



## **Black carbon concentration trends in Helsinki during 1996?2005**

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## Black carbon trends in Helsinki

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# Black carbon concentration trends in Helsinki during 1996–2005

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## Abstract

The black carbon (BC) concentration trends were studied during ten years in Helsinki, Finland. Measurements were made in three campaigns between 1996 and 2005 at an urban area locating two kilometres from the centre of Helsinki. The first campaign was from November 1996 to June 1997, the second from September 2000 to May 2001 and the third from March 2004 to October 2005. In this study, only data from winter and spring months was analysed. The effect of traffic and meteorological variables on the measured BC concentrations was studied by means of a multiple regression analysis, where the meteorological data was obtained from a meteorological pre-processing model (MPP-FMI).

During the ten years, the campaign median BC concentrations were found to decrease slightly from 1.11 to 1.00  $\mu\text{g m}^{-3}$ . The lowest campaign median concentration (0.93  $\mu\text{g m}^{-3}$ ) was measured during the second campaign in 2000–2001, when also the lowest traffic rates were measured. The strongest decrease between campaigns 1 and 3 was observed during weekday daytimes, when the traffic rates are highest. The variables affecting the measured BC concentrations most were traffic, wind speed and mixing height. On weekdays, traffic had clearly the most important influence and on weekends the effect of wind speed diluted the effect of traffic. The affecting variables and their influence on the BC concentration were similar in winter and spring. The separate examination of the three campaigns showed that the effect of traffic on the BC concentrations had decreased during the studied years. This reduction was caused by cleaner emissions from vehicles, since between years 1996 and 2005 the traffic rates had increased. A rough estimate gave that vehicle number-scaled BC mass concentrations have decreased from 0.0028 to 0.0020  $\mu\text{g m}^{-3}$  between campaigns 1 and 3.

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## 1 Introduction

It is well known that atmospheric particulate matter (PM) has adverse health effects, especially in urbanized areas (Pope et al., 2002; Keeler et al., 2005). One of the main constituents of PM has been identified to be black carbon (BC), particularly in primary particles originating from combustion sources. The major anthropogenic source of BC is incomplete combustion of fossil and biomass fuels. These include, e.g., residential heating, traffic and power plants. In traffic, especially diesel engines are known to emit BC (e.g. Watson et al., 1994). Szidat et al. (2006) estimated that almost all BC in Zürich originates from anthropogenic sources and the contribution of biogenic emissions is insignificant. Characteristic to BC is its ability to absorb solar radiation very effectively. Thus, it plays an important role in climate change by warming the atmosphere similarly to greenhouse gases (Jacobson, 2001; Novakov et al., 2003). In addition to this global effect, black carbon has also a local effect on visibility (Jiang et al., 2005) and small carbonaceous particles are considered to have severe effects to human health, for instance to cardiopulmonary and respiratory diseases (Stoeger et al., 2006). Because of its global and local effects, it is important to understand the nature and sources of BC particles. To common knowledge black carbon is a primary emission and it is not produced in secondary reactions. It is inert in the atmosphere and can be considered as a good tracer for combustion emissions (Kendall et al., 2001). Most BC is found in small particles (e.g. Ruellan and Cachier, 2001). Kerminen et al. (1997) studied diesel vehicle exhausts and found that BC mass concentration peaked with aerodynamic diameter of  $0.1\text{ }\mu\text{m}$ . Due to their size, the average time these particles spend in the atmosphere is around 6 days and thus can be transported some thousands of kilometres (Khan et al., 2006).

The crucial question is how urban BC concentrations have developed during the last decades. The main sources of BC have changed because of fuel and technology improvements, both in industrial and commercial sectors. In the 20th century, BC emissions decreased due to the changes in coal usage and improvements in diesel

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technology (Novakov et al., 2003). In the United Kingdom, the decrease in BC emissions and concentrations has been detected in the last 40 years (Novakov and Hansen, 2004). The reductions can be explained by the improved technology, which decreases the emission factors (emitted BC per unit mass of fossil fuel) and cleaning methods. In Europe, the yearly BC emissions decreased from 0.89 to 0.68 Tg between 1990 and 2000 (Kupiainen and Klimont, 2007). This change was mainly due to the development in Eastern Europe, where significant decrease was observed especially between 1990 and 1995. The BC concentration changes during the past ten years were studied in Tokyo and a yearly decrease of  $0.88 \mu\text{g m}^{-3}$  was observed (Minoura et al., 2006). The main reason was reported to be the decreased emissions from vehicles.

In Helsinki, Finland, BC mass concentration measurements have been conducted in three campaigns since 1996. Measurements have been made at the same site in Vallila located next to one of the main roads leading to downtown of Helsinki. In Vallila, the annual contribution of BC to  $\text{PM}_{2.5}$  and  $\text{PM}_{10}$  was found to be 14% and 7% in 2000 and 2001, respectively (Viidanoja et al., 2002). The most important source of BC has been identified to be local traffic, which contributes 63% to the concentrations on working days (Pakkanen et al., 2000). Other main contributors are long-range transport and other local sources than traffic. The traffic emissions have been affected by an increase in the number of vehicles during ten years. On average, traffic rates have increased 12% in Helsinki between 1996 and 2005 (Lilleberg and Hellman, 2006). It is reasonable to assume that especially the number of diesel vehicles has increased in Helsinki, since the fraction of diesel powered cars, vans, busses and lorries has increased from 20 to 30% in Finland during this time (Mäkelä, 2006). At the same time, exhaust emissions from vehicles have decreased due to developments in the after-treatment and cleaning methods of car exhausts.

