



## Contribution of the road traffic to air pollution in the Prague city (busy speedway and suburban crossroads)

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### ABSTRACT

Two measurement campaigns near busy freeway and suburban crossroad (different traffic intensity) were carried out in Prague city. Both of the two traffic related campaigns were supported with simultaneous measurements at suburban background site to compare measured values with corresponding city background. Extensive set of aerosol instrumentation was used during both campaigns at both measurement sites. Two aerosol spectrometer sets (SMPS and APS) were used to monitor aerosol particle number size distribution. Two BLP1 provided information about mass size distribution and chemical composition of collected size resolved aerosol samples. The crossroad campaign was also supported by two EC/OC field analysers providing information about elemental and organic carbon concentrations. Consequent chemical analysis of collected size resolved samples comprised of ion chromatography (water soluble ions), PIXE (elements) and RBS (carbon). Results obtained from spectrometers and BLP1s showed that the main contribution of traffic in ultrafine size range can be attributed to direct exhaust emissions, while the coarse fraction was dominated mainly by regional background aerosol with small traces of brake and tyre abrasion as well as the resuspension of the road dust. Chemical analysis demonstrated that most of the water soluble ions can be found in fine fraction of mass distribution and mostly comes from regional background and long range transport. Most of major elements were found in coarse fraction of mass size distribution and it can be attributed to three different sources: abrasion of different vehicle parts (Fe, Cu, Mn and Zn), resuspension of the road dust (Si, Al, Ca) and long range transport or regional background (Ca and K). Elemental carbon concentration (diesel engine emissions) was found to be proportional to traffic intensity.

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### 1. Introduction

Aerosol particles belong among major air pollution sources. Moreover, it is generally known that aerosol particles have chronic and acute adverse health effects (Pope et al., 1991, 1995; Schwartz et al., 1993; Oberdörster et al., 1995; Pope, 2000; Kampa and Castanas, 2008), they damage cultural heritage (Zappia, 2000), reduce visibility (Chan et al., 1999) and cause a climate change (Jacobson, 2002).

A special type of environment is the urban area (Mönkkönen et al., 2004). Aerosol particles emitted from the road traffic significantly contribute to high levels of air pollution. Today, these particles represent one of the main sources of air pollution in the urban environment.

The particles emitted from road traffic can be divided into two categories – direct and indirect emissions. Direct emissions are emissions from vehicle exhausts (Mulawa et al., 1997; Sagebiel et al., 1997), abrasion of car breaks, tyres and pavement (Rogge et al., 1993; Moosmüller et al., 1998; Garg et al., 2000; Kupianen et al., 2005) and particle resuspension (Nicholson et al., 1989; Sternbeck et al., 2002). The indirect emissions include secondary aerosol particles originated by chemical reactions from the gaseous emissions. Apportionment of road traffic particles according to their size can be made taking into account the process of particle formation. Nanoparticles (<50 nm) and ultrafine particles (<100 nm) are produced by high temperature and chemical processes and thus can be linked to exhaust emissions and secondary aerosol particles. The coarse particles (>1 µm) are usually formed by mechanical processes, which include the abrasion and the resuspension mechanisms.

The exhaust emissions depend on the type and age of the engine, the type of used fuel and oil, the gearshift mode (idling, gear) as well

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as on the configuration of the exhaust system (Rönkkö et al., 2006; Morawska et al., 1998). The measurements show that the emissions from diesel engines are four orders of magnitude higher than the emissions from gasoline engines, namely  $10^8$  particles  $\text{cm}^{-3}$  and  $10^4$  particles  $\text{cm}^{-3}$ , respectively. The size of particles produced by gasoline engines usually vary between 40 and 80 nm, while the diesel engines emit the particles in the range 50–120 nm (Morawska et al., 1998; Ristovski et al., 1998; Harris and Maricq, 2001). The emissions from modern diesel engines can go even lower in particle size, down to units of nanometres, namely 2–4 nm (Beatrice et al., 2010). The smaller particle size of aerosol emitted from gasoline engines can be caused by lower coagulation growth due to the lower total concentrations  $\sim 10^4$  particles  $\text{cm}^{-3}$  (Harris and Maricq, 2001). Another measurements of gasoline engine emissions show bimodal size distributions having nucleation mode  $<30$  nm and the accumulation mode about 60 nm (Giechaskiel et al., 2005; Kittelson et al., 2006a, b; Rönkkö et al., 2006; Weimer et al., 2009).

