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**Evaluation of a
coupled dispersion
and aerosol process
model**

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Evaluation of a coupled dispersion and aerosol process model against measurements near a major road

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A field measurement campaign was conducted near a major road 'Itäväylä' in an urban area in Helsinki in 17–20 February 2003. Aerosol measurements were conducted using a mobile laboratory "Sniffer" at various distances from the road, and at an urban background location. Measurements included particle size distribution in the size range of 7 nm–10 μ m (aerodynamic diameter) by the Electrical Low Pressure Impactor (ELPI) and in the size range of 3–50 nm (mobility diameter) by Scanning Mobility Particle Sizer (SMPS), total number concentration of particles larger than 3 nm detected by an ultrafine condensation particle counter (UCPC), temperature, relative humidity, wind speed and direction, driving route of the mobile laboratory, and traffic density on the studied road. In this study, we have compared measured concentration data with the predictions of the road network dispersion model CAR-FMI used in combination with an aerosol process model MONO32. The vehicular exhaust emissions, and atmospheric dispersion and transformation of fine and ultrafine particles was evaluated within the distance scale of 200 m (corresponding to a time scale of a couple of minutes). We computed the temporal evolution of the number concentrations, size distributions and chemical compositions of various particle size classes. The atmospheric dilution rate of particles is obtained from the roadside dispersion model CAR-FMI. Considering the evolution of total number concentration, dilution was shown to be the most important process. The influence of coagulation and condensation on the number concentrations of particle size modes was found to be negligible at this distance scale. Condensation was found to affect the evolution of particle diameter in the two smallest particle modes. The assumed value of the concentration of condensable organic vapour of 10^{12} molecules cm^{-3} was shown to be in a disagreement with the measured particle size evolution, while the modelling runs with the concentration of condensable organic vapour of 10^9 – 10^{10} molecules cm^{-3} resulted in particle sizes that were closest to the measured values.

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1 Introduction

Elevated particle concentrations have been associated with short and long term health effects (e.g. WHO, 2004) There is also evidence that the ultrafine particle fraction is associated with deterioration of respiratory health (Peters et al., 1997). Therefore high number concentrations of fine and ultrafine particles in urban environments, especially in the vicinity of major streets and roads, have raised the interest to study the physical and chemical transformation of PM in the urban environment. This work is a continuation of our effort to analyse particle transformation and dilution in a local scale. In our earlier work, we have studied numerically the transformation and dilution in distance scale of less than 100 m from a road with a combination of the aerosol process model MONO32 and a simple plume model (Pohjola et al., 2003 and 2004). Dilution was found to be the most important process affecting the particle number concentrations, and effect of condensation was seen to depend on the available concentration of condensable organic vapour. However, only a small set of experimental data was available for the evaluation of the numerical computations.

Of the aerosol processes, nucleation was not considered in this work. According to the theoretical work of Zhang and Wexler (2004) the first stage of dilution during 1–3 s would be accompanied with nucleation. At least for diesel exhaust nucleation is likely (Maricq et al., 2002; Zhang and Wexler, 2004). The modelling of nucleation would require more detailed information about the environmental conditions very near the tailpipe (e.g. temperature gradient, information about chemical composition and concentrations of volatile nucleating vapours), which was not available. We also did not assess the influence of dry deposition, as it was previously shown to be irrelevant in this time scale (Ketznel and Berkowicz, 2004).

Dispersion of particle number concentrations and size distributions at various distances from roads have been measured by several groups, e.g. in the United States (Zhu et al., 2002a, 2002b, 2004), in Australia (Morawska et al., 1999; Hitchins et al., 2000; Gramotnev and Ristovski, 2004), in United Kingdom (Shi et al., 1999, 2001)

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and in Finland (Pirjola et al., 2006). In most of those studies the measurements were taken at 15 m and further up to 300 m downwind from the highway. However, in the last mentioned case, during the Finnish LIPIKA campaign, a mobile laboratory was available, and the dispersion measurements were conducted directly on a highway and downwind to around a distance of 150 m. These measurements therefore provide a comprehensive data set for model evaluation.

Recently, the evolution of particle number concentrations and size distributions as function of distance to a road or street, and the importance of aerosol processes has been analysed via the concept of aerosol process time scales by Zhang and Wexler (2004) and Ketzel and Berkowicz (2004), and with aerosol dynamics model simulations by Jacobson and Seinfeld (2004).

The aims of this study were 1) to model the evolution of particle number concentrations and size distributions as functions of distance to a road, 2) to study the importance of the concentration of volatile condensable vapour for this evolution, and 3) to determine, which of the included aerosol processes (dilution and mixing with ambient air, coagulation and condensation) are important in the studied distance scale (the modelled distances extend up to 350 m). Compared to the previous work of the present authors in this area (Pohjola et al., 2003 and 2004), the modelling of atmospheric dilution has been substantially improved, and the modelling also contains proper atmospheric boundary layer scaling (using the roadside dispersion model CAR-FMI combined with the meteorological pre-processing model MPP-FMI). The experimental dataset that is utilised in the evaluation of the predicted results is also substantially more extensive both in terms of the number of data and the measured parameters.

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2 Experimental

2.1 Measuring locations and equipment

In this work, we consider the aerosol measurements conducted by a mobile laboratory, Sniffer, at various locations near the highway Itäväylä, a major road in an urban area of Helsinki, during the LIPIKA campaign in 17–20 February 2003, weekdays from Monday to Thursday (Pirjola et al., 2006). A schematic description of the measurement site is presented in Fig. 1. The highway consists of six lanes, three lanes to both directions (the total width of three lanes is 12 m), and a 6 m wide central grass area between the lanes to both directions. The speed limit is 80 km h^{-1} and the average vehicle speed in the vicinity of the measurement site was approximately 70 km h^{-1} .

The measurement site is located about 5 km northeast of the city centre in a suburban area with substantial small-scale industrial capacity. The closest factory building with the height of approximately 7 m (2 floors) is located in the next block north or northeast of the measurement sites. However, to south and southwest of the street, in which the measurements were conducted (Fig. 1) there are no buildings, only some individual deciduous trees of an average height of 4 m. The factory periodically emits particles which contribute to the local background aerosol. However, the data presented in this paper include only north-westerly wind sectors, which allowed for the measurements of the traffic exhausts originated from the road of Itäväylä.

The urban background was measured at the site of Saunalahti bay that is located approximately at a distance of 600 m northwest of the road. There are no major pollution sources in the immediate vicinity of this station. The urban background measurements were conducted before and after the roadside measurements a few times each day: in the morning before and after the morning rush hours, in the afternoon, and in late afternoon after the rush hours, altogether 189 min. The time resolution of the urban background measurements was 10 min or better.

The mobile laboratory used was constructed into a Volkswagen LT35 diesel vehicle (Pirjola et al., 2004). Particle size distribution in the size range of 7 nm – $10 \mu\text{m}$ (aerody-

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5 namic diameter) with 12 channels is measured by the Electrical Low Pressure Impactor (ELPI, Dekati Ltd, Keskinen et al., 1992) with a time resolution of 1 s. The nucleation mode particle size distribution was measured with a high size resolution by a Hauke type Scanning Mobility Particle Sizer (SMPS), where particles are first neutralised, then classified by a Differential Mobility Analyser (DMA) based on their electrical mobility, and counted by a CPC 3025 (TSI, Inc.). Measurement size range was 3–50 nm (mobility diameter), with a resolution of 20 channels and a scan-up time mostly 30 s. Additionally, the total number concentration of particles larger than 3 nm is detected by an ultrafine condensation particle counter CPC 3025 (TSI, Inc.). A passive clean air dilution system (ratio 1:3) was installed, in order to keep inside the measuring range of the instrument. The particle measurement inlet is located at the height of 2.4 m.

10 The weather station is located at the roof of the van. Temperature and relative humidity (Model HMP45A, Vaisala) as well as relative (in case of a moving vehicle) wind speed and direction by an ultrasonic wind sensor (Model WAS425AH, Vaisala) were measured at the height of 2.9 m. Additionally, a global position system (GPS V, Garmin) detected the van speed and the driving route, and a video camera in the cab recorded visually the traffic situations.