The purpose of this work is to study BC concentration trends during the past ten years and to analyze reasons to the observed behaviour. The analysis is done by means of data from three measurement campaigns made between 1996 and 2005. In Sect. 2, the measurements and used methods are introduced. The campaign trends

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with weekly and diurnal cycles are studied in Sect. 3.1. Sect. 3.2 summarizes traffic trends during the measurement periods and that, how much of the BC concentration changes can be explained by traffic and how much by sole meteorology, are studied via multiple regression analysis in Sect. 3.3. Finally, conclusions are presented in Sect. 4.

5 **2 Measurements and methods**

2.1 Measurement site

Helsinki (60°10'N, 24°56'E) itself is located on a relatively flat land on the coast of Gulf of Finland. The area of the city is 686 km<sup>2</sup> with 560 000 inhabitants. Helsinki together with the neighbouring cities (Vantaa, Espoo, Kauniainen) forms the Helsinki metropolitan area with a total area of 1460 km<sup>2</sup> and 1 million inhabitants. The measurements took place in Vallila about 2 km northeast from the downtown of Helsinki (Fig. 1). The measuring site represents a typical urban area in Helsinki and it is in a small opening surrounded by 5–7 storey buildings. It is situated 14 m away from the nearest road, Hämeentie. This is one of the main roads leading to the city centre and its traffic loads are high especially during rush hours. The number of buses using diesel fuel is considerable on the road.

The traffic rates are monitored by the Helsinki City Planning Department. The nearest automatic traffic counting point is by the Itäväylä road on the Kulosaari Bridge, about 1.7 km southeast from the BC measuring site (Fig. 1). Traffic data is logged once an hour, and during rush hours four times per hour.

2.2 Black carbon measurements and data selection

The BC measurements were made during three campaigns between 1996 and 2005. The first campaign was from November 1996 to June 1997, the second from September 2000 to May 2001 and the third from March 2004 to October 2005. Data periods

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during these campaigns were incomplete due to, e.g., power cuts and maintenance operations. Thus, comparison between different years was done for selected periods, when data from all campaigns existed. In the selection, more weight was given to campaigns 1 and 3, which had 75% percent of data coverage. For campaign 2, the percentage was 55%. Four comparable periods were found, two at wintertime and two at springtime. All together 82 whole days from all campaigns were chosen (Table 1).

All measurements were carried out with the aethalometers (Hansen et al., 1984). It is an optical instrument where aerosol particles are collected on a filter (quartz tape) and the light transmittance through the filter is measured. To avoid the optical saturation, the filter spot changes at a certain light transmittance value. A more detailed description about the measurement technique is presented in Hansen et al. (1984). Sample air was taken using an inlet about 3.5 m above ground level and fed into the aethalometer inside a measurement container. Only particles with aerodynamic diameter smaller than  $2.5\text{ }\mu\text{m}$  were sampled. The aethalometer model, the measuring time resolution and the flow rate varied between different campaigns. In 1996 and 1997, measurements were made with a one-wavelength (880 nm) aethalometer (Magee Scientific Aethalometer, model AE-14) with a 10-min time resolution. The flow rate was 16.7 lpm. Two different aethalometers operated during campaign 2. From September 2000 to mid-January 2001, a model AE-20 with a 20-min time resolution was used and from mid-January to May 2001 measurements were carried out with a 10-min resolution with a multiwavelength model AE-30 (Magee Scientific Aethalometer). In both cases, the same wavelength 880 nm was used for BC measurements and the flow rates were 5.6 lpm and 5.4 lpm, respectively. During campaign 3, measurements were made with a one-wavelength (880 nm) aethalometer (Magee Scientific Aethalometer, model AE-16) at a 5-min time resolution and with a flow rate of 5.2 lpm. The mass absorption cross section of BC used to convert the aethalometer raw signals to BC mass concentrations was  $16.6\text{ m}^2\text{ g}^{-1}$  during all campaigns.

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## 2.3 Effect of black carbon loading and the correction method

With an aethalometer, the BC concentrations are calculated from the rate of change of attenuation of the light beam. The aethalometer software assumes that the relationship between the rate of change of attenuation and the BC concentration is linear. However, it has been observed that this relationship is usually nonlinear (e.g. Weingartner et al., 2003; Arnott et al., 2005; Virkkula et al., 2007). This nonlinearity usually leads to underestimation of BC when the filter gets darker, i.e., when attenuation increases. Virkkula et al. (2007) developed a simple algorithm to correct the data. The corrected BC concentrations were calculated with the equation

$$BC_{\text{CORRECTED}} = (1 + k \cdot ATN) BC_{\text{NON-CORRECTED}}, \quad (1)$$

where ATN is the light attenuation defined as  $-100 \ln(I/I_0^{-1})$ , where  $I_0$  and  $I$  are the light intensities before and after the filter, respectively, and  $k$  is a constant calculated from the concentration difference before and after the filter spot change. The algorithm assumes that the concentration stays stable during the filter spot change (Virkkula et al., 2007). This is not always true, so the median  $k$  values were calculated for different campaigns and were used to correct the data. For the aethalometer used during campaign 1,  $k$  was  $0.0050 \pm 0.0003$  (s.e) and during campaign 3 it was  $0.0060 \pm 0.0004$ . The  $k$  values were not possible to calculate for aethalometer used at the beginning of the campaign 2 due to the low measuring frequency (20 min). Same applies for the second aethalometer used during campaign 2, since  $k$  values for multiwavelength aethalometers may be incorrect. Therefore, the same  $k$  value, which was valid during campaign 1 and was also obtained by Virkkula et al. (2007), was used to correct the data during campaign 2. A sensitivity test of the  $k$  values showed that a difference of 0.001 would cause a 4 % and 3.9 % error in BC concentrations for the aethalometers used in 2000 and 2001, respectively.

