**Atmospheric
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Evaluation and modeling of the size fractionated aerosol number concentration measurements near a major road in Helsinki

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Evaluation and modeling of the size fractionated aerosol number

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Abstract

This study presents an evaluation and modeling of the size fractionated aerosol number concentrations that were measured near a major road of Itäväylä in Helsinki, during 23 August – 19 September 2003 and 14 January – 11 February 2004. The measurement system contained also electronic traffic counts, on-site meteorological measurements, and urban background concentration measurements. We have evaluated the temporal variations and the dependencies on local meteorological conditions of the measured aerosol number concentrations and size distributions. The ultrafine particle (UFP) number concentrations at the roadside site were approximately an order of magnitude higher than those at the urban background site during daytime, due to vehicular emissions from the road. We also determined the statistical correlations of the sequential time series of the particle number size distributions at the roadside site, and the traffic densities. The computed Pearson correlation coefficients for the downwind cases were substantially high for UFP's (>0.6), and low for accumulation mode particles; the latter is due to the substantial contribution of long-range transported particles in that size range. We also utilized this dataset for evaluating the performance of a modeling system that consists of a roadside dispersion model CAR-FMI (Contaminants in the Air from a Road – Finnish Meteorological Institute), a meteorological pre-processing model MPP-FMI and an aerosol process model UHMA (University of Helsinki Model for Aerosol processes). Model simulations underpredicted the particle number concentrations at the measurement site, which was caused by uncertainties in the emission modeling, especially in the UFP size range.

1 Introduction

There is a strong indication that the 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.

Aerosol particle concentrations in urban areas are originated from several source categories, such as local vehicular traffic, long-range transport (LRT) and industrial emissions. Also so called regional nucleation (new particle formation) events contribute on particle number concentration (e.g. Kulmala et al., 2004). In Nordic countries, atmospheric LRT constitutes an important part of the total urban background $PM_{2.5}$ concentrations (e.g. Karppinen et al., 2004; Johansson et al. 1999). In Helsinki Metropolitan Area, there are clear seasonal differences in the characteristics of particle number size distributions (e.g. Hussein et al., 2004). For example ultrafine particle (UFP, diameter <100 nm) number concentrations show a clear diurnal variation. 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. However, these dependencies are more moderate for $PM_{2.5}$, caused mainly by the substantial long-range transported background (e.g. Pohjola et al., 2000, 2002 and 2003; Laakso et al., 2003).

Traffic is the most important source of air pollution in urban centers. Vehicular dispersion models are therefore essential computational tools in modern municipal and urban planning. There is, therefore, an urgent need for good-quality experimental data that has been specifically produced for model validation and evaluation purposes, especially regarding particulate matter. For model evaluation, air quality data have to be combined with simultaneous measurements of traffic flow and meteorological parameters, together with information about local topography and buildings. Evaluation of roadside dispersion models is seriously hindered by the scarcity of appropriate experimental datasets (e.g. Kukkonen et al. 2001). The evaluation of road dispersion models against experimental datasets have previously been presented by, e.g., Luhar and Patil (1989); Benson (1992); Kukkonen et al. (2001); Oettl et al. (2001); Tiitta et al. (2002); and Levitin et al. (2005). However, these model evaluation studies do not contain the measurements of fine particulate matter, except for one study (Tiitta et

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al. 2002), and none of these studies addresses size-segregated aerosol data. Wåhlin et al. (2001) and Ketzel et al. (2003) have conducted measurements of fine particles (FP, diameter <0.1 μm) and UFP at various urban and rural locations in Denmark by using a differential mobility analyzer. A significant correlation at street level in two cities was observed between the concentrations of CO, NO$_x$, and UFP which indicates that the local traffic is a major source of UFP in busy streets. They also applied inverse modeling for evaluating the emission factors of UFP originating from various pollution source categories.

The main objectives of this study are to investigate, evaluate, and simulate size-fractionated aerosol number concentrations that were 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 flows were also measured electronically, and meteorological parameters were measured on-site. The results can be utilized especially for investigating particulate matter originating from urban vehicular traffic. We also evaluated a modeling system for dispersion and aerosol processes that contains the roadside dispersion model CAR-FMI (e.g., Kukkonen et al., 2001) and the latest version of the aerosol dynamic model UHMA (Korhonen et al., 2004). Within the SAPPHIRE project, corresponding measurement campaigns have also been conducted in other European cities including Athens, Birmingham, Copenhagen and Oporto.

2 Materials and experimental setup

2.1 Measurement site locations

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

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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.

5   The measurements were performed simultaneously at two locations: an urban background station (Kumpula) and a roadside site (Herttoniemi), as presented in Fig. 1.

2.1.1 Roadside site (Herttoniemi)

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
10 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 is located in fairly flat, homogeneous terrain; there are no obstacles towards the direction of the major road (Itäväylä); however, there were some low buildings and trees in the other directions at a distance of a few tens of meters. There were no residential buildings in the vicinity of the measure-
15 ment site. A minor plastic factory is situated at about a distance of 35 m northeast of the measurement site that may have occasionally provided emissions of aerosol particles. In the subsequent analysis of the measured data in terms of the wind direction, we have therefore excluded from the analysis the sector that is in the direction of this installation.