15 Additional meteorological data were measured at a height of 10 m from the ground at the roof of a container that was located at a distance of 9 m from the edge of the road. Wind speed and direction data obtained from the container measurements were compared with the measurements by the mobile laboratory. Due to the disturbance of the flow caused by the measuring van itself and the surrounding buildings, especially at low altitudes, we decided to use the wind data measured at the roof of the container.

2.2 Traffic measurements

25 The traffic flow data at Itäväylä was recorded continuously using electronic loops installed below the street surface by the City of Helsinki and the Finnish Road Administration at a site located about 1.8 km southwest of the Itäväylä measurement site (Pirjola et al., 2006, Virtanen et al., 2006). However, there is one major junction between

the traffic density count site and the measurement site. Additional manual three-minute vehicle counts (including the total traffic flow, and the fractions of light- and heavy-duty vehicles) were therefore performed at selected intervals at the mobile laboratory in order to check the representativity of the continuous traffic flow measurements. The agreement of the visual and the continuous electronic counts was excellent, indicating that the continuous measurements are well representative (Pirjola et al., 2006).

The measured traffic densities are presented in Fig. 2. The average daily traffic count during the time period considered was 46063 vehicles per day, 50% of which was directed towards the city center on average. The average daily total traffic count varied between a maximum of 46477 veh/day on 20 February and a minimum of 45581 veh/day on 18 February, and the variations in the diurnal hourly traffic patterns were small within the four days studied. On all the days, the busiest traffic occurred during the morning and evening rush hours. The traffic during the morning hour contained on average about 8% of the daily traffic count (and 71% of the traffic during this hour was directed towards the city centre). The traffic at 04:00–05:00 p.m. contained on average about 9% of the total daily traffic count (and 64% of the traffic within this one hour was directed away from the centre).

3 Modelling methods

3.1 Modelling of the vehicular emissions

Emission factors for particles were calculated for the whole vehicle fleet characteristic of Itäväylä. In the Helsinki Metropolitan Area (HMA), light duty (LD) vehicles constitute about 90% of all traffic, 80% of which are petrol vehicles and 20% diesel vehicles (Kauhaniemi, 2003). It was assumed that all heavy duty (HD) vehicles are diesel operated. These percentiles were then used in calculating the emission factor for the total number concentration $EF(N_{\text{tot}})$ based on the corresponding LD ($\sim 1.1 \times 10^{14}$ particles $\text{veh}^{-1} \text{ km}^{-1}$) and HD ($\sim 5.2 \times 10^{15}$ particles $\text{veh}^{-1} \text{ km}^{-1}$) values, as reported by

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Gidhagen et al. (2004). As a result we obtained $EF(N_{\text{tot}}) = 6.23 \times 10^{14}$ particles $\text{veh}^{-1} \text{km}^{-1}$.

To be able to estimate modal emission factors we have plotted the average size distribution of particles measured while driving on Itäväylä (Fig. 3). The diameters along with the lower and upper limits of the six modes were prescribed to vary according to this figure, in the range of $1 \text{ nm} < d < 7.5 \text{ nm}$ for the nucleation mode 1 (nuc1), $7.5 \text{ nm} < d < 43.2 \text{ nm}$ for the nucleation mode 2 (nuc2), $43.2 \text{ nm} < d < 0.122 \mu\text{m}$ for the Aitken mode (Ait), $0.122 \mu\text{m} < d < 0.321 \mu\text{m}$ and $0.321 \mu\text{m} < d < 2.5 \mu\text{m}$ for the accumulation modes 1 and 2 (acc1, acc2), respectively, and $d > 2.5 \mu\text{m}$ for the coarse mode particles. The partitioning percents to each mode were calculated based on the measured total number concentration and concentrations of each size bin resulting in 9.5, 80.5, 8.7, 1.0, 0.3 and 0.001 percent, respectively. The modal emission factors were then obtained by multiplying $EF(N_{\text{tot}})$ by these numbers. Since the particle number concentration on Itäväylä is positively correlated with the traffic flow as seen in the Figure 4, the emission factors also depend on the traffic flow. Thus we have finally extended the modal emission factors to allow for the temporal variation by multiplying them by the measured traffic density profiles, separately to both directions, to obtain Q_i in particles $\text{m}^{-1} \text{s}^{-1}$.

For comparison, we also used the modal emission factors calculated based on the ELPI measurements by Sniffer. Yli-Tuomi et al. (2004) estimated the emission factor $EF(N_{\text{tot}})$ of 9.3×10^{15} particles per kg fuel for the highway conditions in HMA. A rough estimation of $EF(N_{\text{tot}}) = 8.7 \times 10^{14}$ particles $\text{veh}^{-1} \text{km}^{-1}$ can be obtained by assuming that the average fuel consumption for light duty vehicles (90%) is 8 l/100 km and for heavy duty (10%) around 20 l/100 km. The results are discussed in Sect. 4.1.

For the modal chemical composition of the exhaust particles LIPIKA data for exhaust particle number concentrations measured while driving on the highway was used, combined with the chemical composition data extracted from Norbeck et al. (1998) (considering exhaust particle emissions for light duty vehicles) and Shi et al. (2000) (containing data on exhaust particle emissions for heavy duty vehicles). The properties of the par-

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ticulate modes in the vehicle exhaust are presented in Table 1.

3.2 Evaluation of the particle size distribution and chemical composition of urban background

We utilized an average urban background particle size distribution (presented in Fig. 3) that is based on the LIPIKA-measurements at the site of Saunalahti. The average total number concentration of urban background particles (N_{tot}) was $1.1 \times 10^4 \text{ cm}^{-3}$. The diameters of particles and the limits of the six modes were fitted to be the same as in the case of emission modelling (presented in Fig. 3).

The evaluation of the chemical composition of the background particles was based on Pakkanen et al. (2001a, b), concerning ultrafine particles and Viidanoja et al. (2002), concerning fine and coarse particles. The properties of the particulate modes in urban background air are presented in Table 2. The nucleation mode particles were assumed to be mainly composed of organic carbon, whereas the Aitken and accumulation mode particles included the most abundant components, organic and elemental carbon (OC and EC), in a ratio of $\text{OC/EC} = 1.2$.

3.3 Atmospheric dispersion and aerosol dynamics models

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 utilises the meteorological input data evaluated with the meteorological pre-processing model MPP-FMI. The dispersion equation is based on a semi-analytic 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 Härkönen et al. (2002) and Karppinen et al. (2000a, b). The performance of the CAR-FMI model has previously been extensively evaluated both against the results of field measurement campaigns regarding NO_x , NO_2 and O_3 (e.g., Kukkonen et al., 2001), $\text{PM}_{2.5}$ (Tiitta et al., 2002), and the data of urban concentration monitoring networks (e.g., Karppinen et al., 2000b,

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Kousa et al., 2001).

The aerosol dynamics model MONO32 is a box model, which includes gas-phase chemistry and aerosol dynamics, and can be applied under clear sky conditions. The model uses monodisperse representation for particle size distribution with an optional number of size modes. In this work we have used six modes, two nucleation modes, one Aitken mode, two accumulation modes and one coarse mode. All particles in a mode are characterised by the same size and the same composition. Particles can consist of soluble material such as sulphuric acid, ammonium sulphate, ammonium nitrate and sodium chloride, organic carbon which can be soluble, partly soluble or insoluble, and insoluble material like elemental carbon and mineral dust. Size and composition of particles in any class can change due to multicomponent condensation of sulphuric acid and organic vapours as well as due to coagulation between particles. For a more detailed description of the MONO32 model and its evaluation against measurement data, the reader is referred to Pirjola and Kulmala (2000), Pirjola et al. (2003) and Pohjola et al. (2003).

3.4 Coupled modelling of the emissions, dispersion and aerosol processes

The dispersion model (CAR-FMI) is a three-dimensional Eulerian model with a temporal resolution of one hour, i.e., the emissions, background concentrations and meteorological conditions are assumed to be constant during that period. The aerosol dynamics model (MONO32) is a one-dimensional Lagrangian model; this model addresses the temporal evolution of aerosols and gases within a constant volume of air. The temporal resolution can be selected, but it is commonly of the order of seconds. The combination of these two models in order to form an integrated system is therefore not a trivial task. However, the principle of combining the two models is simple: the dispersion model should provide for the dilution of the Lagrangian air parcel, and the aerosol process model should then allow for the aerosol transformation within this temporally expanding volume.