An example about the loading effect correction is shown in Fig. 2 for two days of data in January 2005. The discontinuities in uncorrected data are due to the filter spot changes. At the end of each filter spot, the uncorrected concentrations are much

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smaller than concentrations at the beginning of the next filter. This effect is due to the loading effect of the filter. The correction algorithm makes the data more continuous and the decrease due to the loading effect disappears. It is evident that the corrections made are usually significant and can be as high as  $1 \mu\text{g m}^{-3}$ .

## 5 2.4 Meteorological pre-processing model

The meteorological data used in this study was calculated with the meteorological pre-processing model (MPP-FMI) (Karppinen et al., 1997, 2001), which is based on the energy budget method originally developed by van Ulden and Holtslag (1985). The method evaluates turbulent heat and momentum fluxes in the boundary layer from  
 10 synoptic weather observations. The MPP-FMI estimates the hourly time series for so-called pre-processed meteorological variables, which are more representative for the whole Helsinki area than variables from a single point measurements made, e.g., at the airport. The pre-processed meteorological variables include also important, not  
 15 directly measured variables such as Monin-Obukhov length and mixing height, which are important for determining the dispersion conditions.

## 2.5 Multiple regression analysis

A multiple regression analysis is used to define, which meteorological parameters influence the BC concentrations, and how much of the concentration variation can be explained by traffic and how much by the meteorological parameters.

20 In multiple regression analysis, a relationship between dependent variable and several independent variables is studied. In our case, BC concentration is the dependent variable and the traffic rate and meteorological parameters are the independent variables ( $X_1 \dots X_n$ ). The idea is to construct a model, which follows equation

$$\text{BC}_{\text{pred}} = b_0 + b_1X_1 + b_2X_2 + \dots + b_nX_n, \quad (2)$$

25 where  $b_0$  is the intercept and  $b_1 \dots b_n$  are the regression coefficients (Hair et al., 2006).

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Variables to the final (optimized) regression model are chosen by minimizing the difference between the measured BC concentrations and BC concentrations from the model. The normalization of the dependent and independent variables before the multiple regression model enables one to get also the so called beta coefficients, which tell the importance of the corresponding variable to the dependent variable in relation to the other variables in the model. We used bootstrapping to obtain error estimates to model parameters and performance indices. In bootstrapping, the used data is divided into 100 subsamples each including arbitrary 5/6 from the original data. The MLR model is constructed to each of the subsets and the final regression coefficients, beta coefficients, squared R and root mean square error are calculated as arithmetic mean and standard deviation of these 100 subsamples. The use of bootstrapping give better estimate for model parameters (regression and beta coefficients), eliminates the error caused by outliers and offers more generalized model as an output.

MLR model has an assumption that the variables are normally distributed. However, in our case most of the variables had a lognormal distribution and in order to avoid violating the assumptions of normal distribution, logarithmic transformations were done for the BC concentration, traffic rate, wind speed, mixing height and relative humidity. If one wishes to compare beta values of different sampling periods or weather conditions, the distributions have to be similar. This is ensured using Levene's test (Hair et al., 2006). The data used in all calculations had hourly time resolution.

### 3 Results and discussion

#### 3.1 Median BC concentrations

The BC time series (hourly medians) for the chosen periods were plotted in Fig. 3. High concentration episodes can easily be seen and it is evident that those episodes deviate from year to year depending, e.g., on meteorological conditions. The median BC concentrations experienced a slight decrease from 1.11 to 1.00  $\mu\text{g m}^{-3}$  between

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campaigns 1 and 3 (Table 2). The concentrations were lowest during campaign 2 with a median value of  $0.93 \mu\text{g m}^{-3}$ . The separate examination of different periods showed no clear systematic trend between the campaigns, but the highest concentrations were most often measured during campaign 1 (periods 1, 2 and 3). The highest median concentration  $1.43 \mu\text{g m}^{-3}$  was measured during period 1 of campaign 1 (Table 2), when no especially high peaks were observed (Fig. 2). This indicates, on average, higher daily concentrations and/or higher background concentrations. The lowest concentration  $0.68 \mu\text{g m}^{-3}$  was measured during period 2 of campaign 3.

The measured values correspond well with other measured BC concentrations in Helsinki. Pakkanen et al. (2000) used partly the same data as in campaign 1 and got an average value of  $1.38 \mu\text{g m}^{-3}$ . This value is somewhat higher than our  $1.11 \mu\text{g m}^{-3}$ , even though the used loading effect correction raised BC concentrations in this study. Deviations between these two studies rise from the partly different time periods and from the median value used in this study. Between July 2000 and July 2001 an annual average value of  $1.2 \mu\text{g m}^{-3}$  was measured by Viidanoja et al. (2002), being again somewhat higher than our result during campaign 2. The observed decrease  $0.11 \mu\text{g m}^{-3}$  during the ten years in Helsinki is much smaller than the yearly decrease of  $0.88 \mu\text{g m}^{-3}$  observed in Tokyo, Japan (Minoura et al., 2006). In Tokyo, the traffic rates are much higher and thus the effect of lower traffic emissions is more evident. In Helsinki, the concentration  $1.11 \mu\text{g m}^{-3}$  in 1996 and 1997 was already very small compared to Tokyo's  $11 \mu\text{g m}^{-3}$  for the year 1996. In general, concentrations are lower in Helsinki than in many European cities, where BC concentrations have been measured. Salma et al. (2004) measured a mean BC concentrations of  $2.9 \mu\text{g m}^{-3}$  at Budapest in spring 2002. In London, winter and spring BC concentrations were measured to be 3.2 and  $2.7 \mu\text{g m}^{-3}$ , respectively, in 1995 and 1996 (Kendall et al., 2001). Much higher BC concentrations were measured in Birmingham by Castro et al. (1999) in winter 1994 ( $3.4 \mu\text{g m}^{-3}$ ) and in Vienna by Hittenberger and Tohno (2001) during winter 1998–1999 ( $6.58 \mu\text{g m}^{-3}$ ).

