During summer, late spring and, early autumn these trees have enough leaves to act as an active deposition surface for UFP and significantly reduces their concentrations before they arrive to the background site. In contrast, during the winter season, the deciduous trees allowing the background site to be directly influenced by the traffic emissions on the nearby road. Indeed, we still think that cases I–III are valid for our analysis because during downwind conditions (Cases I and II) the background site was not influenced by traffic or industrial emissions. Case III was during the early autumn season when the trees had enough trees to shield the background site from the direct influence by the traffic emissions.

The road contribution to the particle number concentration (defined as the difference between the concentrations at the roadside and the urban background site for the downwind cases) is substantial during the daytime, and especially high during the traffic rush hours (Figs. 3 and 4). The number
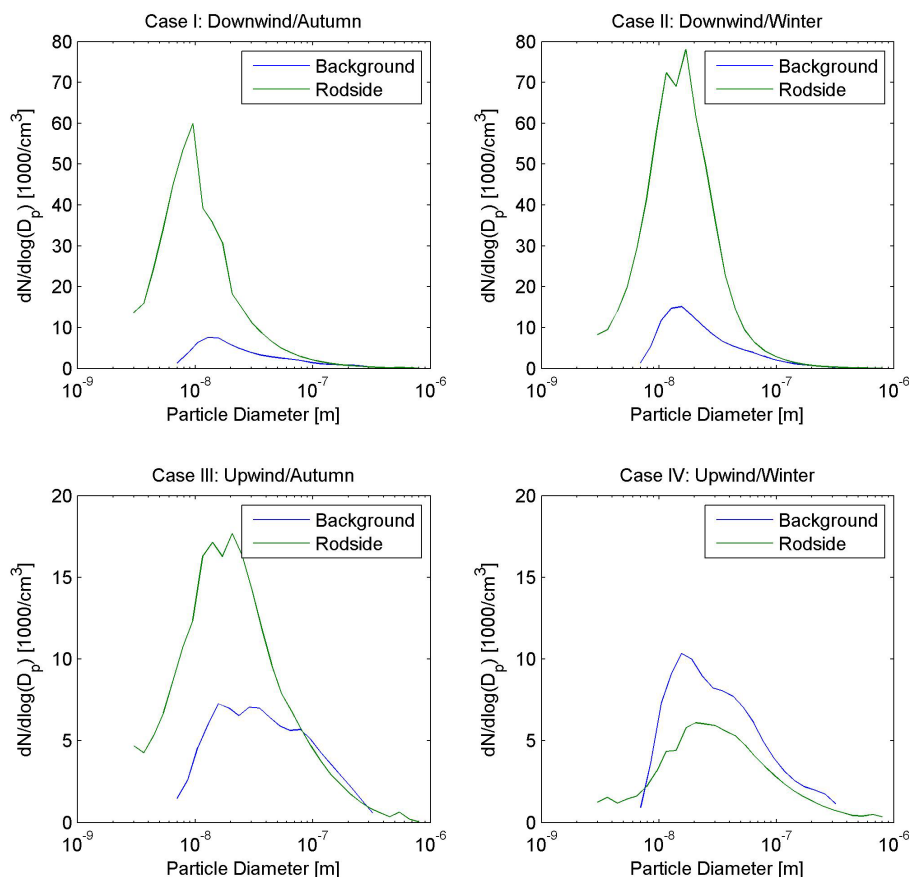
Fig. 5. Median particle number size distributions during downwind conditions (upper panel) and upwind conditions (lower panel). Cases I and III are both in the autumn season and cases II and IV are both in the winter season.

concentrations of fine aerosol particles larger than 100 nm were rather similar at both sites indicating that both sites have similar accumulation particles that are most likely regional. On the other hand, the particle number concentration differences were significant for UFP. The UFP number concentration difference between the roadside site and the background site was as high as 2×10^5 cm^{-3} regardless to the case season as autumn or winter. These high concentrations of UFP were mainly observed during 06:00–18:00 for case I (autumn) and during 06:00–22:00 for case II. It is therefore evident that these particles are originated from traffic combustion emissions nearby a road. It is also clear that the start time when the UFP number concentration increased was around 06:00 regardless to the time of the year; whereas, UFP particles remained airborne for a longer time during the winter period indicating that atmospheric dilution processes are more efficient during the autumn.