20 ### 2.1.2 Urban background station (Kumpula)

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. The department is located on the Kumpula campus area of the University of Helsinki (on a hilltop of approximately 20 m height from the nearby). The campus is located
25 5 km northeast the centre of Helsinki (Fig. 1). The measurement site is surrounded with residential and university buildings in the northeastern side. To the west there

are greenswards, mainly forested area and some scattered houses. At a distance of less than 200 m to the east, there is a densely trafficked major highway (Lahdentie). However, 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.

2.2 Aerosol particle measurements

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) (e.g. Aalto et al., 2001), whereas at the roadside site they were measured with a twin scanning mobility particle sizer (SMPS). The differential mobility analysis of aerosol particles relies on bipolar charging of the particles (Liu and Pui, 1974) followed by classification of particles due to electrical mobility by a differential mobility analyzer (DMA) (Knutson and Whitby, 1975) and then counting the particles 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).

2.2.1 DMPS setup at the background station

The sample flow rate in the DMPS was 1.5 liters per minute and the sheath flow was 8.5 liters per minute. The sheath flow was 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 DMA voltage change was 15 s and after that particles were counted over a three second period. One measurement cycle took five minutes.

The particle number size distributions were extracted by inversion. The inversion was

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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).

2.2.2   SMPS setup at the roadside sitev

The twin SMPS system consists of two differential mobility analyzers. In the twin SMPS system, the first unit measured particle number size distributions between 3–50 nm and it consists 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 liter per minute and the sheath flow was 17 liter per minute. In the second unit that measures particle number size distributions between 10–800 nm we used a DMA (HAUKE –type 28 cm in length) with a CPC TSI 3010. The sample flow in this unit was 1.0 liter per minute and the sheath flow was 5 liter per minute. 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 liter per minute. There was no special inlet in the main sample line. One measurement cycle took one minute.

   We implemented laboratory calibrations for the detection efficiency of the CPC instruments. 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 included CPC and DMA calibration and thorough cleaning. The CPC calibration included the particle detection efficiency calibration and the concentration calibration against the aerosol electrometer. DMA calibration included transport loss calibration and the sizing accuracy calibration with PSL particles. The CPC was serviced and calibrated at the factory when needed.

2.3 Meteorological measurements

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 a height of 5 m from the ground; wind speed and direction along with temperature were measured at a height of 8 m from the ground. We used this on-site meteorological data for the wind sector analysis to define the downwind and upwind cases.

Atmospheric stability parameters and the mixing height were evaluated using the meteorological pre-processing model developed at the Finnish Meteorological Institute, MPP-FMI (Karppinen et al., 2000b), utilizing the meteorological database of the FMI, which contains routine weather and sounding observations. We used a combination of the data from the stations at Helsinki-Vantaa airport (north of Helsinki) and Isosaari (an island south of the centre of Helsinki). The mixing height of the atmospheric boundary layer was evaluated using the meteorological pre-processor, based on synoptic observations and the sounding observations made at Jokioinen (in a rural area in Southern Finland).

2.4 Traffic density data

The traffic density data was available from the Helsinki City Planning Department. The traffic densities on the nearest major roads (Itäväylä and Lahdentie) at both sites were similar regarding the average traffic flows and their temporal variation; the relative differences in the daily average traffic densities at these two roads were less than 5 %. The daily average traffic density on Itäväylä varied between 32 000–54 000 vehicles per day during the investigated periods (Fig. 2). Based on the traffic data analysis for the year 2001, about 85% of the vehicles were light duty personal vehicles (of which 11% were diesel), about 12% were vans (of which about 84% diesel), and about 4% heavy duty vehicles. The traffic density was highest between 07:00–10:00 and 15:00–18:00

on workdays.

3 Modeling

3.1 Modal structure analysis: multi-lognormal fitting

It is often convenient and mathematically straightforward to use a modal representation of the size distribution (e.g. Whitby, 1978). The distribution can be described as the sum of several number of lognormal modes, each is characterized with D_{pg} (geometric mean diameter), σ_g (geometric standard deviation), and N (number concentration);

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

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. 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 estimate the multi-lognormal parameters for the experimentally measured size distributions. The modified fitting program first analyzes the measured size distribution in order to decide how many modes should be fitted to the data. It then makes a preliminary guess of the modal parameters, and uses the least-square minimization method to fit from one up to five modes to the measured size distribution.

The recent modifications of the DO-FIT algorithm were to optimize and enhance its performance while fitting the modes and to introduce a new set of conditions on the decision of the suitable number of modes. The original algorithm could accurately fit about 80% of the data set discussed by Hussein et al. (2004), whereas the modified

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algorithm can fit more than 90% of the same data set, and the accuracy of the fitting has improved.

3.2 Modeling of the particle number size distributions

We utilized a modified version of the aerosol dynamic model UHMA (Korhonen et al. 2004) to simulate the particle number size distributions. The model simulates the time evolution of particle size and composition distribution; it includes mathematical treatments for Brownian coagulation, and the condensation of sulphuric acid, organic vapors, and water.