In Helsinki, the BC concentrations were similar in winter and spring (not shown).

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However, the concentrations had a weekly cycle with higher concentrations on weekdays and lower on weekends (Fig. 4). This is apparently related to the lower traffic rates on weekends. On weekdays, the daily median BC concentrations were between 1.3 and 1.5  $\mu\text{g m}^{-3}$  during campaign 1, between 0.9 and 1.1  $\mu\text{g m}^{-3}$  during campaign 2 and between 1.1 and 1.3  $\mu\text{g m}^{-3}$  during campaign 3. Thus, the weekday concentrations were again lowest during campaign 2, and decreased from campaigns 1 to 3 following the overall medians. On weekends, the concentrations were between 0.6–1.1  $\mu\text{g m}^{-3}$  with slightly lower values on Sundays. During ten years, the weekend concentrations systematically decreased.

The diurnal cycle of BC concentrations was examined separately for weekdays and weekends (Fig. 5). The effect of traffic was evident on weekdays with maximum concentration during morning rush hours between 5 and 9 a.m. (Fig. 6a). The peaks related to morning rush hours were the same 2.4  $\mu\text{g m}^{-3}$  during campaigns 1 and 3, and lower (1.7  $\mu\text{g m}^{-3}$ ) during campaign 2. The afternoon maximum, related to afternoon rush hours, was most evident during campaign 1 reaching a value of 2  $\mu\text{g m}^{-3}$ . The afternoon peaks were systematically lower than the morning peaks due to the stronger turbulent mixing and increased mixing layer heights. The daytime BC concentrations were evidently higher during campaign 1 suggesting that traffic had a greater effect to the BC concentrations ten years ago. The daytime concentrations were lowest during campaign 2 following the total concentrations. The night-time concentrations were similar, around 0.5  $\mu\text{g m}^{-3}$ , during the campaigns.

On weekends, the diurnal cycle differed considerably from that on weekdays. Minimum values were measured in the early morning and maximum values at night and afternoon. Friday and Saturday are the most typical days for people to go out in Helsinki, and the raised concentrations at night time are caused by the numerous taxis and buses, which are typically diesel powered (see also Fig. 6b). In addition to the intense diesel-powered traffic, low mixing heights elevate the night time concentrations. The diurnal cycle of BC was similar to the diurnal behaviour of accumulation mode particle concentrations measured in Helsinki (Laakso et al., 2003; Hussein et al., 2004). This

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was the case especially during weekends, when diurnal variations were rather small and maximum was measured at night.

### 3.2 Traffic trends

The major source of BC is the local traffic, which in our case refers mainly to the vehicles on the road (Hämeentie) next to measurement site. To estimate the effect of traffic on the measured concentrations, it is important to know the traffic intensity on the Hämeentie road. Online traffic monitoring was only made at Itäväylä road, so the traffic rates on Hämeentie road needed to be derived from these. The traffic rates have been counted in campaigns at Hämeentie, and the comparison of these campaign traffic rates with online traffic measurement at Itäväylä, enabled us to calculate transformation coefficients between the roads. The coefficients were determined by fitting a straight line to the data measured at these two locations. To get the traffic rates on Hämeentie, the traffic data from Itäväylä needed to be multiplied by values 0.576 ( $R^2=0.998$ ), 0.553 ( $R^2=0.995$ ) and 0.564 ( $R^2=0.997$ ) for campaign 1, 2 and 3, respectively. This shows that the traffic rates on these two roads are correlating well, but are about 50% less on Hämeentie than on Itäväylä.

Overall, the daily traffic rates have increased from 29 700 to 30 000 vehicles day<sup>-1</sup> on Hämeentie between campaigns 1 and 3. This increase occurred mainly on weekdays, while on weekend, the traffic rates were similar (19 750 vehicles day<sup>-1</sup>) during campaigns 1 and 3. The traffic rates were lowest (29 600 vehicles day<sup>-1</sup>) during campaign 2, agreeing with the lowest BC concentrations during campaign 2. However, the changes in traffic rates were marginal and this correlation should be considered cautiously.

On weekdays (Fig. 6a), the traffic rate followed a typical rush-hour related pattern with maxima in the morning (5–0 a.m.) and afternoon (3–6 p.m.). The traffic pattern on weekends (Fig. 6b) showed highest traffic rates at daytime during all campaigns with slightly higher traffic intensities during campaign 1. The weekend night-time traffic rates seemed to have decreased from 1996 to this decade. The differences in the

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diurnal cycle of traffic rates between the campaigns were small compared to the diurnal behaviour of BC concentrations (Fig. 6) implying that the number of vehicles is not the only factor affecting the concentrations. On weekends, the diurnal behaviour of BC concentrations was not as evidently related to the diurnal cycle of traffic rates as on weekdays.