There is no analytical solution for combining the equations of the CAR-FMI and the

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MONO32 models. However, the problem can be solved mathematically, if we use the dispersion model to provide for a more simple function that describes the dilution of the air parcel within a sufficiently good accuracy. We have therefore defined numerical power functions, $y=ax^{-b}=a(ut)^{-b}$, in which the coefficients a and b are positive, for approximating the dilution process, x (in m) refers to the distance from the source and u is the horizontal wind speed perpendicular to the highway. The coefficients a and b are determined separately for each meteorological case, using the CAR-FMI model.

The dilution was therefore first computed for an inert pollutant by the FMI-CAR model. The road was assumed to consist of a set of two line sources that were located in the middle of the three lanes, separately for both directions of the traffic. Only the dilution of vehicle exhaust was computed, background was not included in the dilution calculations.

The emission and dilution rates were taken into account in the aerosol process model by appropriately modifying the differential equations of the number and mass concentrations of particles. Dilution rate was obtained by derivating the above mentioned numerical power function as a function of time. To calculate the modal emission rates (particles $\text{cm}^{-3} \text{s}^{-1}$) the modal emission factors Q_i (particles $\text{m}^{-1} \text{s}^{-1}$) have to be divided by the width of three lanes to one direction to obtain the portion of emissions caught by the box, and by the box height since air in the box is assumed to be well mixed.

The time evolution of particle number concentrations (N_i) and the total mass concentration of different compounds $j(=1,2)$ ($M_{i,j}$) in i th ($=1,6$) size mode are given by

$$\frac{dN_i}{dt} = -0.5K_{ii}N_i^2 - \sum_{k=i+1}^6 K_{ik}N_iN_k - \lambda_{\text{dep},i}N_i + \frac{Q_i}{HW} - \frac{b}{t}(N_i - N_{bi}) \quad (1)$$

and

$$\frac{dM_{i,j}}{dt} = N_i I_{i,j} + \sum_{k=1}^{i-1} K_{ik} N_i N_k m_{k,j} - \left(\sum_{k=i+1}^6 K_{i,k} N_i N_k \right) m_{ij} - \lambda_{\text{dep},i} N_i m_{i,j} + \frac{Q_i}{Hw} m_{ei,j} - \frac{b}{\tau} (N_i m_{i,j} - N_{bi} m_{bi,j}) \quad (2)$$

where K_{ik} is the coagulation coefficient, $\lambda_{\text{dep},i}$ is the dry deposition velocity (s^{-1}) of the particles in size class i , H is the box height, $w=12$ m is the width of the lanes to one direction, Q_i is the modal emission factor (in particles $\text{m}^{-1} \text{s}^{-1}$), b is the dilution coefficient, N_{bi} is the number concentration of particles in background air in size class i given by Table 2. $I_{i,j}$ is the mass flux of condensable vapour j (sulphuric acid or organic vapour) to the particles in size class i given by Pohjola et al. (2003). In Eq. (2) $m_{i,1}$ refers to the sulphuric acid mass and $m_{i,2}$ to the organic mass in a particle in size class i ; subscripts b and e refer to the urban background and emitted exhaust particles, respectively. Due to different origin the background and exhaust particles are expected to have different composition.

One additional procedure is needed for integrating the emission modelling (in terms of the total emissions from the traffic flow) and the combined dispersion and aerosol dynamics modelling. The combined dispersion and aerosol process model addresses expanding air parcels that are transported along with the air flow over the road. For simplicity, we address here only cases, in which the wind direction is perpendicular to the road. First, one needs to assume an initial height of the air parcel 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. Due to the rapid temperature decrease immediately after the exhaust, nucleation has already occurred within this time-scale (Rönkkö et al., 2006); we have therefore assumed that the diameter of the smallest size mode was 4.7 nm. The initial dispersion is dependent on both the traffic-induced and atmospheric turbulence. Clearly, the height of the initial mixing volume cannot be smaller than the effective release height of the

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emissions. We have assumed that this initial air parcel height is equal to 0.80 m.

There is a continuous flux of vehicular emissions to the moving air parcel as long as the parcel is transported over the lanes; this is also the case for the modelling. The height of the parcel was assumed to increase linearly above the traffic lanes, in order to account for the effect of dilution during transport above the lane. For this linear dependence we have performed a semi-empirical fitting in the following way. The slope of the linear increase of the height of the air parcel was adjusted so that the total particle number concentration based on one of the 14 dispersion cases (case 10: 19 February at 14) agreed with the measured value, which was $1.83 \times 10^5 \text{ cm}^{-3}$ with $\text{SD} = 8.98 \times 10^4 \text{ cm}^{-3}$ (Pirjola et al., 2006). This numerical fit has been presented in Fig. 5. This height increases linearly above the lanes; this takes into account atmospheric dispersion. During the grass area between the lanes as well as downwind from the road, dilution is modelled using the CAR-FMI model and the height of the air parcel is therefore constant.

This procedure resulted in a slope of 0.17 m s^{-1} and this value was applied to all the simulations. The x-axis in Fig. 5 is the distance from the roadside instead of time, and since the slope of the air parcel height also depends on wind speed (1.6 m s^{-1} in this case), the slope in the figure is 0.106. The height of the air parcel is assumed to be constant while being transported over the central grass area of the road and after it has crossed the road; dilution and mixing with the background air are subsequently taken into account as described above. As a result, above the downtown direction the box height was increased from 80 cm to 1.56–2.84 m depending on the wind speed ($2.7\text{--}1.0 \text{ m s}^{-1}$), and during the eastern direction to 2.31–4.88 m, respectively.

In the model, particle growth was assumed to occur by condensation of H_2SO_4 and an organic vapour (Rönkkö et al., 2006); however, this vapour is not yet identified and its concentration and thermodynamic properties are therefore not well known. H_2SO_4 is formed photochemically from its precursor SO_2 whose concentration in the background air was assumed to be 1 ppb. The concentration of SO_2 in the exhaust gas was assumed to be $\sim 5.5 \times 10^{11} \text{ molecules cm}^{-3}$ ($\sim 20 \text{ ppb}$), in mega cities it might be

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even several hundred ppb (Seinfeld and Pandis, 1998), Condensable organic vapour concentration at the roadside was an unknown parameter, and therefore it was varied from 10^9 to 10^{12} molecules cm^{-3} . Hellén et al. (2006) have shown that the average winter concentrations of different VOC compounds at an urban site in Helsinki are $\sim 4.9 \times 10^4$ ng m^{-3} corresponding to 2.9×10^{11} molecules cm^{-3} . We assumed that 10% of these compounds are condensable. The background VOC concentration was assumed to be 2.5×10^8 molecules cm^{-3} (~ 10 ppt). SO_2 and organic vapour were diluted in the similar way as particles.

4 Results and discussion

The meteorological data used was from the period 17–20 February 2003, at time periods when the wind was directed from the road towards the measurement sites (total of 14 h with these conditions).

4.1 Modelled and measured concentrations

The scatter plot of the measured and predicted hourly total number concentrations (at 14 hourly cases) at all roadside measurement points (5–9 locations) is presented in Fig. 6. The overall average Index of Agreement (which is a measure of correlation of two sets of data) between the measured and predicted values of N_{tot} was 0.69. When looking at each day separately, IA varied from 0.96 on 17 February (2 cases) to 0.42 (4 cases) on 20 February while the Fractional Bias (a measure of the agreement of mean concentrations) varied between 1.02 on 17 February and -1.02 on 20 February.

Three example cases were chosen for a more detailed examination. These specific cases were selected in order to be able to address various traffic situations, prevailing wind speeds and atmospheric stratifications. The cases are: Case 1 on 17 February at 06:00–07:00 p.m. (Monday, evening hour after busiest rush hours, lower wind speed of 1.3 m s^{-1} , neutral stratification), Case 8 on 19 February at 08:00–09:00 a.m. (Wednes-

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day, morning rush hour, higher wind speed of 2.7 m s^{-1} , neutral stratification) and Case 11 on 19 February at 04:00–05:00 p.m. (Wednesday, afternoon rush hour traffic, lower wind speed of 1.3 m s^{-1} and stable stratification).