The abrasion of brakes produces particles characterized by high concentrations of Cu, Ba, Zn and Fe (Sanders et al., 2003; Hjortenkrans et al., 2007). In the study made in Sweden, the authors found out that 90% of Cu traffic emissions come from the brake abrasion (Johansson et al., 2008). The other typical “brake” elements are Sb, K and Ti. Kukutschova et al. (2011) studied emissions from low metallic breaks using BLPI and PIXE analysis and they found most of particles in the coarse mode, where Fe formed 20% of mass and Cu, Sn, Zn, and S formed about 2.6%, 1.7%, 1.7%, and 0.9% of coarse mode mass. Other chemical compounds are added to some brake types as lubricants (e.g.  $\text{Sb}_2\text{S}_3$ ) to prevent excessive wear by heavy brake duty (Jang and Kim, 2000),  $\text{K}_2\text{O} \cdot n\text{TiO}_2$  is sometimes used to improve the thermal resistance and to lower the brakes wear (Hee and Filip, 2005). Elevated concentrations of these elements were observed in numerous studies dealing with the emissions from road traffic (Legret and Pagotto, 1999; Sternbeck et al., 2002; Sanders et al., 2003; Wählin et al., 2006; Canepari et al., 2008; Johansson et al., 2008; Handler et al., 2008). Brake abrasion produces mainly coarse aerosol particles (Abu-Allaban et al., 2003; Wählin et al., 2006; Sanders et al., 2003; Iijima et al., 2007; Garg et al., 2000).

Particles released by mechanical abrasion of tyres are characterized especially by high concentration of Zn. ZnO is used to accelerate the process of vulcanization during the tyre production (Sörme et al., 2001). The elevated concentrations of Zn from road emission measurements have been documented in several studies (Lagerwerff and Specht, 1970; Hopke et al., 1980; Legret and Pagotto, 1999; Wählin et al., 2006; Johansson et al., 2008; Handler et al., 2008). The high Zn concentrations in aerosol samples from road traffic are also partially explained by the presence of this element in the engine oils (Sörme et al., 2001). Johansson et al. (2008) found out, based on measurements and modelling, that about 40% of Zn emissions from road traffic come from exhaust gases, engine oils, fuel burning and the engine wear. Hjortenkrans et al. (2007) proved that the predominant source of Zn particles in the urban environment is the tyre abrasion. Furthermore, the emissions from tyre abrasion were found to be a dominant source of Cd probably as a contaminant of ZnO. Moreover, the abrasion of tyres can contribute to the total “traffic” mass also with particles containing high concentrations of Si, Ti, Cr, Ni, Cu, Sb and Pb (Sörme et al., 2001; Hjortenkrans et al., 2007; Canepari et al., 2008).

Substantial source of aerosol particles from the road traffic is also the mechanical abrasion of the pavement. These particles typically contain high concentrations of Si, Al, Ca, Mg and Fe. The elevated concentrations of above mentioned elements in the coarse mode of road traffic emissions were observed in many studies (Sternbeck et al., 2002; Abu-Allaban et al., 2003; Wählin et al., 2006; Canepari et al., 2008; Handler et al., 2008). Further, several studies drew the attention to the influence of the pavement surface on particle

emissions. The particles emitted from the asphalt pavement are characterized mainly by high concentrations of Zn, Cr, Ni and Pb (Sörme et al., 2001). In Italy, Canepari et al. (2008) showed that for asphalt pavement the typical elements can be also As, Ti and V. Furthermore, the granite pavement releases about 70% higher emissions of PM10 than quartz pavement (Gustafsson et al., 2008).