As already was mentioned, the traffic rates itself did not explain all variations in the BC concentrations. The measured BC concentrations are also affected by the other local and distant sources. The effect of meteorology on the measured concentrations is studied in Sect. 3.3. Some of the differences between the BC concentrations and traffic can be explained by the uncertainties in traffic counts, which do not take into consideration whether vehicles are diesel or gasoline powered, and differences in vehicle emissions. The effect of cleaner fuels and improved technology in diesel powered vehicles could be examined by comparing the night-time BC concentrations and traffic rates on weekends between the campaigns, when vehicles are mainly expected to be diesel-powered taxis and buses. The taxi pool is always fairly new in Helsinki and the effect of improved technology is easier to detect during intensive taxi traffic. Typically, the average age of the car pool is high in Finland. Passenger cars mean age was 10.4 years in Finland in 2003, while in UK and France corresponding ages were 6.8 and 8.0 years (<http://www.autoalantieto.fi/vanhauusi.asp>), respectively. A very rough estimate for diesel vehicle emissions was obtained by dividing the hourly BC concentrations with hourly traffic rates between 2 and 4 a.m. We found that the vehicle number-scaled BC mass concentrations were  $0.0028 \pm 0.0005$  (s.e),  $0.0022 \pm 0.0005$  and  $0.0020 \pm 0.0002 \mu\text{g m}^{-3}$  for campaign 1, 2 and 3, respectively. Thus, the diesel vehicle emissions seem to have decreased, following the engine development and better exhaust after treatment. These estimates have a large uncertainty and should be considered with caution, since night time traffic includes also some gasoline cars and BC concentrations include an contribution of other local sources and long-range transport.

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### 3.3 Multiple regression analysis

Besides traffic, meteorology has a strong effect on the measured BC concentrations. The local meteorology affects the dispersion of pollutants and long-range transport has been identified to be one of the main factors affecting BC concentrations, with an average value of  $0.4 \mu\text{g m}^{-3}$  (Pakkanen et al., 2000). A multiple regression analysis was made to identify the relationships between BC concentrations, traffic and meteorological variables.

Multiple regression models were constructed for different situations, including whole data, winter and spring separately, weekdays and weekends separately, and for each campaign. The optimized models were in most cases obtained with three independent variables. Traffic and wind speed were always present in the models and the third variable varied between mixing height, pressure, temperature and relative humidity. The median values of these affecting variables during every period are shown in Fig. 7. The change of the third variable in the multiple regression models did not have a great effect and in the final optimized models the same three variables were used to get the different cases more comparable. Traffic and wind speed were automatically included in the optimized multiple regression models and the third one was chosen to be the mixing height. These three variables gave most often the best models.

The multiple regression analysis for all data ( $R^2=54\%$ ) confirmed traffic having the most important effect on BC concentrations with a beta coefficient of 0.63 (Table 3). Of the meteorological variables, wind speed had the highest influence on the BC concentrations with a beta coefficient of -0.48. The negative sign means inverse relationship between the wind speed and BC concentrations: the lower the wind speed, the higher the concentration due to the mixing of pollutants. The beta coefficient for mixing height was also negative (-0.17). High mixing height allows the air pollution mix into a larger air volume and lower concentrations at the surface are measured. Division into weekdays and weekends showed traffic having the greatest effect on BC concentration changes on weekdays but not on weekends, when the wind speed had the strongest

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influence (Table 3). The latter, was already suggested by the diurnal cycle of BC and traffic rate, which seemed to have a low connection on weekends (Figs. 5 and 6). The multiple regression model was poorer for weekend than for weekdays. Adding more variables did not improve the model on weekends, suggesting that there was some missing variable or variables, which were not included to the available variables such as the long-range transport. On the other hand, also the number of weekend samples was much lower than the number of workday samples, which increases the uncertainty.

A multiple regression analysis was also made separately for winter and spring to get more information about the seasonal changes (not shown). The optimized model was better for winter ( $R^2=62\%$ ) than for spring ( $R^2=52\%$ ) and differences were small between the seasons. The effect of traffic was slightly larger during winter than spring (beta coefficients of 0.66 and 0.62, respectively), while the influence of wind speed was the same. The mixing height was more important factor during spring than in winter.

To compare whether the contribution of traffic on the measured BC concentrations had changed during the ten years, multiple regression models were made separately for different campaigns (Table 3). The Levene's test showed that with 95% confidence the variances of traffic were the same between the campaigns, meaning comparable beta coefficients between the campaigns. The squared R's were between 53 and 60 % during the campaigns. In all cases, traffic had the highest influence on the BC concentrations before the wind speed. The effect of wind speed and mixing height did not have any systematic trend between the campaigns and for both, the lowest effects were observed during campaign 2. The effect of traffic, on the other hand, had a decreasing trend between the beta coefficients from 0.69 to 0.62 indicating a decreased influence of traffic to the BC concentration changes. The number of vehicles had increased in Helsinki and thus, the decreasing effect needs to be related to the cleaner emissions from vehicles. This supports the rough estimate for the diesel vehicle emissions obtained in Sect. 3.2.

The decreasing BC concentration trend between the campaigns 1 and 3 could be explained by the decreased emissions from the (diesel) vehicles. The decrease in

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concentrations occurred especially during rush hours, when the traffic rates are highest and the decreased vehicle emissions most visible. The traffic rate was the most important variable affecting the BC concentrations during campaign 2, so the BC concentration minimum was at least partly caused by the lower traffic rates, even though the deviations between the campaigns were small.