No emission measurements or modelling were performed within the SAPPHIRE campaign in Helsinki. However, the emissions of particle number concentration from vehicular traffic were evaluated within the so-called LIPIKA measurement campaign that was conducted previously on the same site (e.g. Pirjola et al., 2004). These evaluations were performed using measured concentrations and inverse dispersion modelling. The model simulations were limited to the cases in winter, due to the availability of the LIPIKA emission data. Although the LIPIKA campaign was not conducted during the same period as the cases considered in this study, it is expected to represent the emission rates for winter conditions. The measured emission rates were scaled according to the measured traffic densities on the simulated days, i.e., multiplied by the ratio of the measured traffic density in the SAPPHIRE campaign and the traffic density of the LIPIKA campaign, separately for each hour. The nocturnal time period between 01:00 and 03:30 was assumed to be representative for the regional background particle number size distribution regarding UFP, as the local traffic density was very low during that time (Hussein et al., 2005b).

In the modified aerosol process model UHMA requires as input parameters for the rate of pollutants dilution, traffic densities, and the initial mixing volume height; we set these parameters according to (Pirjola et al., 2006). The combined dispersion and aerosol process model addresses expanding air parcels that are transported along with the air flow over the road. First, one needs to assume an initial height of the air parcel

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volume, which corresponds to the situation where vehicular exhaust gases and particles have been diluted in a time scale of approximately 0.5 s, after their discharge from the tailpipe. The initial dispersion is mainly dependent on traffic-induced turbulence (Pirjola et al., 2006).

5　　The dilution of traffic exhausts were 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).

4　Results and discussion

The aerosol data was first checked for consistency and correctness. The SMPS instruments typically measure with a higher time resolution, compared with the DMPS instruments. In order to compare the aerosol data measured at the two sites, we therefore calculated the 5-min averages of the SMPS data measured at the roadside site. As the measured size ranges were different at the two sites, we performed the comparison for the particle size range between 8–320 nm.

20　　We selected five time periods, during which the wind direction did not change substantially, for a more detailed analysis. The variation of some relevant meteorological parameters measured on-site during four of these cases are summarized in Table 1. These cases can be classified based on the prevailing wind direction to represent downwind (cases I and II) and upwind (cases III and IV) conditions at the roadside site. These cases include winter (cases II and IV), summer (case I) and autumn (case III) periods. The variation of the relevant meteorological parameters and the measured particle number size distributions at both sites are presented in Figs. 3–6. The parti-

cle number concentrations show a clear diurnal variation at both sites; this variation is closely correlated to the corresponding variation of the traffic density.

As expected, particle number concentrations at the roadside site (panels d in Figs. 3–6) are clearly higher for the downwind cases (Cases I and II), compared with the upwind cases (Cases III and IV), mainly caused by the vehicular emissions originated from the highway. The road contribution to the particle number size distribution (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 (panels e in Figs. 3–6). For example, the UFP number concentrations at the roadside site were approximately an order of magnitude higher than those at the urban background site during daytime. However, in downwind conditions at night, the total particle number concentrations were only slightly higher at the roadside site in comparison to the background site.

These findings are illustrated in another manner by the particle number size distributions averaged over the time period for each case (Cases I–IV), which are presented in Figs. 7a–d. The roadside concentrations were substantially higher than those at the urban background site for the downwind cases, and these were comparable for the upwind cases. However, the accumulation mode particle number concentrations are only slightly affected by the road contribution.

The maximum value in the particle number size distribution was found to be between 10–30 nm. This is in agreement with previously reported observations nearby main roads, street canyons, and car-chasing measurements. For example, Wehner et al. (2002) reported a maximum in the particle number size distributions at 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 urban 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

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of about 60–80 m from the road.

Both the decrease of the number concentration and increase of the location of the maximum in the 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 the vicinity of 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 '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 for depletion of nano particles, when second-stage dilution is slow.

We have presented an analysis of the modal structure of aerosol particles measured at the roadside and background sites in Fig. 8. The number of modes of urban aerosols is dependent on the local sources (e.g. Hussein et al., 2004 and 2005b). The modal structure of urban background aerosol particles can be fairly well characterized by 2–3 lognormal modes (Fig. 8a and e): an accumulation mode (GMD>100 nm) that represents regional background characteristics, in addition to two UFP modes (GMD<100 nm) that represent urban characteristics. An additional mode (GMD<25 nm) with concentrations over 50 000 cm^{-3} is required to describe the size distributions at the roadside site during downwind conditions (Fig. 8b). A similar modal structure for both sites was obtained during upwind conditions (Fig. 8e–f). In order to separate the fresh nucleation mode (traffic contribution) and the urban background modes, we have presented the frequency plots for nighttime and daytime measurements at the roadside site during 25 and 26 August 2003 (Case I) in Fig. 8c–d. This illustrates the major traffic contribution in daytime to the particle number size distri-

bution at the roadside site for downwind conditions, in contrast to the corresponding results measured at night.

We have also investigated the statistical correlation between the particle number size distributions at the roadside site and the traffic densities. The computed Pearson correlation coefficients were largest for the downwind cases; these have been presented in Fig. 9. In general, the correlation coefficients were higher than 0.6 for UFP for Case I and they were higher than 0.7 for Case II. The correlation coefficients for accumulation mode particles are low; this is caused by the substantial contribution of long-range transported particles in that size range.