Another important source of aerosol particles from the road traffic is the dust resuspended from the pavement surface. These particles comprise mainly the salt and crushed stones, deposited during winter road maintenance. Such particles usually contain high concentrations of Cl. In Copenhagen, it was proved that the Cl concentrations correlate with the road dust concentrations during winter, while during summer the concentration of Cl was almost zero (Wählin et al., 2006). In winter, Legret and Pagotto (1999) observed elevated concentrations of Pb, Cd, Zn, and attributed them to corrosion of surfaces caused by de-icing salts. Measurement of particles in two tunnels in Sweden was carried out at the end of winter. The elevated concentrations of Si, Al, Ca and Fe particles were observed only in the tunnel, which was not cleaned. It confirms the origin of these particles as a resuspended road dust. On the other hand, aerosol sampled in both tunnels was enriched to the same levels with Ba, Cd, Cu, Pb, Sb and Zn. This observation proves the origin of these particles in car emissions (Sternbeck et al., 2002). In Vienna, a strong correlation among concentrations of coarse mode Ca, Mg, Fe, Si and Al particles was observed, pointing to the identical source of these particles (Handler et al., 2008). The emissions due to resuspension of particles are also highly influenced by the car speed. Gustafsson et al. (2008) found that the increase of a car speed by  $10 \text{ km h}^{-1}$  causes the increase of PM10 by  $680 \mu\text{g m}^{-3}$  in one of the simulator experiments. Decreasing the speed limit from 100 down to  $80 \text{ km h}^{-1}$  on the Amsterdam city circuit led to a provable decrease of PM10 and PM1 concentrations (Dijkema et al., 2008).

Measurements of ultrafine particles from the road traffic, performed in the traffic flow directly on the roads or nearby the roads, show that the measured concentrations depend primarily on the distance from the road, site position regarding the wind direction and also on temperature and relative humidity. The concentration of aerosol particles decreases with an increasing distance from the road, mainly due to the particle dispersion and coagulation. In case of long distances of measurement site from the road or, when the wind blows in the opposite direction, the measured concentrations usually correspond to background concentrations (Hitchins et al., 2000; Zhu et al., 2002a, 2004; Buonanno et al., 2008; Mejía et al., 2008). In case of decreasing temperature, concentration of the nucleation mode particles rises, most probably due to an increased condensation of organic vapours (Bukowiecki et al., 2002; Zhu et al., 2002a,b, 2004; Giechaskiel et al., 2005; Pirjola et al., 2006; Yao et al., 2007).

The aim of our work was to compare the contribution of road traffic emissions to air pollution in places with different traffic density and, at the same time, to identify the individual sources of “traffic” aerosol according to their size resolved chemical composition.

## 2. Experimental setup

Two road traffic measurement campaigns were conducted during our study. The first campaign was performed nearby a busy freeway in Prague (see Fig. 1). The freeway is a part of Prague's circumferential highway. The other campaign was conducted next to the crossroads in the suburban area of Prague (see Fig. 1). During both campaigns, simultaneous measurement of the same parameters was carried out in the campus of the ICPF (Institute of Chemical Process Fundamentals, AS CR v.v.i.), in Prague (see Fig. 1). This measurement site is considered to be a suburban background site.

Both campaigns consisted of measurements of aerosol particle number size distribution, mass size distribution (including