#### 4 Conclusions

The purpose of this study was to investigate BC concentration trends in Helsinki between 1996 and 2005. Measurements were made during three campaigns in Vallila, which is located two kilometres from the centre of Helsinki. One of the main roads leading to the city centre is located 10 m away from the measurements site. The first campaign took place from November 1996 to June 1997, the second from September 2000 to May 2001 and the third from March 2004 to October 2005. The studied dates were chosen to be the same for all three campaigns, with four measurement periods and a total number of 82 days from every campaign. Two periods were at winter and two at spring, respectively.

The BC concentrations showed a slightly decreasing trend from 1.11 to 1.00  $\mu\text{g m}^{-3}$  during ten years. The decrease occurred especially on daytime during the weekdays, when traffic intensities were highest. Systematically, lower concentrations were measured during campaign 2 following the slightly lower traffic rates. The only exception was the weekend concentrations when the concentrations decreased between the campaigns.

The optimized multiple regression models to predict the BC concentrations were obtained with three variables: traffic, wind speed and mixing height. On weekdays, traffic had clearly the highest influence before the wind speed, and on weekends, the effect was the other way around. On weekends, the traffic rates were much lower causing the meteorological factors becoming more important. Of the three variables, the mixing height was always the least explanatory one. The separate examination of all three

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campaigns by means of a multiple regression model, showed a decreased influence of traffic on the BC concentrations. The beta coefficients decreased from 0.69 to 0.62. Since the traffic rates have increased between campaign 1 and 3, the decreasing effect is very likely caused by cleaner emissions from vehicles due to engine and fuel development, and exhaust after treatment. A rough estimate for BC emissions from diesel vehicles was calculated on weekend nights, when vehicles are mainly diesel powered buses and taxis. At our sampling site, the vehicle number-scaled BC concentrations have decreased from 0.0028 to 0.0020  $\mu\text{g m}^{-3}$  between campaigns 1 and 3. It is worth of mentioning, that these values are very rough and may be different for different sites. The decreased BC emissions from traffic most probably caused the decrease of the BC concentrations during the ten years, while the lowest concentrations during campaign 2 were at least partly influenced by lower traffic rates.

*Acknowledgements.* For financial support we would like to thank Research Foundation of the University of Helsinki, Maj and Tor Nessling Foundation, National Technology Agency (TEKES, Grant # 40462/03) and the Ministry of Environment. T. Koistinen and H. Seppälä from City Planning Department and M. Dal Maso and L. Sogacheva from University of Helsinki are acknowledged for their help in this study.

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**Table 1.** The chosen data periods, the number of measurement days and the percentage of data included on every period.

| Time period     | Nro of days | Campaign 1<br>1996–1997<br>(%) | Campaign 2<br>2000–2001<br>(%) | Campaign 3<br>2004–2005<br>(%) |
|-----------------|-------------|--------------------------------|--------------------------------|--------------------------------|
| 1.12–17.12 (P1) | 17          | 88                             | 100                            | 76                             |
| 26.12–8.1 (P2)  | 14          | 86                             | 71                             | 79                             |
| 6.3–3.4 (P3)    | 29          | 79                             | 97                             | 100                            |
| 13.4–4.5 (P4)   | 22          | 100                            | 55                             | 100                            |

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**Table 2.** The median black carbon concentrations ( $\mu\text{g m}^{-3}$ ) and quartile deviations (half of the difference between lower and upper quartiles) measured during different campaigns and periods.

| Period | Campaign 1<br>(1996–1997) | Campaign 2<br>(2000–2001) | Campaign 3<br>(2004–2005) |
|--------|---------------------------|---------------------------|---------------------------|
| P1     | 1.43 (0.62)               | 0.95 (0.35)               | 1.12 (0.71)               |
| P2     | 1.14 (0.54)               | 0.97 (0.36)               | 0.68 (0.46)               |
| P3     | 1.11 (0.61)               | 0.92 (0.40)               | 1.04 (0.56)               |
| P4     | 0.86 (0.54)               | 0.90 (0.51)               | 1.05 (0.50)               |
| All    | 1.11 (0.60)               | 0.93 (0.40)               | 1.00 (0.56)               |

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**Table 3.** The results from multiple regression analysis for all data, weekdays and weekends separately, and for campaigns 1, 2 and 3 ( $p < 0.05$ ). Optimized models were obtained with three variables: traffic (Tr), wind speed (U) and mixing height ( $H_m$ ). The model parameters squared R, root mean square error (rmse), intercept (int), and regression coefficients  $b$  and beta coefficients  $\beta$  for used variables are also listed.