5   The effect of wind speed on the particle number concentrations

On 23 January 2004 the weather conditions including temperature, wind direction, and relative humidity remained rather similar in the afternoon (Fig. 10); it was upwind conditions at the roadside site. The wind speed was nearly zero before noon and it increased in a step-wise manner during the afternoon (Table 2). These situations provide a sight on how fast urban aerosols are diluted and dispersed in the urban atmosphere.

6   Modeling of the particle number size distribution

The evolution of particle number size distribution as a function of distance from the road was modeled using the coupled modelling system that contains a sectional aerosol dynamic model, a roadside dispersion model and a meteorological pre-processing model, for Case II. We selected two periods for simulations: 10 February between 12:00–17:00 and 11 February between 12:00–20:00. During both of these periods, the direction of the air flow was fairly stable, from the highway towards the measurement site. We used hourly averages of dilution coefficients, which were obtained from the CAR-FMI and MPP-FMI model results, and the measured traffic densities for the model simula-

tions. The initial mixing volume height was set to 40 cm, and the modelling of emissions was performed according to Pohjola et al. (2007).

Model simulations for the selected periods on 10 and 11 February substantially under predict the particle number concentrations at the measurement site (Fig. 11a–b and

5 e–f); the ratio of simulated to measured total concentration varies from 0.31 to 0.49 on 10 February and from 0.14 to 0.35 on 11 February. Due to the substantial uncertainties in the emission modeling, these curves have also been presented using a three-fold emission factor. There was also a systematic under prediction of the measured particle number size distributions, and the location of distributions was systematically over

10 predicted in terms of the particle size (Figs. 11c–d, and g–h). The under prediction of number concentrations is, therefore, predominantly due to the model failure to correctly simulate the high concentration of traffic-emitted particles in the UFP size range. The background particle number concentrations are very low in comparison to the traffic-originated particle concentrations and therefore the background particles do not

15 substantially affect the simulated particle size distributions.

The size distribution and the amount of traffic emissions were modeled here according to Pohjola et al. (2007) for the same highway and the same season, but for a different year (winter of 2003–2004). The most probable reason for the differences of the modeled and measured size distributions is that the characteristics of the traf-

20 fic emissions in terms of the size distribution and number concentrations are sensitive to the ambient conditions. The currently available estimates for the detailed aerosol emission characteristics vary widely (e.g. Ketzel and Berkowicz, 2004; Pohjola et al., 2003).

7 Conclusions

25 This work is 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. This study investigates the size fractionated aerosol number concentrations that were measured near a major road in Helsinki. The measurement system contained also electronic traffic counts, on-site meteorological measurements, and urban background concentration
5 measurements. A limitation of the present campaign was that vehicular emissions were not measured on-site, as was the case in a previous measurement campaign that was conducted on the same location (Pohjola et al., 2007).

We evaluated the temporal variations and the dependencies on local meteorological conditions of the measured aerosol number concentrations and size distributions at the
10 roadside and the urban background locations. We selected five time periods, during which the wind direction did not change substantially, for a more detailed analysis. Two of these cases represent downwind, two upwind conditions at the roadside site in winter, summer and autumn. As expected, the total particle number concentrations show a clear diurnal variation at both sites; this variation is closely correlated to the
15 corresponding variation of the traffic density.

The road contribution to the particle number size distribution was substantial in daytime 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 in daytime. For the upwind cases, the roadside concentrations
20 were comparable to those at the urban background site. However, the accumulation mode particle number concentrations were only slightly affected by the road contribution, due to the substantial long-range transported fraction for that particle mode. We also determined the statistical correlations of the sequential time series of the particle number size distributions at the roadside site, and the traffic densities. The computed
25 Pearson correlation coefficients for the downwind cases were substantially high for UFP's (>0.6), and low for accumulation mode particles.

The maximum values of the particle number size distributions varied from 10 to 30 nm. This is in agreement with previously reported observations nearby main roads, street canyons, and car-chasing measurements (e.g., Wehner et al., 2002). In addi-

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tion to dilution, aerosol dynamics, such as condensation and evaporation of vapors, are also responsible for the evolution of aerosol particles while transported from the vicinity of a road to the urban background (e.g. Zhang and Wexler, 2004).

We also utilized this dataset for evaluating the performance of a coupled modeling system that consists of the models CAR-FMI, MPP-FMI and UHMA. Model simulations under predicted the particle number concentrations at the measurement site, which was caused by uncertainties in the emission modeling, especially in the UFP size range.

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Table 1. Selected cases that were considered in the analysis and the variations of some meteorological parameters during these cases. WD = wind direction, WS = wind speed, T = ambient temperature, and RH = relative humidity.

	Time period	WD	WS [m/s]	T [°C]	RH%	Location of the station
Case I	August 23–28, 2003	−90 – 30	<4	10–15	50–95	Downwind
Case II	February 9–11, 2004	−90 – 0	<5	−15 – −4	55–92	Downwind
Case III	14–16 September 2003	170–235	<4	10–18	80–95	Upwind
Case IV	24–30 January 2004	45–270	<5	−7.5 – −12.5	75–95	Upwind

Table 2. Average values for the weather conditions during Case V.