Measured and modelled total number concentrations of particles as a function of distance from the edge of the road for three example cases are presented in Figs. 7–9a). The measured total number concentration is calculated as the sum of particles smaller than 50 nm recorded by SMPS and particles larger than 50 nm recorded by ELPI (see details in Pirjola et al., 2006). As an example in the 17 February case, the measured total number concentration decreased by 43% between the closest measurement point at 9 m distance and 65 m distance from the edge of the road, and by 58% between the distances of 37 m and 95 m from the edge of the road. Number concentration in the nucleation 1 and 2 and Aitken modes decreased by 40–54% between the closest measurement point at 9 m distance and 65 m distance from the edge of the road, and by 55–60% in the nucleation modes and 36% in the Aitken mode between the distances of 37 m and 95 m from the edge of the road. In the accumulation modes, the modewise number concentration decrease between the distances of 9 m and 65 m was 18–20%, and between the distances of 37 m and 95 m the decrease was 14% for the accumulation 1 mode while the accumulation 2 mode number concentration increased by 1%.

Clearly, such results are also dependent on atmospheric conditions, and could therefore be different, e.g., for various climatic regions and the seasons of the year. However, these results are similar to the findings of Zhu et al. (2002b) and Hitchins et al. (2000), who found that total number concentration decreased to approximately half of its original value at a distance of 30 m at somewhere between the distances of 90 and 150 m. They also found that the number concentration of small particles decreased more rapidly than that for large particles with increasing distance from a freeway.

In all cases, the total number concentrations are in maximum at the roadside ~35% higher by using the emission factors calculated based on the results by Yli-Tuomi et al. (2004) than by Gidhagen et al. (2004). As the distance from the road increases up to 170 m the difference decreases to 25–27%. Neither of the emission factors by

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Yli-Tuomi et al. (2004) or by Gidhagen et al. (2004) produced consistently better results in comparison with the measured data.

Predicted and measured size-number concentrations distributions at the measurement distances from the road are presented in panels b-c of Figs. 7–9. To be able to compare the results, the first seven and the next (12) size bins by SMPS were summed to form two size bins and the number concentration was calculated from the lognormal values. Note that Figs. 7–9b–c gives the concentrations in each mode, not lognormal size distribution, as the particles in a mode are of exactly one diameter (monodisperse model).

During the 17 February case period, the measured N_{tot} was on average (over all the measurement distances) 87% of the modeled N_{tot} values, during the 18 February case period it was 129%, and during the 19 February case period, about 76%. A shift towards larger diameter is seen in the modeled smaller nucleation mode size bin in all three cases, by 14%, 19% and 14% in chronological order of the cases. This shift is less pronounced in the second nucleation bin (2–3%) and the Aitken bin (1–2%), and a bit more pronounced in the two accumulation modes (3.6–4.3% for first accumulation bin and 2.4–3.9% for the second accumulation mode).

Ketzel et al. (2004) found that for a roadside site in Copenhagen, the maximum in size distribution is at about 22 nm in traffic, 20–30 nm at kerbside and 50–60 nm at a rural site. As presented in Fig. 3, the maximum in the size distribution at the Itäväylä site was at 18 nm in traffic, about 19 nm at 9 m distance from the edge of the highway, and 16–17 nm at the background site.

We have also evaluated the evolution of particle size distribution assuming various concentrations of condensable vapours for the 17 February case. These results are presented in the Fig. 10; however, the results for the coarse mode have not been shown, as there is no variation in terms of the concentration of the condensable vapour. The percentual changes of modewise number concentrations and particle diameters from 9 m distance to 125 m distance from the edge of the road are presented in the Table 3. The highest concentration of condensable organic vapour (C_{org}) used,

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10^{12} molecules cm^{-3} , was clearly too high, as five of smallest predicted size modes at the distance of 37 m had already grown larger than 10^{-7} m in diameter. For the nucleation 1 mode, the $C_{\text{org}}=10^9$ molecules cm^{-3} produced diameter values closest to the measured ones, for nucleation 2 mode, the $C_{\text{org}}=10^{10}$ molecules cm^{-3} was the best, and for the other size bins, the predicted sizes were very similar. Considering all measured data, the changes of the predicted particle number concentrations were of the same order as the corresponding measured values with both values of C_{org} , 10^9 and 10^{10} molecules cm^{-3} in the nucleation and Aitken modes, overestimated by factor of 2 for accumulation 1 mode and by factor of 7 for accumulation 2 mode, and underestimated by factor of 43 for the coarse mode.

The importance of aerosol processes was also evaluated for the 17 February case for the six size bins by modelling runs with all processes included being compared to modelling runs with each aerosol process being excluded from the model equations. As an example, the results considering the particle effects on particle diameter in the case with C_{org} being 10^{10} molecules cm^{-3} are presented on the right side of the Table 3. The effect of condensation is most pronounced for the nucleation 1 mode, where the model runs with no processes or only coagulation included seems to be more accurate. For the nucleation 2 mode, the results with condensation only included in the model run are more accurate. For the other size bins, the model runs produce similar results whether condensation and coagulation are included or not.

The measured decrease of the diameters of the nucleation 2 and coarse mode particles is not reproduced by the modelling. Jacobson and Seinfeld (2004) found that at 30 m downwind, the difference in number concentration between total number concentrations in the no-coagulation and the coagulation case suggested that about 2% of the decrease in particle concentration between 0 and 30 m was due to coagulation and most of the rest was due to dilution (some was due to dry deposition as well). This result, though, depends substantially on the emission rate. In our example case on 17 February, the difference in total number concentration (not presented) in model runs with all aerosol processes included and coagulation excluded, the effect of coagulation

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was about 1% decrease in particle total number concentration at 37 m distance from the edge of the road. In the model run with condensation excluded the particle number concentration was increased by 0.3% at 37 m distance compared to the run with all aerosol processes included.

5 4.2 Comparison of modelled values and the background

The total particle number concentration of the urban background used in the modeling was $1.11 \times 10^4 \text{ cm}^{-3}$, while at the farthest modelled distance of 350 m (wind speed in this case was 5.2 m s^{-1} and deviation of wind speed from perpendicular to the highway was about 8°) the total number concentration was still above the background with the value $1.32 \times 10^4 \text{ cm}^{-3}$. The corresponding size number distributions are presented in Fig. 11. The modelled particle mode sizes and mode wise number concentrations are larger (by 6–50% and 4–145%, respective) than the input values with particle sizes smaller than 120 nm, while modelled particles larger than this value were 35% smaller in size and 22% more in number concentration.

Taking the sensitivity to wind speed and the used dilution model into account, this is similar to the results of Zhu et al. (2002a), who found in their study that UF particle number concentration measured at 300 m downwind was indistinguishable from the upwind background number concentration. Zhang and Wexler (2004) also state, based on theoretical analysis, that road plumes become ambientlike in their particulate matter characteristics at about 300 m distance from the road. However, neither of these studies assessed the evolution of the particle diameters in the comparison.

5 Conclusions

This study has applied a recently made measurement campaign that contains data on particle number concentrations, size distributions and the concentrations of NO_x performed in a roadside environment in Helsinki, for the evaluation of combined dispersion

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and aerosol process models. The experimental dataset is extensive and versatile both in terms of the measured aerosol quantities and the locations of measurement sites. The data contains both measured particle number concentrations and size distributions at several distances from the road, as well as urban background measurements and number concentration measurements on the road. The main limitations of the experimental campaign are probably associated with the fairly limited temporal duration of the campaign, i.e., two weeks. In addition, there were only four days suitable for modelling of the influence of the emissions on the road (downwind cases on 17–20 February 2003). Clearly, the importance of various aerosol processes depends on the spatial scale; this study addresses the scale that is smaller than 200 m regarding measured data and 360 m concerning the model computations.

In this study, we have compared the measured data with the predictions of the road network dispersion model CAR-FMI used in combination with an aerosol process model MONO32. Compared to previous studies in this area (e.g., Pohjola et al., 2003 and 2004), we have utilised a state-of-the-art atmospheric dispersion model and atmospheric boundary layer scaling. It is straightforward to show that there is no analytic mathematical solution for combining the equations of these two models. We have therefore developed an approximative mathematical method that (i) presents the dilution of pollution as simplified equations that are based on the roadside dispersion model, and (ii) combines these equations mathematically with the set of differential equations that constitute the aerosol process model. The same or similar mathematical procedure could probably be used also for coupling other dispersion models representing various spatial scales, and aerosol process models. Regarding the application of the models, the probably most uncertain aspect is the assessment of the initial air parcel mixing height, and the modelling of its increase over the road.