Considering the downwind cases only, the maximum value of the particle number size distribution was observed at around 10 nm during the autumn (case I) and at around 20 nm during the winter (case II). This is in agreement with previously reported observations nearby main roads,

street canyons, and car-chasing measurements. For example, Wehner et al. (2002) reported 15 nm in a street canyon and around 20–25 nm for urban background in Leipzig (Germany). In general, the shift in the maximum towards larger diameters is typically accompanied with a significant decrease in the total particle number concentration, as the air mass is transported from the roadside to the background. Pirjola et al. (2006), Gramotnev et al. (2003), and Zhu et al. (2002) showed that the total particle number concentration decreased by 35–45% while transported from the roadside to a distance of about 60–80 m from the road. Both the number concentration decrement and the increment of the location of the maximum of the particle number size distribution have been attributed to dilution process (e.g. Ketzel and Berkowicz, 2004; Shi et al., 1999). In addition to dilution, aerosol dynamics such as condensation and evaporation of vapors are also responsible for the evolution of aerosol particles while transported from a road to the urban background (e.g. Zhang and Wexler, 2004; Zhang et al., 2004; Pohjola et al., 2003). According to Zhang and Wexler (2004), the evolution of the vehicular exhausts near roadways usually experiences two distinct stages: "tailpipe-to-road" and
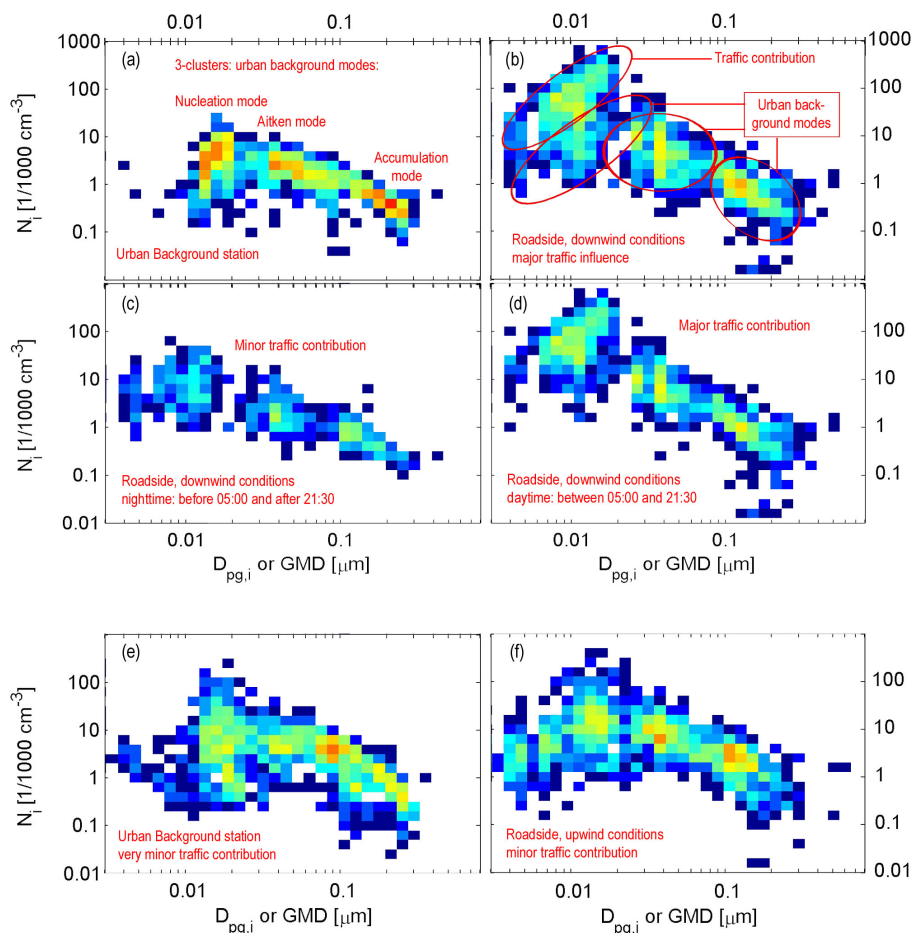
Fig. 6. Frequency of occurrence of the all modes obtained with the multi-lognormal fitting analysis. The frequency of occurrence was evaluated within N_i–$D_{pg,i}$ ranges during case I that represents downwind conditions at the roadside site: **(a)** the urban background site during the same period, **(b)** the roadside site, **(c)** nighttime and early morning at the roadside site, and **(d)** daytime period at the roadside site. Background conditions are presented for case III in **(e)** at the urban background site and **(f)** at the roadside site during upwind conditions. The color scale is arbitrary but it is the same for all subfigures.