	Time period	WD	WS [m/s]	T [°C]	RH%
P1	10:00–12:30	183	0.05	−11	90
P2	12:30–16:30	195	2.15	−3	87
P3	16:30–23:59	189	3.43	−2	87
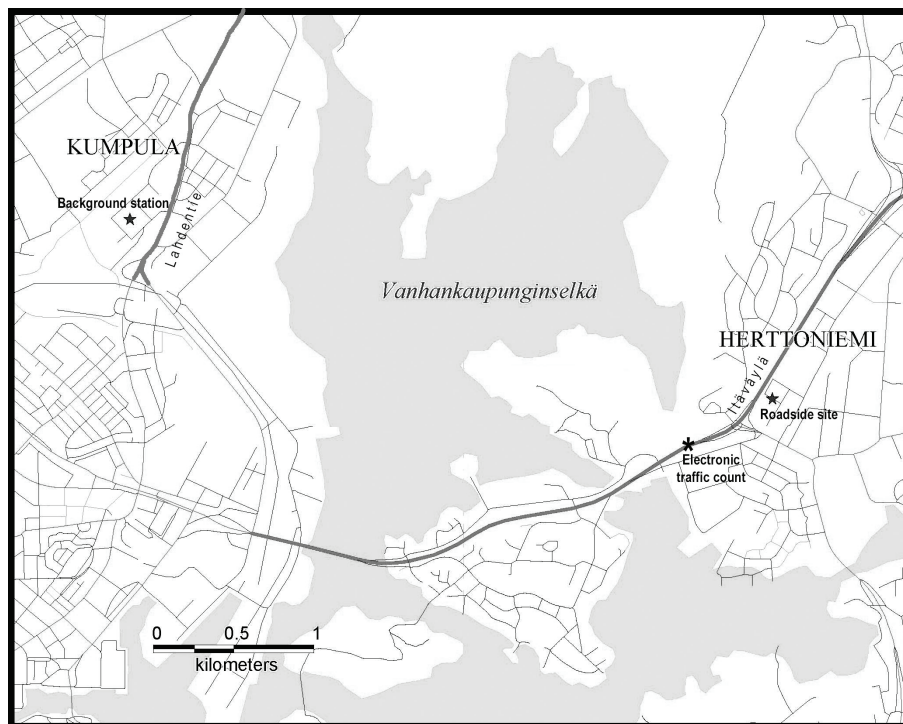
Fig. 1. Measurement sites and traffic count locations (marked with stars). The map also shows the road and street network; the nearby major roads are indicated with dark lines. Grey area is a part of the Baltic Sea. The north is directed upward in this map. This map was provided by Ground map material (C) National Land Survey of Finland, licence number 158/MYY/06.
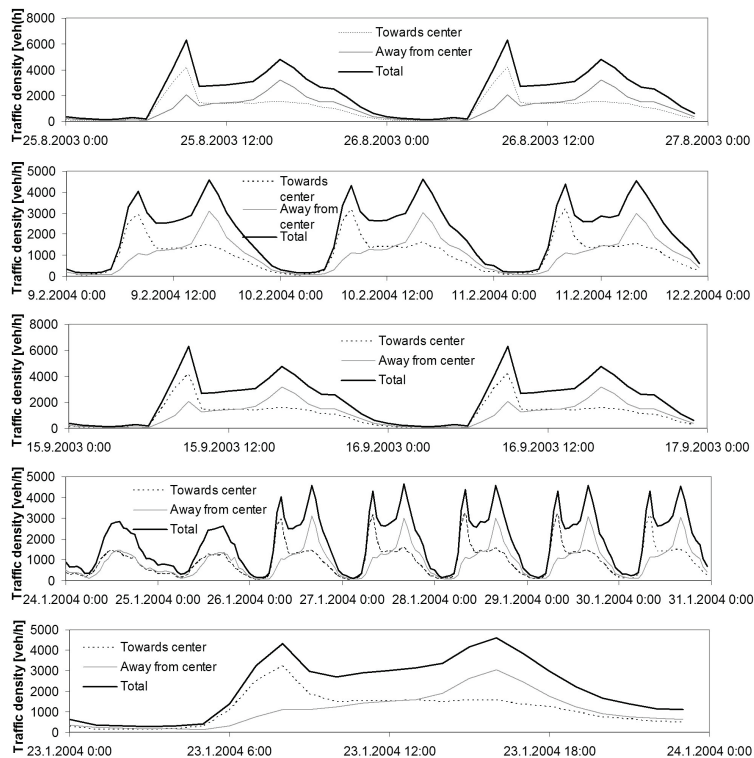
Fig. 2. Traffic densities on the major road in the vicinity of the roadside site at Herttoniemi. The time periods correspond to the selected cases I–V.
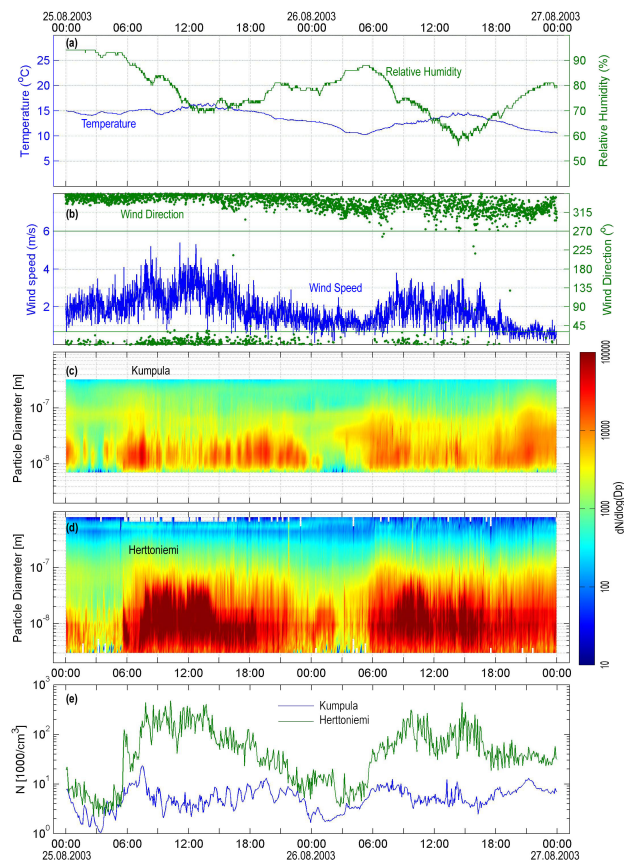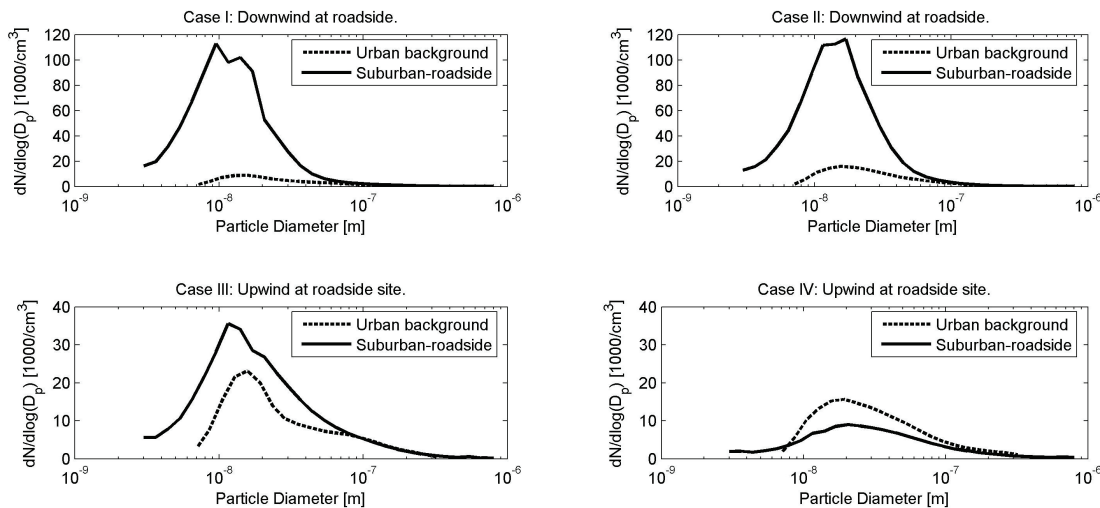
Fig. 3. Case I: **(a)**–**(b)** relevant meteorological variables, **(c)**–**(d)** the particle number distribution spectra at the roadside site (Herttoniemi) and the urban background site (Kumpula), and **(e)** particle number concentrations between 8–320 nm at both sites. This case represents the direct influence of traffic emissions on the particle number size distributions at the roadside site in autumn.