Considering the evolution of total number concentration, dilution was shown to be the most important process in the considered distance scale. The influences of coagulation and condensation on the number concentrations of particle size modes were found to be negligible on this distance scale. Condensation was found to affect the evolution

of particle diameter in the two smallest particle modes. The assumed value of the concentration of condensable organic vapour of 10^9 – 10^{10} molecules cm^{-3} resulted in particle sizes that were closest to the measured values.

The particle number size distribution of the plume originated from the traffic emissions in the major road studied approached closely that of the urban background at a distance of approximately 300 m. Clearly, this result depends on the magnitude of the urban background, the traffic density on the road studied and the dispersion conditions. However, the result obtained in this study was very similar to the previous estimates by Zhu et al. (2002a) and Zhang and Wexler (2004).

Three example cases were chosen for a more detailed examination. These specific cases were selected in order to be able to investigate various traffic situations, prevailing wind speeds and atmospheric stratifications. In Case 11 (at 04:00–05:00 p.m. on 19 February 2003), the total number concentration at the edge of the road was the highest, mainly due to the highest traffic density (busy evening traffic) and the fact that there was more traffic on the lane closest to the measurement sites.

A specific limitation of the campaign is that both the measurements of the particle size distributions and those of the total number concentrations (N_{tot}) at various distances from the road are not simultaneous, but from subsequent time periods, as only one measurement van was available for the campaign. The concentrations at different distances were measured during approximately 5–10 min of parking at that particular location, and the measurements were started at the site that is closest to the edge of the road. This causes a stochastic variation to the measured concentration data, when the data at various distances from the road are inter-compared. Clearly, the measured concentrations can therefore randomly vary in terms of the short-term variations of the traffic densities, driving speeds and meteorological conditions, while the modeling results are based on hourly averaged input values of the meteorological and traffic flow data. The commonly occurring short-term variation of the measured concentrations at the stationary measurement site (the site at the distance of 9 m from the edge of the road) was about 30% during one measuring cycle of approximately one hour (Pirjola et

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al., 2004).

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Table 1. The properties of the vehicle exhaust particle emissions.

| Particle size mode | Dry radius (nm) | % of total number concentration (cm ⁻³) | Mass composition (%) | | | | | | |
|--------------------|-----------------|---|----------------------|----------------|------------------|--------------|---------|-------------------|------------------|
| | | | Sulphuric acid | Organic carbon | Elemental carbon | Mineral dust | Seasalt | Ammonium sulphate | Ammonium nitrate |
| Nucleation1 | 2.35 | 9.5 | 5 | 90 | 0 | 0 | 0 | 5 | 0 |
| Nucleation2 | 9.0 | 80.5 | 5 | 90 | 0 | 0 | 0 | 5 | 0 |
| Aitken | 36.4 | 8.7 | 0 | 27 | 64 | 7 | 0 | 2.2 | 0.3 |
| Accumulation1 | 103 | 1 | 0 | 26 | 63 | 9 | 0 | 3 | 0.3 |
| Accumulation2 | 226 | 0,3 | 0 | 25 | 63 | 9 | 0 | 3 | 0.3 |
| Coarse | 1010 | 9e-4~0 | 0 | 0 | 0 | 100 | 0 | 0 | 0 |

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Table 2. The properties of particulate matter in the urban background air.

| Particle size mode | Dry radius (nm) | Number concentration (cm ⁻³) | Total mass (ng m ⁻³) | Mass composition (%) | | | | | | |
|--------------------|-----------------|--|----------------------------------|----------------------|----------------|------------------|--------------|----------|-------------------|------------------|
| | | | | Sulphuric acid | Organic carbon | Elemental carbon | Mineral dust | Sea salt | Ammonium sulphate | Ammonium nitrate |
| Nucleation1 | 2.35 | 1.32e2 | 1.22e-2 | 7 | 68 | 19 | 0 | 0 | 0 | 6 |
| Nucleation2 | 9.0 | 7.75e3 | 4.02e+1 | 7 | 68 | 19 | 0 | 0 | 0 | 6 |
| Aitken | 36.4 | 2.63e3 | 9.03e+2 | 21 | 24.5 | 19 | 12 | 2.5 | 0 | 21.3 |
| Accumulation1 | 103 | 5.66e2 | 4.34e+3 | 21 | 24.5 | 19 | 12 | 2.5 | 0 | 21.3 |
| Accumulation2 | 226 | 2.94e1 | 2.4e+3 | 21 | 24.5 | 19 | 12 | 2.5 | 0 | 21.3 |
| Coarse | 1010 | 1.7e-1 | 1.0e+4 | 2 | 23.4 | 4.56 | 59 | 7 | 0 | 4 |
| Total | | 1.11e4 | 1.77e+4 | 10.2 | 24 | 10.8 | 38.5 | 5 | 0 | 11.5 |

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Table 3. The percentual changes of modewise number concentrations and particle diameters from 9 m distance to 125 m distance from the edge of the road in the Case 1, 17 February at 06:00–07:00 p.m.. $\Delta X = (X(125\text{ m}) - X(9\text{ m})) / X(9\text{ m}) * 100$, where $X = \#$ or D . Less or smaller particles at 125 m; $\Delta < 0$, more or larger particles at 125 m; $\Delta > 0$.

| Set of C_{org} and processes included | | | Change of umber concentration in mode: $\Delta\#$ | | | | | | Change in diameter of mode: ΔD_p | | | | | |
|--|------------------|------------|---|------|-----|------|------|-----|--|------|-----|------|------|------|
| | C_{org} | Proc-esses | Nuc1 | Nuc2 | Ait | Acc1 | Acc2 | Coa | Nuc1 | Nuc2 | Ait | Acc1 | Acc2 | Coa |
| Varying C_{org} | 1e9 | all | -77 | -69 | -57 | -46 | -69 | -1 | 4.3 | 0.9 | 1.7 | 3.9 | 3.3 | 0.2 |
| | 1e 10 | all | -76 | -69 | -57 | -46 | -69 | -1 | 14 | 1.9 | 1.0 | 4.3 | 2.9 | 0 |
| Varying set of processes included | 1e 12 | all | -75 | -69 | -57 | -46 | -69 | -1 | 190 | 144 | 59 | 13 | 10 | 0 |
| | 1e 10 | none | -76 | -69 | -57 | -46 | -69 | -1 | 0 | 0.1 | 1.6 | 4.6 | 3.5 | 0 |
| | 1e 10 | cond only | -76 | -69 | -57 | -46 | -69 | -1 | 14 | 1.9 | 1.0 | 4.5 | 3.0 | 0 |
| | 1e 10 | coag only | -78 | -69 | -57 | -46 | -69 | -1 | 0.4 | 0.2 | 1.6 | 3.6 | 3.5 | 0 |
| Measured data | | | -76 | -80 | -56 | -23 | -10 | -43 | 1.7 | -1.8 | 0.1 | 15 | 0.3 | -7.5 |

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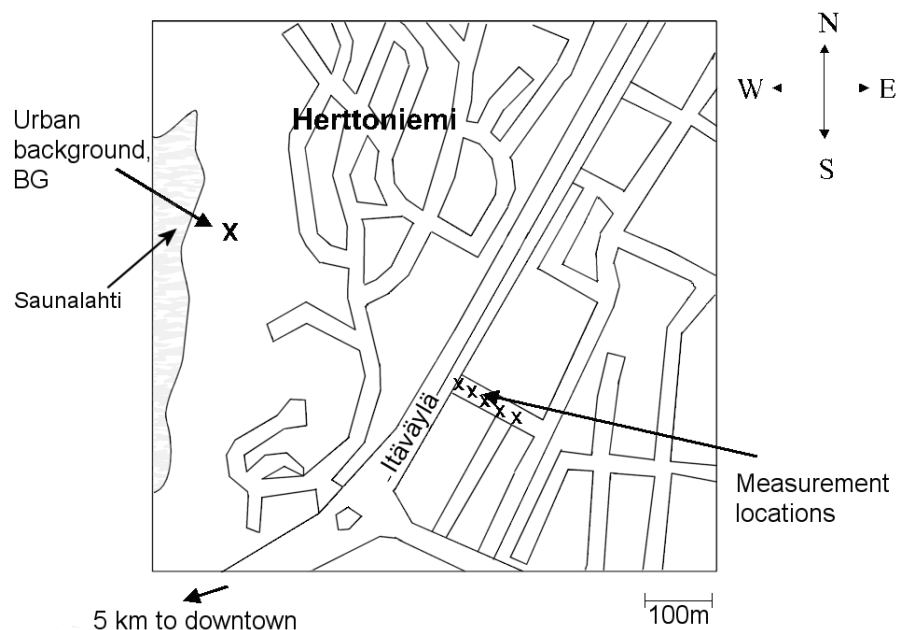


Fig. 1. Schematic map of the measurement site and its surroundings (Pirjola et al., 2006). The mobile laboratory measurements were conducted while the van was parked in several locations on a minor road (Työnjohtajankatu) that is approximately perpendicular to the major road studied (Itäväylä) and while driving on the major road.