|            | $R^2$ | rmse $\pm$ std  | int             |       | $b$   | $\beta$ | std ( $\beta$ ) |
|------------|-------|-----------------|-----------------|-------|-------|---------|-----------------|
| All        | 54    | 0.22 $\pm$ 0.00 | 0.91 $\pm$ 0.01 | Tr    | 0.50  | 0.63    | 0.01            |
|            |       |                 |                 | U     | −0.73 | −0.48   | 0.01            |
|            |       |                 |                 | $H_m$ | −0.17 | −0.17   | 0.01            |
| Weekdays   | 59    | 0.21 $\pm$ 0.00 | 0.83 $\pm$ 0.01 | Tr    | 0.50  | 0.69    | 0.01            |
|            |       |                 |                 | U     | −0.70 | −0.46   | 0.01            |
|            |       |                 |                 | $H_m$ | −0.12 | −0.12   | 0.01            |
| Weekends   | 48    | 0.20 $\pm$ 0.00 | 0.97 $\pm$ 0.02 | Tr    | 0.36  | 0.39    | 0.01            |
|            |       |                 |                 | U     | −0.81 | −0.60   | 0.01            |
|            |       |                 |                 | $H_m$ | −0.22 | −0.28   | 0.01            |
| Campaign 1 | 58    | 0.21 $\pm$ 0.00 | 1.06 $\pm$ 0.02 | Tr    | 0.54  | 0.69    | 0.01            |
|            |       |                 |                 | U     | −0.71 | −0.47   | 0.01            |
|            |       |                 |                 | $H_m$ | −0.21 | −0.22   | 0.01            |
| Campaign 2 | 53    | 0.18 $\pm$ 0.00 | 0.68 $\pm$ 0.02 | Tr    | 0.42  | 0.65    | 0.01            |
|            |       |                 |                 | U     | −0.56 | −0.37   | 0.01            |
|            |       |                 |                 | $H_m$ | −0.13 | −0.14   | 0.01            |
| Campaign 3 | 60    | 0.23 $\pm$ 0.00 | 1.07 $\pm$ 0.03 | Tr    | 0.55  | 0.62    | 0.01            |
|            |       |                 |                 | U     | −0.90 | −0.57   | 0.01            |
|            |       |                 |                 | $H_m$ | −0.21 | −0.22   | 0.01            |

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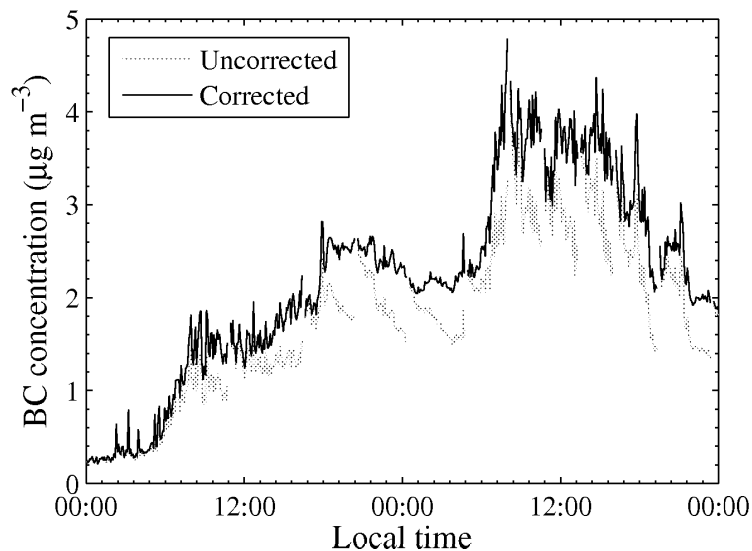


**Fig. 1.** The measuring locations on Helsinki area. The black circle presents the Vallila site and the black square shows the place of the automatic traffic counts at Itäväylä road.

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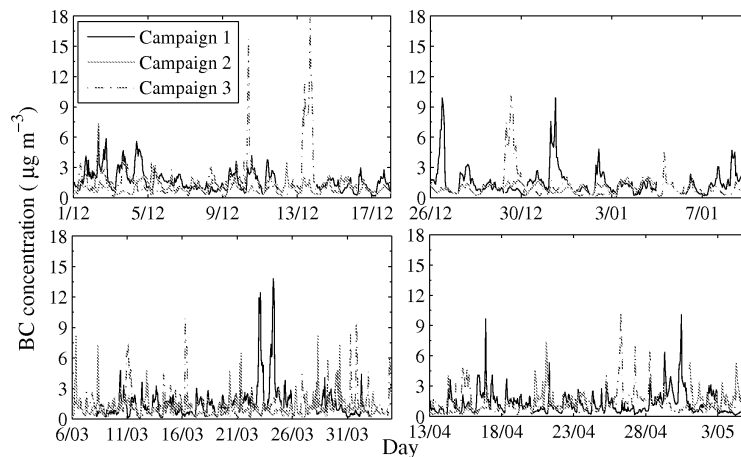


**Fig. 2.** The black carbon mass concentrations ( $\mu\text{g m}^{-3}$ ) before (grey line) and after (black line) the loading effect correction on 17–18 Jan 2005.

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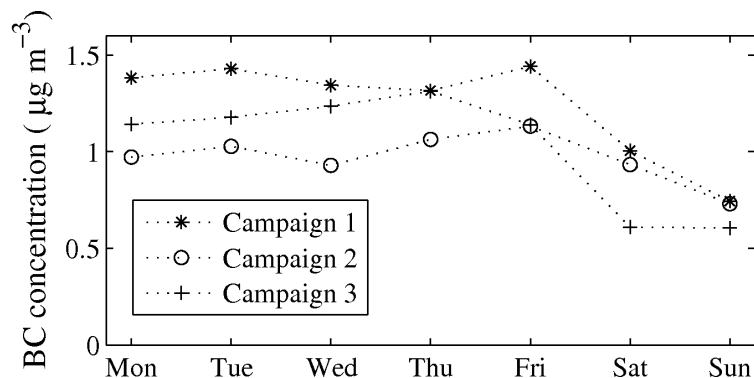
**Fig. 3.** Black carbon time series during chosen periods. Black solid line stands for campaign 1, light-gray solid line for campaign 2 and dark-gray dash-dotted line for campaign 3. Values are hourly medians calculated from the raw data.