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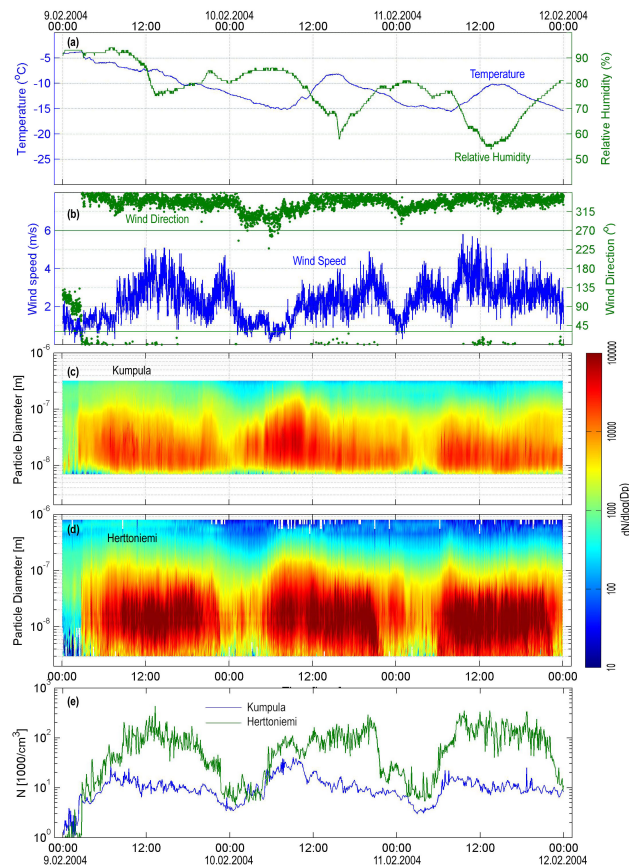
Fig. 4. Case II: **(a)**–**(b)** relevant meteorological variables, **(c)**–**(d)** the particle number distribution spectra at the roadside site (Herttoniemi) and the urban background site (Kumpula), and **(e)** particle number concentrations between 8–320 nm at both sites. This case represents the direct influence of traffic emissions on the particle number size distributions at the roadside site in winter.