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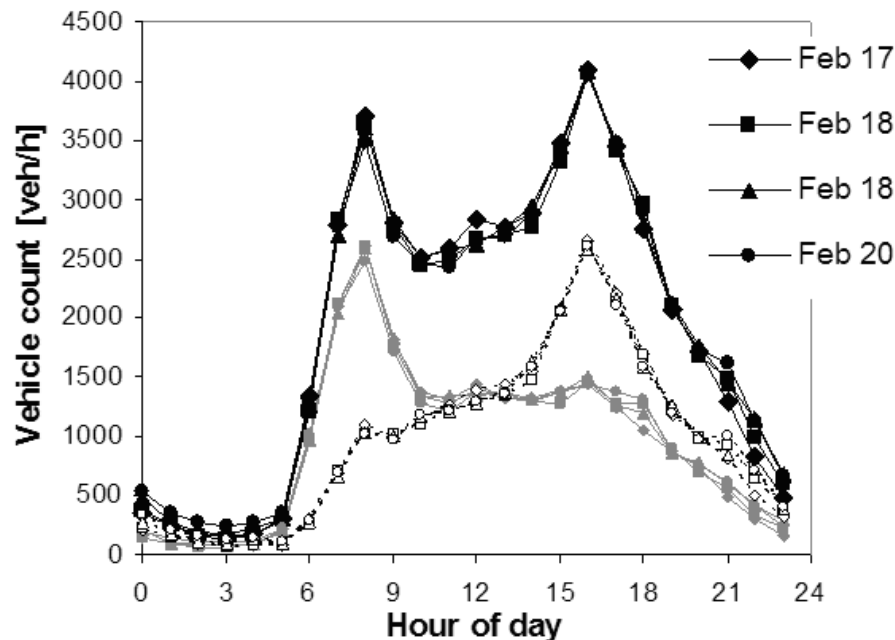


Fig. 2. The traffic densities measured at the traffic count site of Kulosaari for the days 17–20 February 2006. Total hourly traffic counts are indicated with the four bold black lines, traffic counts towards the city center are indicated with gray lines, and traffic counts directed away from the city are indicated with black dotted lines.

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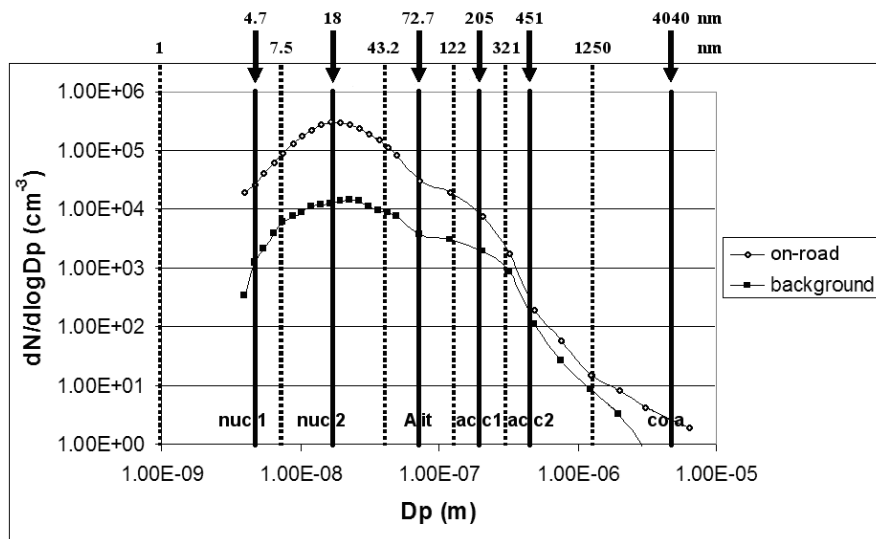


Fig. 3. Average size distribution of particles measured while driving on Itäväylä (open circles), and that for the average urban background measured at the station of Saunalahti (black squares). The solid vertical lines show the assumed initial diameters of the six size modes used in the model, and the dashed vertical lines show the initial boundaries between these size modes. The numerical particle diameter values that correspond to these vertical lines have also been shown in the upper part of the figure.

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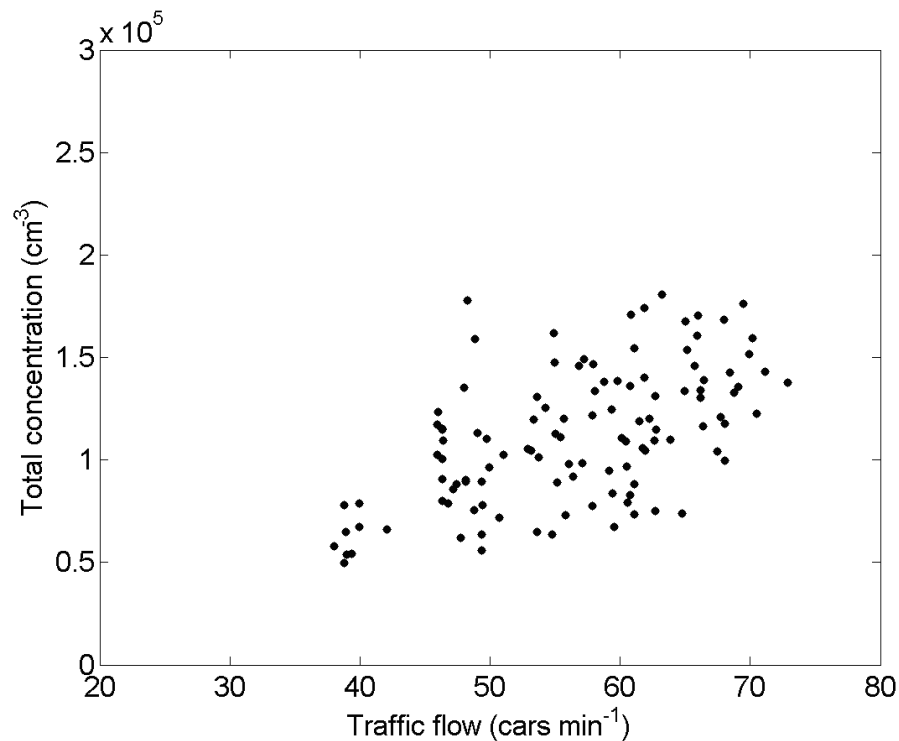


Fig. 4. Measured total particle number concentration at a distance of 9 m from the roadside as a function of traffic flow of the major road (Itäväylä).