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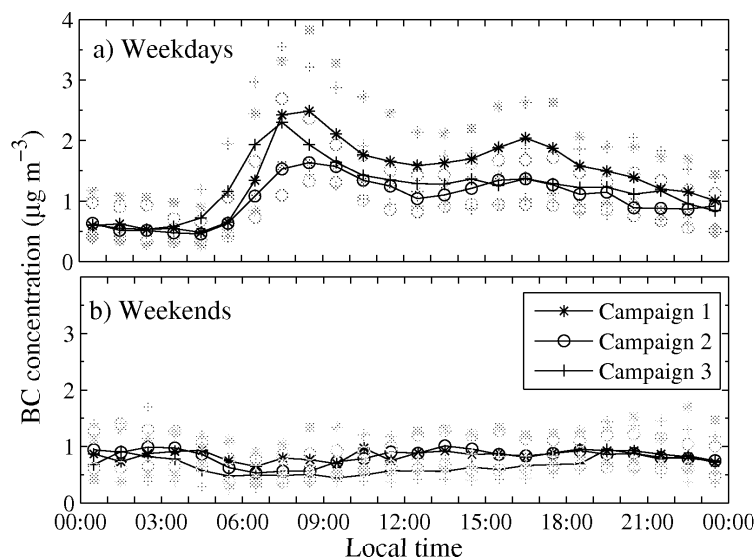
**Fig. 4.** Median black carbon concentrations on different weekdays during campaigns 1 (stars), 2 (circles) and 3 (crosses).

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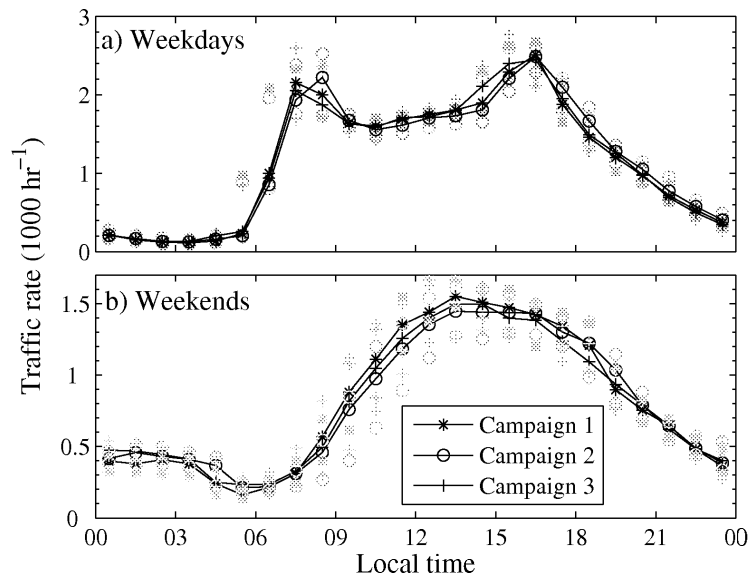


**Fig. 5.** Diurnal median BC concentrations (black lines) together with lower and upper quartiles (gray marks) on **(a)** weekdays and **(b)** weekends. The stars present the BC concentrations during the four periods during campaign 1, circles are the BC concentrations during campaign 2 and crosses represent the BC concentrations during campaign 3.

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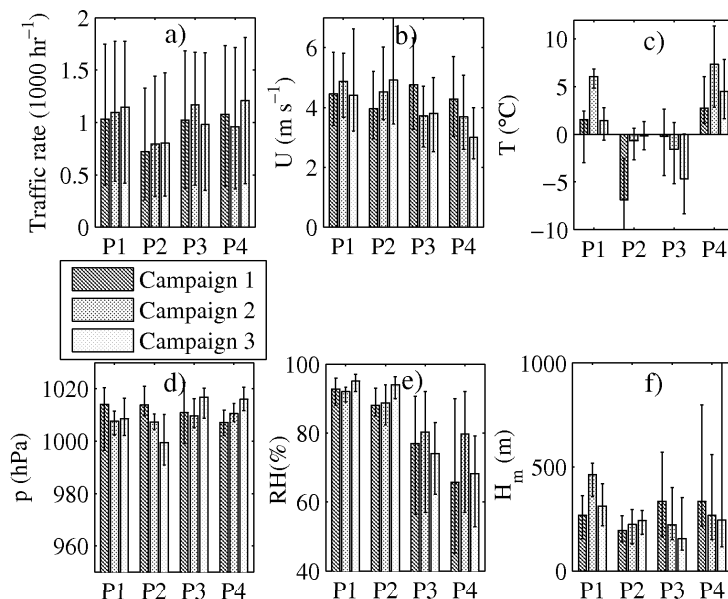


**Fig. 6.** Diurnal median traffic patterns (Black lines) together with lower and upper quartiles (gray marks) on **(a)** weekdays and **(b)** weekends. The stars present the traffic rates during the four periods during campaign 1, circles are the traffic rate during campaign 2 and crosses represent the traffic rate during campaign 3.

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**Fig. 7.** Median (a) traffic rate (1000 vehicles  $\text{hr}^{-1}$ ), (b) wind speed  $U$  ( $\text{m s}^{-1}$ ), (c) temperature  $T$  ( $^{\circ}\text{C}$ ) (d) pressure  $p$  (hPa), (e) relative humidity  $\text{RH}$  (%) and mixing height  $H_m$  (m) for all four periods during campaigns 1, 2 and 3.

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