Fig. 5. Case III: **(a)**–**(b)** relevant meteorological variables, **(c)**–**(d)** the particle number distribution spectra at the roadside site (Herttoniemi) and the urban background site (Kumpula), and **(e)** particle number concentrations between 8–320 nm at both sites. This case represents urban background aerosols at the roadside site in winter.

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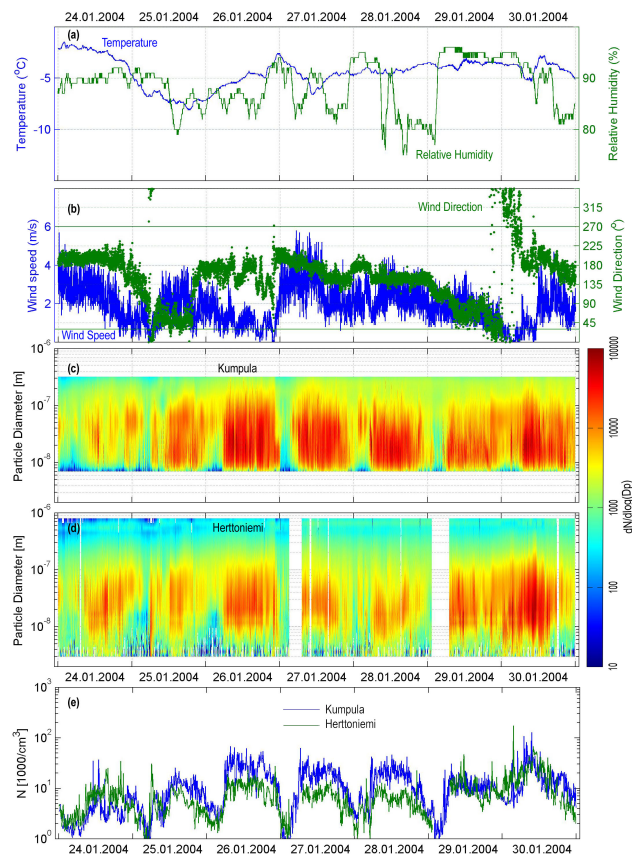
Fig. 6. Case IV: **(a)**–**(b)** relevant meteorological variables, **(c)**–**(d)** the particle number distribution spectra at the roadside site (Herttoniemi) and the urban background site (Kumpula), and **(e)** particle number concentrations between 8–320 nm at both sites. This case represents urban background aerosols at the roadside site in autumn.

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Fig. 7. Mean particle number size distributions during Cases I–IV. Cases I and III are both during autumn and they are respectively downwind and upwind conditions at the roadside site. Cases II and IV are both during winter and they are respectively downwind and upwind conditions at the roadside site.
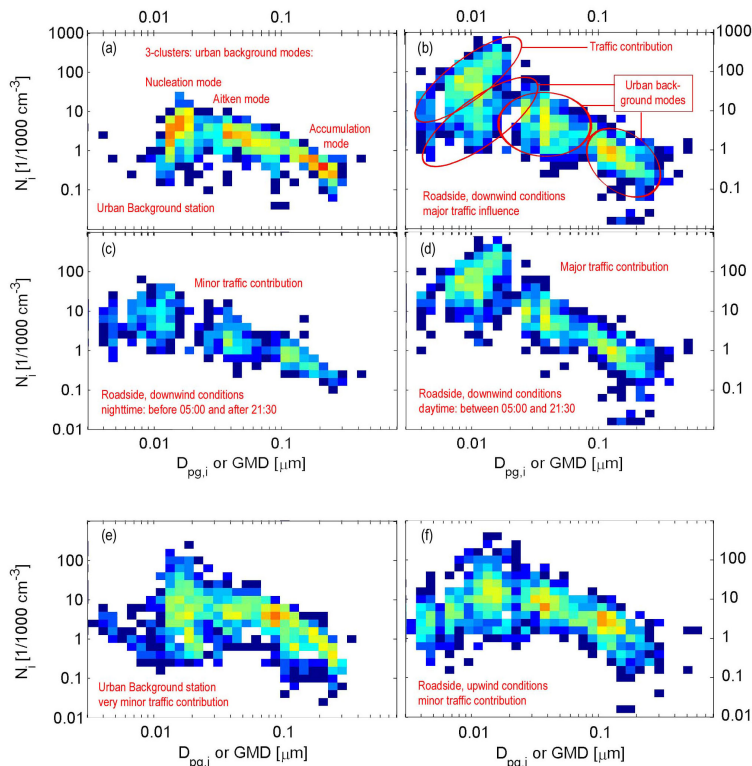
Fig. 8. Frequency of occurrence within a certain mode concentration-GMD range. The frequency plots are generated for a total of two working days as described in **(a)**–**(d)** Case I for downwind conditions at the roadside site (15 and 26 August 2003) and **(e)**–**(f)** Case III for upwind conditions at the roadside site (15 and 16 September 2003). Note that that the frequency scale is arbitrary and it is the same for all plots.