"road-to-ambient". The first stage dilution (1000:1 in around 1–3 s) is induced by traffic-generated turbulence, followed by condensational growth of organic compounds, resulting the rapid growth of the nuclei mode. In the second stage dilution (about 10:1 within 3–10 min) is mainly dependent on atmospheric turbulence and the condensational growth is less pronounced. Commonly coagulation is not important for the first and second stage, but could be significant during the second-stage for depletion of nano particles, when dilution is slow.

3.2 Modal structure of the particle number size distributions

We analyzed the modal structure of the particle number size distributions during cases I and III by using the multi-lognormal fitting (Fig. 6). During case I, the particle number size distribution at the urban background site can be fairly fitted by using three lognormal modes (Fig. 6a): an accumulation mode (GMD>100 nm), which represents regional background aerosol particles, an Aitken mode (GMD between 25–100 nm), and a nucleation mode (GMD<25 nm); these modes represent characteristics of urban background aerosol particle (Table 2). On the other hand, the particle number size distribution at the roadside site also showed three modes but with smaller GMD's (Fig. 6b): accumulation (GMD>65 nm), Aitken (20–65 nm), and nucleation (<20 nm). During upwind conditions at the roadside site (case III), similar modal structure was observed at both sites (Figs. 6e–f).

In order to distinguish the traffic influence on the particle number concentrations, we separated the daytime period (05:00–21:00) from the other times that represent nighttime and early morning hours (Figs. 6b–c). The number concentrations of all modes was higher during the daytime period in comparison with all other times (Table 2). The most

Table 2. Modal structure analysis results of the particle number size distribution showing the variation of the individual mode geometric mean diameter and number concentrations during case I (downwind) at the roadside site and the background site.

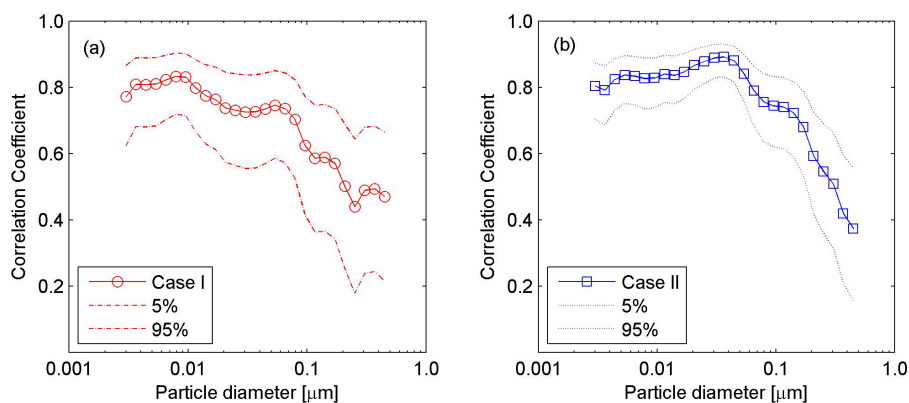| Site	Time	Mode-1 GMD	Concentration	Mode-2 GMD	Concentration	Mode-3 GMD	Concentration
Roadside	All	<20	7000–1 000 000	20–65	6000–50 000	>65	100–4000
	>21:30 and <05:00	<20	10 000–70 000	20–65	6000–10 000	>65	100–3000
	05:00–21:30	<20	7000–1 000 000	20–65	1000–50 000	>65	100–4000
Background	All	<25	500–50 000	25–100	500–90 000	>100	100–2000

Fig. 7. Pearson Correlation coefficients between the particle number size distributions and the traffic density at the roadside site during downwind conditions **(a)** case I and and **(b)** case II.

significant difference was found in the number concentration of the nucleation mode which was about one order of magnitude higher during the daytime than all other times.

We also investigated the statistical correlation between the measured particle number size distribution (at the roadside site) and the traffic density. The computed Pearson correlation coefficients were largest during downwind condition cases (Fig. 7). In general, the correlation coefficients between UFP and the traffic density were higher than 0.55 during case I and higher than 0.75 during case II. The correlation coefficients for accumulation mode particles were low possibly due to the substantial contribution of LRT in that size range. It is very remarkable that the peaks' locations of the correlation coefficient curves as a function of particle diameter (Fig. 7a) closely coincide with the maximum occurrence of the individual modes as a function of the mode GMD and number concentration (Fig. 6b).