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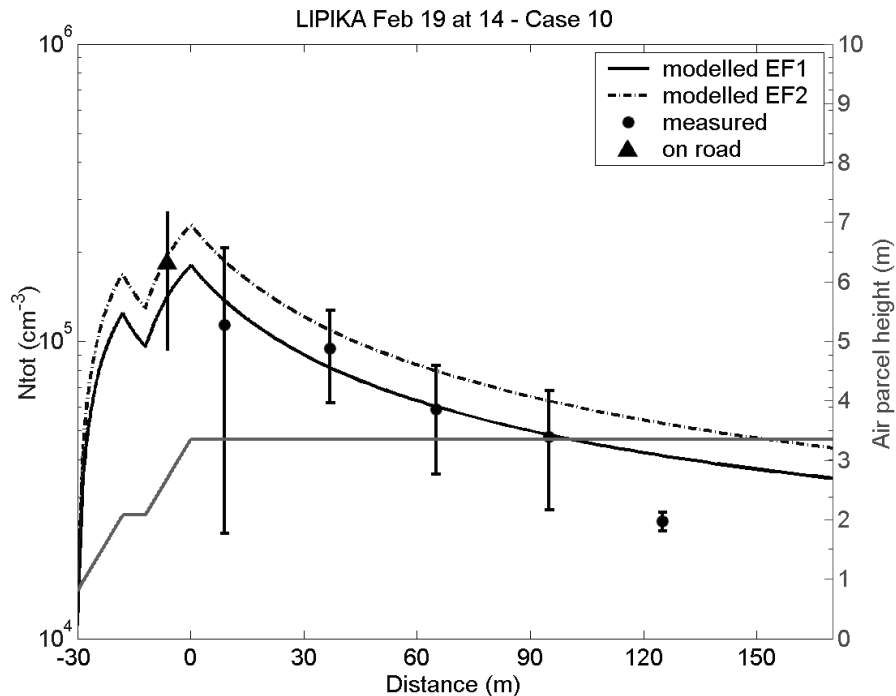


Fig. 5. Measured and predicted results for case number 10 at 02:00 p.m. on 19 February 2003. The predicted results were computed using two sets of emission factors: EF1 refers to those adopted from Gidhagen et al. (2004) whereas EF2 refers to those from Yli-Tuomi et al. (2004). The measured concentrations on road (adopted from Pirjola et al., 2006) and in the roadside are shown as a black triangle and black circles, respectively, and the vertical bars show their standard deviations. The semi-empirical fitting of the air parcel height was made according to this case; the figure therefore also shows the air parcel height. See details in the text.

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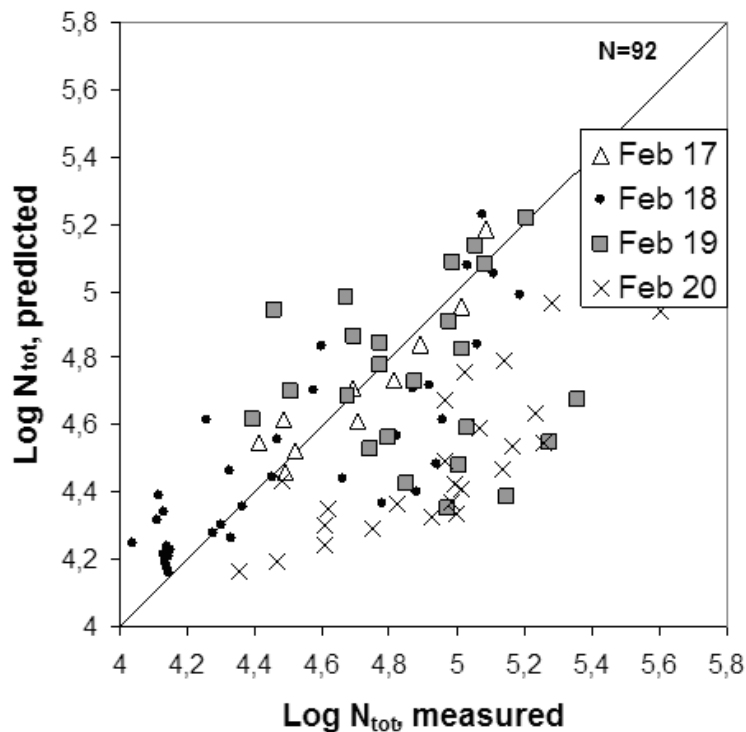


Fig. 6. Predicted total number concentrations versus measured total number concentrations for all studied cases at all roadside measurement locations. The legend shows the measurement days.

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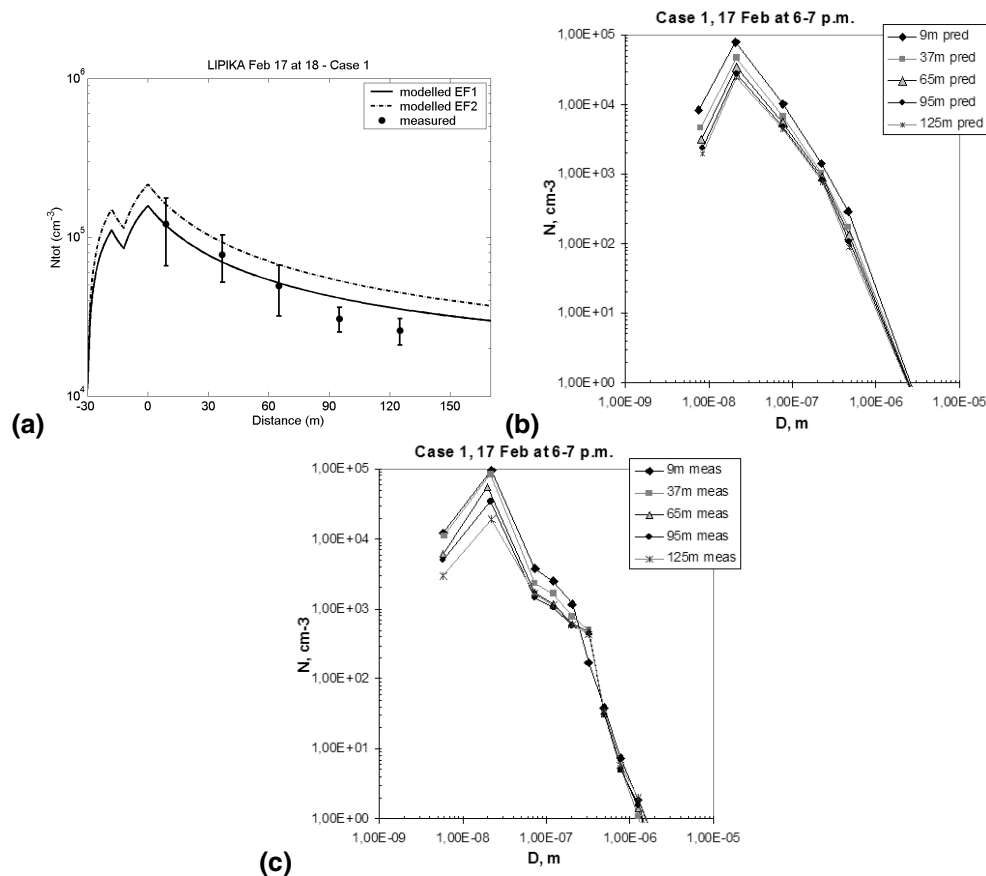


Fig. 7. Measured and predicted results at 06:00–07:00 p.m. on 17 February 2003. **(a)** Measured and modelled total number concentrations of particles as a function of distance from the edge of the road. **(b)–(c)** predicted and measured number concentration distributions at various measurement distances from the road.