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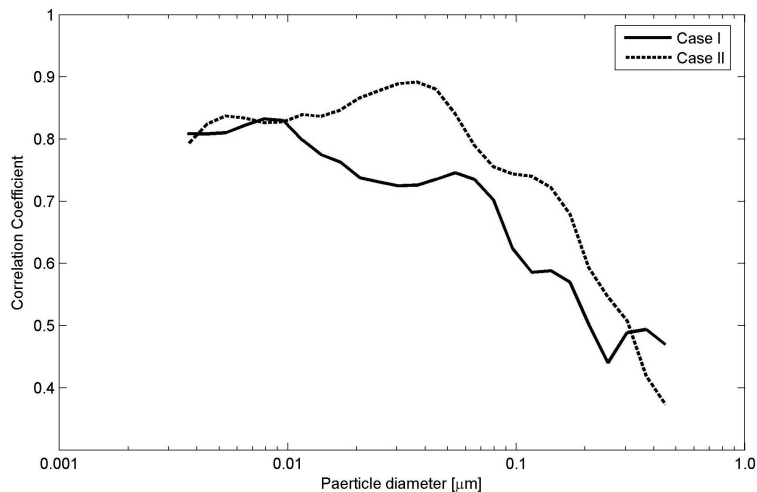
Fig. 9. Pearson Correlation coefficients between the particle number size distributions and the traffic density at the roadside site during downwind conditions (Cases I and II).
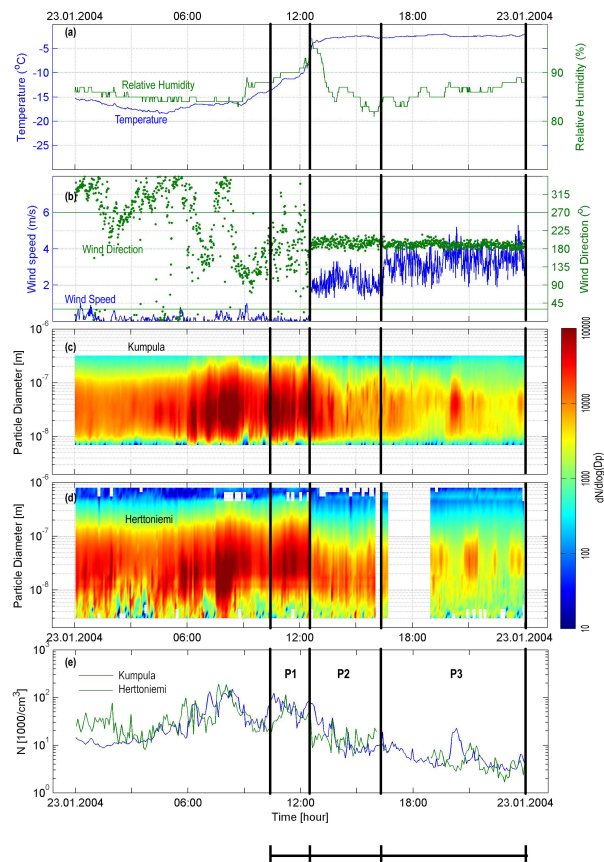
Fig. 10. Case V: **(a)**–**(b)** relevant meteorological variables, **(c)**–**(d)** the particle number distribution spectra at the roadside site (Herttoniemi) and the urban background site (Kumpula), and **(e)** particle number concentrations between 8–320 nm at both sites. This case illustrates the dependency of particle number size distributions on the wind speed when temperature, relative humidity, and wind direction are rather unchanged.
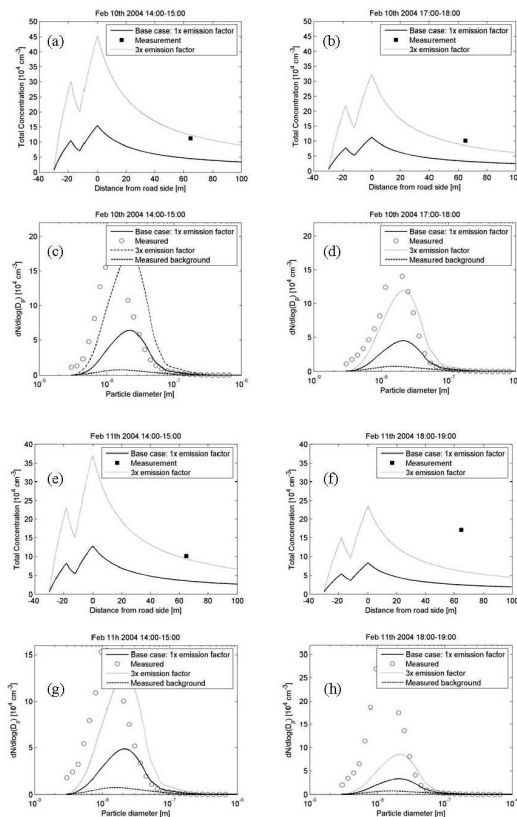
Fig. 11. Model evaluation exercise aiming to predict the particle number size distributions at 65 m from the roadside **(c)**–**(d)** and **(g)**–**(h)**. **(a)**–**(b)** and **(e)**–**(f)** are the corresponding total particle number concentrations; the zero distance was assumed at the roadside. The width of each lane was assumed 4 m (3 lanes in each direction) and 6 m grass area in between both directions.