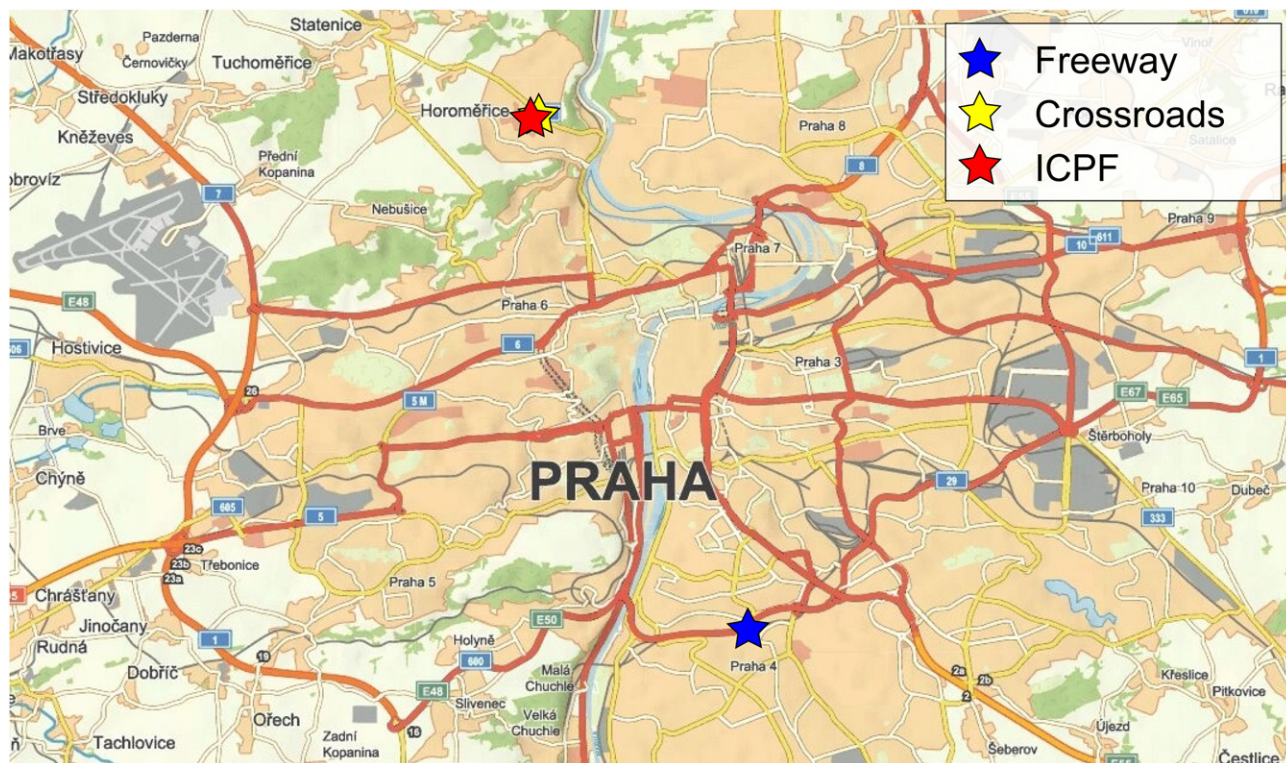


Fig. 1. Measurement sites.

a following chemical analysis) and meteorological conditions. In addition, the crossroads measurement campaign included also the measurement of total number concentration with high time resolution, PM<sub>10</sub> and PM<sub>2.5</sub> total mass concentration and EC/OC concentrations. The number of cars passing (in both directions) next to the measurement site was monitored in both cases (freeway and crossroads) as well.

The freeway measurement campaign was conducted during 3-day period in September 2008 (9th–11th). The selected measurement site is located on a very busy segment of a circumferential Prague's circuit (5900 cars per hour on average during rush hours) with 3 lanes in each direction (see Fig. 2). The instruments were placed in a mobile container located about 2 m from the closest freeway lane. The sampling inlets were located approximately 4–5 m above the ground. The instrumentation included two 10 stage BLPIs (Bernier Low Pressure Impactor, Hauke), one SMPS 3934 (Scanning Mobility Particle Sizer, TSI) and one APS 3321 (Aerodynamic Particle Sizer, TSI). The BLPI covers the range of aerodynamic diameters from 25 nm up to 10 µm. BLPI1 was provided with polycarbonate foils covered with Apiezon L to prevent particle bounce, BLPI2 was loaded with aluminium foils. The sampling with BLPIs was carried out for 23 h. The two spectrometers covered the particle size range from 13.8 nm up to 10 µm (PM<sub>10</sub> sampling head was used as a common inlet of both spectrometers). The scanning interval of both spectrometers was set to 5 min (SMPS up scan and APS scan 3 min). At the same time, the PM<sub>10</sub>, gases (SO<sub>2</sub>, NO, NO<sub>2</sub>, NO<sub>x</sub>, CO and O<sub>3</sub>), the noise level and meteorological conditions (wind direction and speed, temperature, relative humidity and atmospheric pressure) were measured.