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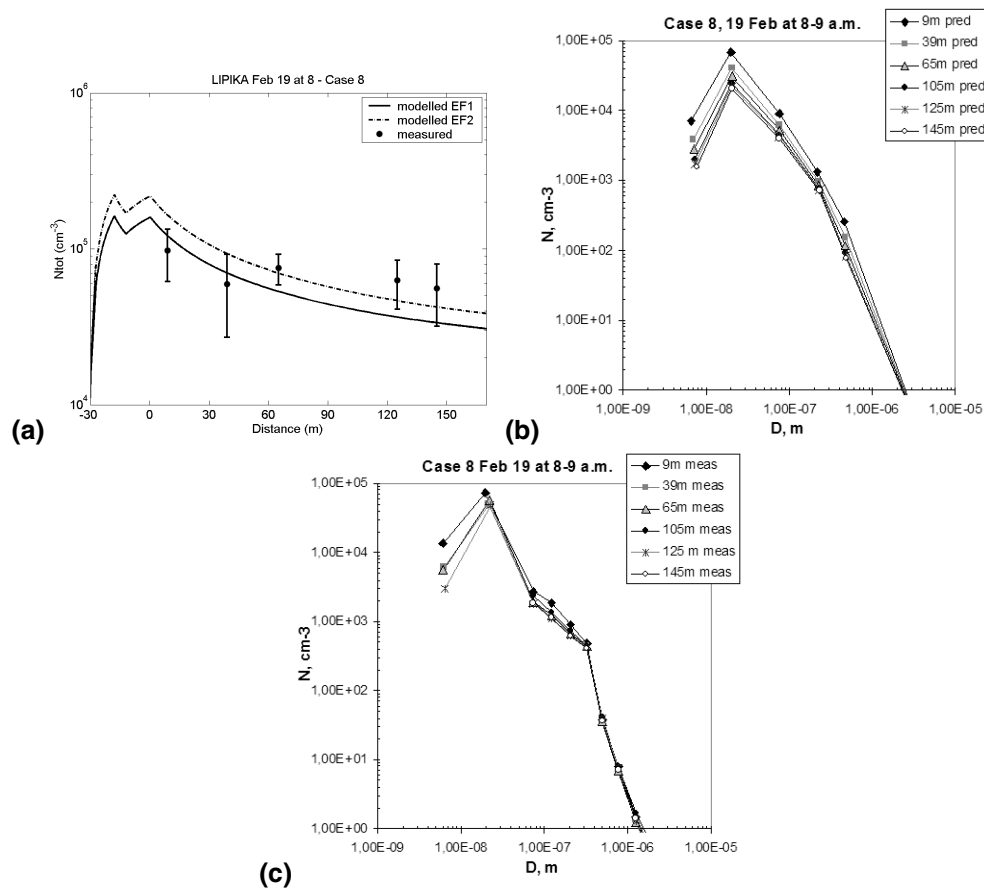


Fig. 8. Measured and predicted results at 08:00–09:00 a.m. on 18 February 2003. **(a)** Measured and modelled total number concentration of particles as a function of distance from the edge of the road. **(b)–(c)** predicted and measured number concentrations distributions at various measurement distances from the road. 2853

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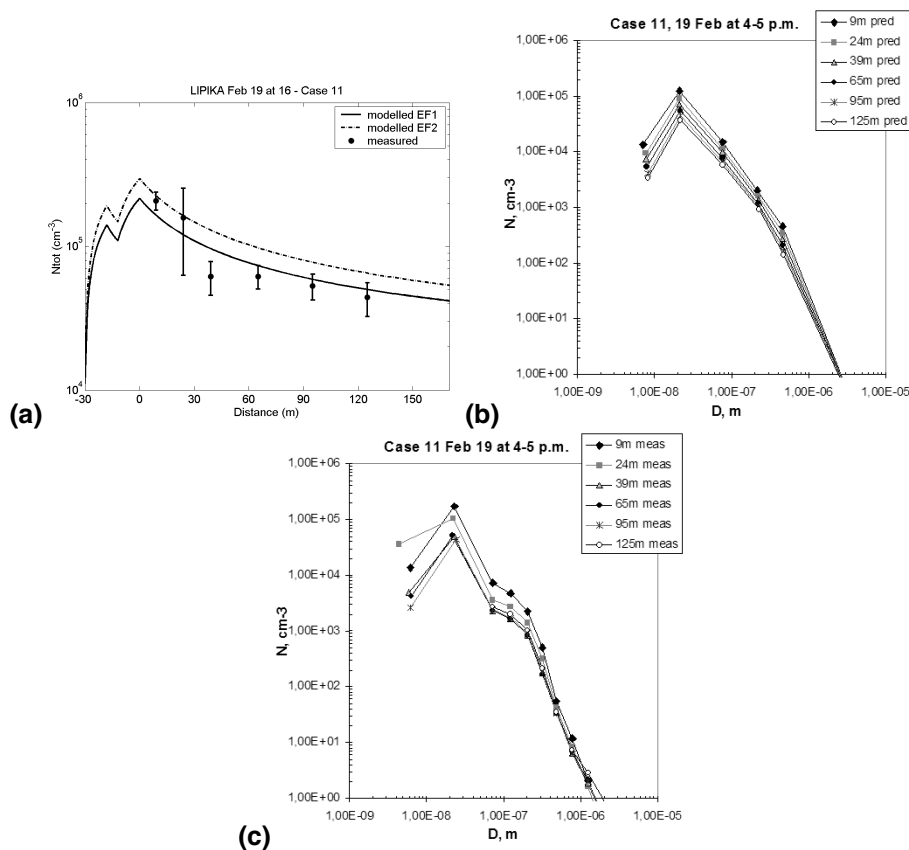


Fig. 9. Measured and predicted results at 04:00–05:00 p.m. on 17 February 2003. **(a)** Measured and modelled total number concentrations of particles as a function of distance from the edge of the road. **(b)–(c)** predicted and measured number concentration distributions at various measurement distances from the road.

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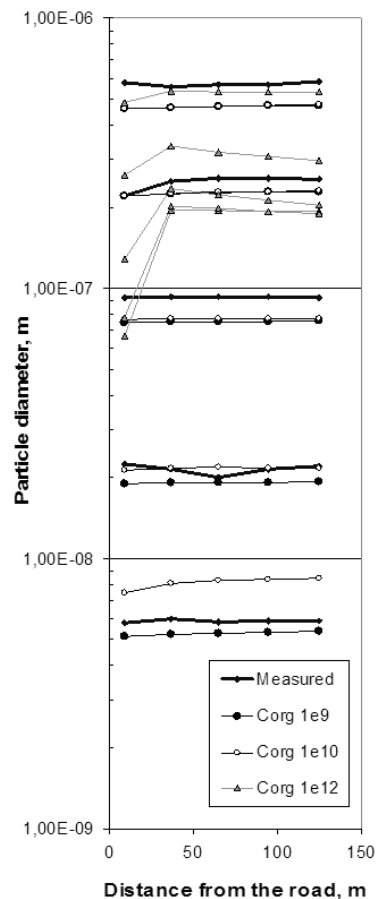


Fig. 10. Measured and modelled particle diameters as a function of the distance from the edge of the road for Case 1 at 06:00–07:00 p.m. on 17 February 2003. The modelled diameters have been computed assuming three different concentrations of the condensable organic vapour.

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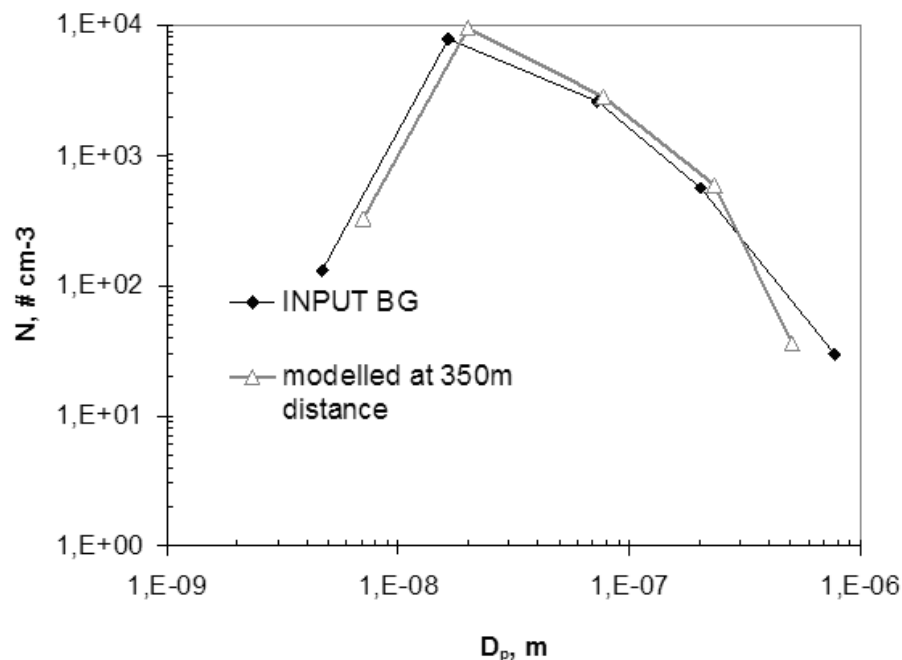


Fig. 11. The particle number size distribution of the urban background (squares) used as input values in the aerosol modelling (Table 2), and the modelled particle number size distribution at the distance of 351 m from the road, both of these on at 10:00–11:00 p.m. on 18 February 2003. The wind speed was 2.7 m s^{-1} and direction $\sim 330^\circ$.

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