3.3 Simulations of the particle number size distributions

Here we considered case II as a modeling exercise to evaluate the modeling scheme presented in Sect. 2.5.2. We selected two periods in case II: 10 February between 12:00–17:00 and 11 February between 12:00–20:00. During these time peri-

ods, the wind speed and direction was fairly stable from the highway towards the measurement site. We used hourly averages of dilution coefficients as obtained from the CAR-FMI and MPP-FMI model simulations. The initial height of the mixing volume was set to 40 cm. The modeling scheme here was then followed according to Part I (Pohjola et al., 2007).

The model simulation exercise by using the emission factor as reported in Part I and the previous study within the LIPIKA campaign on the same site (Pohjola et al., 2007; Pirjola et al., 2004) resulted in significant under prediction of the particle number concentrations at 65 m nearby the roadside (Figs. 8 and 9). The ratio of the simulated to the measured total particle number concentration varied from 0.31 to 0.49 on 10 February and from 0.14 to 0.35 on 11 February. This under prediction can be due several reasons including mainly the validity of the emission factor and the dispersion equation as obtained from the CAR-FMI model. However, due to the substantial uncertainties in the emission factor modeling, we re-simulated the cases by increasing the emission factor with a factor of three, which was the best-fit emission factor.

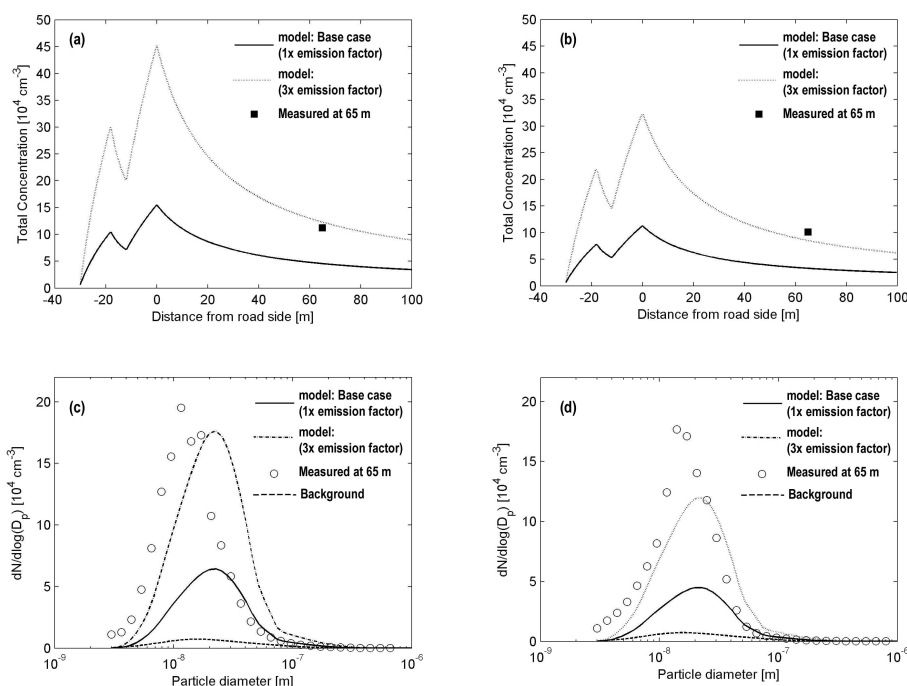Even though the model simulation of the total particle number concentrations improved after using three times the

Fig. 8. Model simulation exercise for the particle number concentration and size distribution (at 65 m) nearby the road on 10 February 2004: **(a)** and **(c)** during 14:00–15:00 and **(b)** and **(d)** during 17:00–18:00. The reference distance (0 m) was assumed at the roadside, road-lane width was assumed 4 m (3 lanes in each direction), and 6 m grass area in between both directions.

emission factor, the particle number size distribution was not well predicted most of the time (Figs. 8c–d and 9c–d). It is therefore clear that the model simulation failed for the selected cases in this study because the emission factors do not seem very well representative for the selected time period. Though the particle number size distributions and the emission factors were previously evaluated for the same site according to Pohjola et al. (2007) for a different year (winter of 2003–2004). Most likely, the evaluation of the emission factors of aerosol particles from traffic combustion are sensitive to the ambient conditions, for example the currently available estimates for the detailed aerosol emission characteristics vary widely (e.g. Ketzel and Berkowicz, 2004; Pohjola et al., 2003).