The crossroads measurement campaign was carried out during the 3-day period in May 2009 (12th–15th). The crossroads is located in Suchbátka, a north-western suburban area of Prague (see Fig. 3). Four pedestrian crossings traverse the crossroads and three

bus stops are located in the vicinity of the crossroads. It means that cars stop, start and change their direction here quite often. During the examined period, the average traffic density was found to be close to 1000 cars per hour. The mobile measurement container was placed on the sidewalk about 3 m apart from the crossroads, just opposite to one of the bus stops. The sampling inlets were placed at the same height as during the freeway measurement (4–5 m above the ground). In terms of on-line measurements an SMPS 3934, APS 3321 and P-Trak (TSI) were deployed inside the mobile container. The on-line instruments measured in the range from 14.1 nm up to 10 µm (PM<sub>10</sub> sampling head was used as a common inlet of both spectrometers). The container included also a BLPI (Hauke) with polycarbonate foils greased with Apiezon L and 2 LVS (Low Volume Samplers, Leckel) loaded with quartz fibre filters for PM<sub>10</sub> and PM<sub>2.5</sub> measurements. The filters in the LVS were arranged in a QBQ (Quartz Behind Quartz) setup in order to correct the results for positive OC artifacts. The off-line sampling was performed for 15-h period during the day time. Furthermore, a field EC/OC analyzer (Carbon Aerosol Analysis Field Instrument, Sunset Lab.) provided with a PM<sub>2.5</sub> sampling head was placed in the container. The EC/OC analyzer sampled with the interval of 2 h.

The campus of ICPF is located in the same area as the above mentioned crossroads (see Fig. 3). There are no major sources of air pollution close to the campus of ICPF except the family houses (with the local heating) and the closest road is about 260 m from the measurement station. The freeway measurement site was located in a distance of about 11.1 km, while the crossroad measurement container was about 0.5 km apart from the ICPF monitoring station. The continuous monitoring station is equipped with SMPS 3034 (TSI) and APS 3321 (TSI). These spectrometers measured in following size ranges: SMPS 3034 10–470 nm, SMPS 3936 10–430 nm and APS 0.523–10 µm (having again common