The above mentioned model simulations represent only a couple of example cases, and the numerical results were compared with data from a single measurement location. These comparisons should therefore be considered as a test of the modeling approach against the available measurement rather than model evaluation.

4 Summary and conclusions

This work was a part of the SAPPHIRE ("Source Apportionment of Airborne Particulate Matter and Polycyclic Aromatic Hydrocarbons in Urban Regions of Europe", 2003–2006) project. Measurement campaigns were conducted in several major European cities, such as Athens, Birmingham, Copenhagen, Helsinki, and Oporto.

In this study we investigated the traffic related size fractionated aerosol particle number concentrations and size distributions measured nearby a major road in Helsinki. We also performed a model exercise to simulate the particle number concentration and size distribution nearby a major road during downwind conditions. The available data and information also included electronic traffic counts, on-site meteorological measurements, and urban background particle number concentration and size distributions.

The contribution of traffic combustion emissions to the particle number size distribution, mainly ultrafine particles (UFP, diameter<100 nm), was substantial during daytime (downwind conditions) and especially high during the traffic rush hours. The UFP number concentrations at the roadside site were approximately an order of magnitude higher than those at the urban background site during these periods. During upwind conditions, the particle number concentrations at the roadside site were comparable to those at the urban background site. On the other hand, the accumulation mode particle number concentrations were only slightly affected by the road contribution.

We also evaluated the statistical correlations between the particle number size distributions and the traffic densities at the roadside site. The computed Pearson correlation coefficients during the downwind cases were (>0.6) for UFP's.
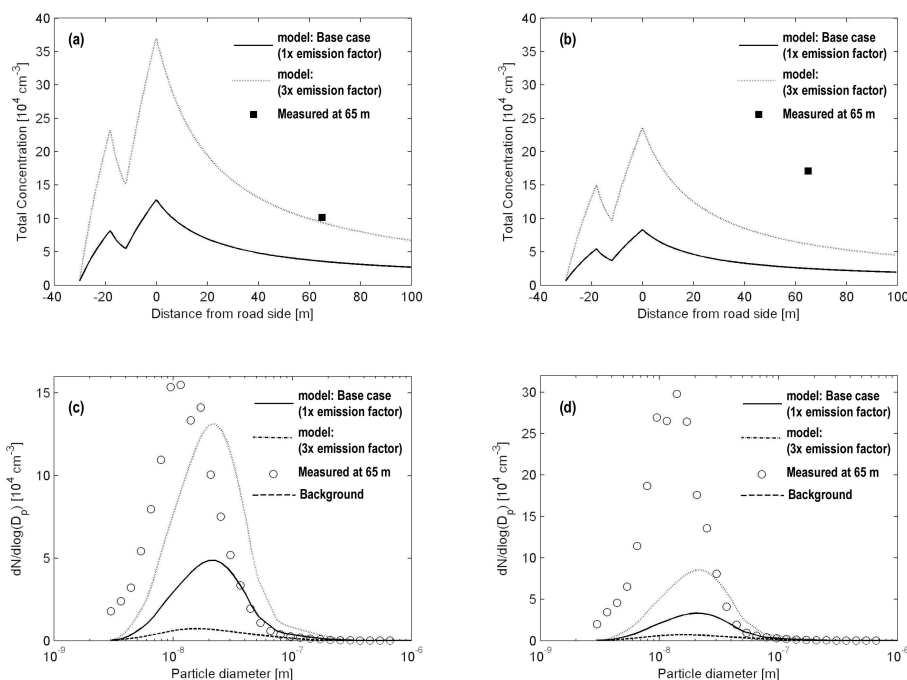
Fig. 9. Model simulation exercise for the particle number concentration and size distribution (at 65 m) nearby the road on 11 February 2004: **(a)** and **(c)** during 14:00–15:00 and **(b)** and **(d)** during 18:00–19:00. The reference distance (0 m) was assumed at the roadside, road-lane width was assumed 4 m (3 lanes in each direction), and 6 m grass area in between both directions.

Along with the modal structure analysis of the particle number size distributions at the roadside site, the UFP are mainly originated from traffic combustion emissions whereas the accumulation mode particles are mainly regional.

The modeling exercise included a dispersion model along with an aerosol dynamic model (UHMA, Korhonen et al., 2004) to simulate the particle number size distribution nearby a road. The main results of this modeling exercise showed that the emission factors of aerosol particles might not be valid for the same site during similar season but different year.

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