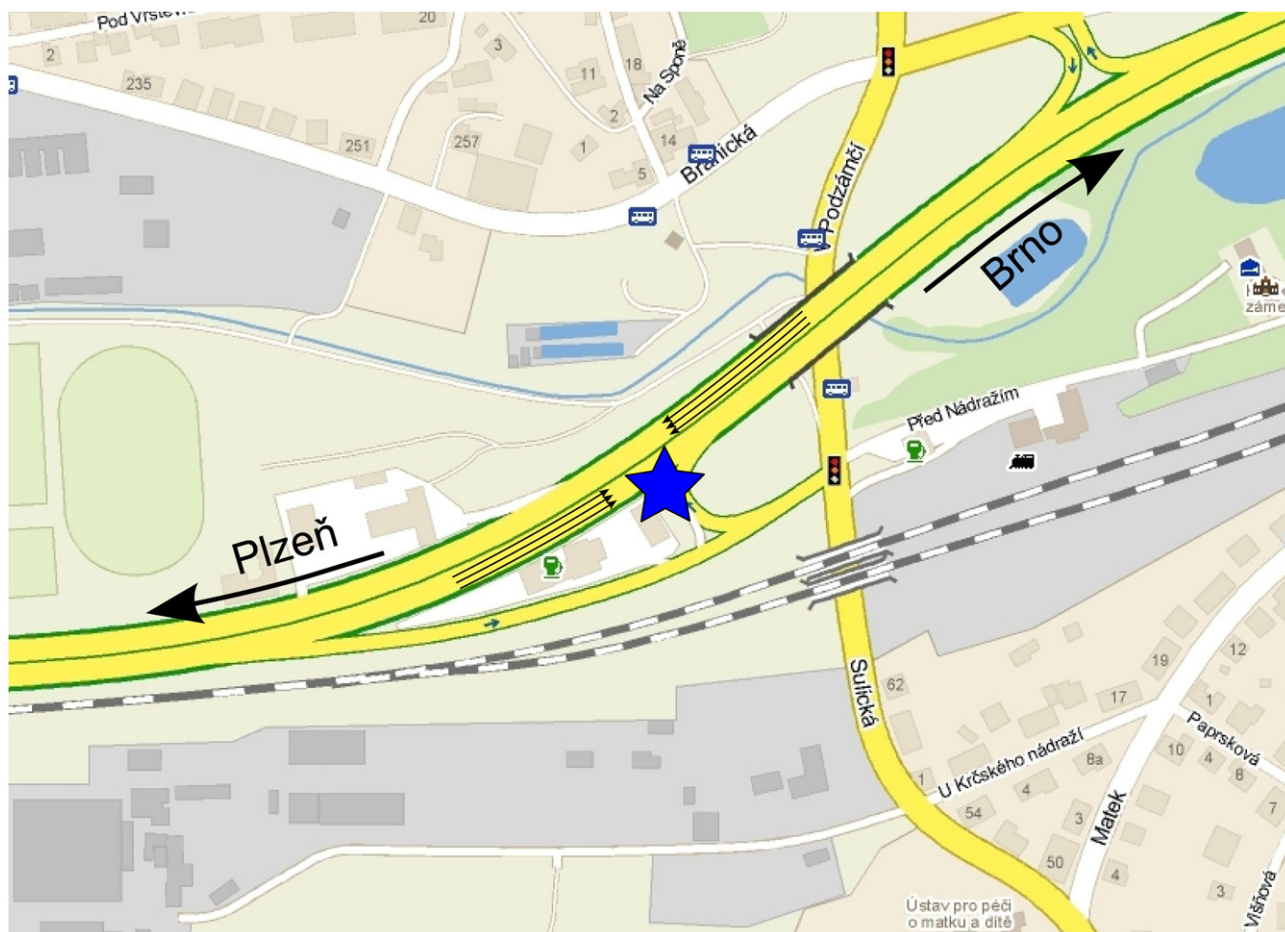


Fig. 2. Freeway measurement site.

PM10 inlet). One scan of both spectrometers was completed in 5 min. Moreover, the monitoring station continuously measures concentrations of  $\text{SO}_2$ ,  $\text{NO}$ ,  $\text{NO}_2$ ,  $\text{NO}_x$ ,  $\text{O}_3$ ,  $\text{PM}_{10}$  and meteorological data (wind speed and direction, relative humidity, temperature and radiation). During the crossroad campaign, the station included also the BLPI filled with polycarbonate foils, EC/OC field analyzer with  $\text{PM}_{2.5}$  sampling head (Sunset Lab.) and 2 LVS (Leckel) measuring  $\text{PM}_{10}$  and  $\text{PM}_{2.5}$  using QBQ technique. The sampling periods were the same as in the mobile container on crossroad.

The size resolved samples were analyzed gravimetrically (Sartorius M5P, 1  $\mu\text{g}$  resolution). Before and after the sampling campaign, the foils were equilibrated at least for 24 h in the weighing room and then the foils were weighted under controlled conditions (temperature and RH). Furthermore, concentration of water soluble cations ( $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{NH}_4^+$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Zn}^{2+}$ ) and anions ( $\text{SO}_4^{2-}$ ,  $\text{NO}_2^-$ ,  $\text{NO}_3^-$ ,  $\text{H}_2\text{PO}_4^-$ ,  $\text{Cl}^-$ ) was determined using IC (Ion Chromatography, WATREX, conductivity detector Shodex CD-5, Transgenomic ICsep AN300 150  $\times$  5.5 mm column for anions and Alltech universal cation 7  $\mu\text{m}$  100  $\times$  4.6 mm for cations). The content of following elements: Al, As, Ba, Br, Ca, Cl, Cr, Cu, Fe, K, Mg, Mn, Ni, Pb, Rb, S, Se, Si, Sr, Ti, V, Zn and Zr, was analyzed with PIXE (Proton Induced X-ray Emission). The samples on aluminium foils were analyzed for total carbon content using an RBS (Rutherford Backscattering Spectroscopy) method. Front and back quartz filters from LVS were analyzed for carbon content using the EC/OC field analyzer.

### 3. Results and discussion

#### 3.1. Traffic monitoring

The freeway traffic intensity survey was performed during the period from 10.9.2008 8:00 to 11.9.2008 8:00 (see Fig. 4). The cars were traced in both directions in each lane separately. The car types were divided into three following groups: passenger car, light truck and heavy truck. The results showed that the total number of cars during the tracked period was  $1.03 \times 10^5$ . The percentage of the different car types was 80.3%, 9.0% and 10.7% for passenger cars, light trucks and heavy trucks, respectively. The time dependence reveals decreasing percentage of the passenger cars during the night hours (between 2:00 and 5:00) down to 50% with 400 passenger cars per hour, while during the morning and evening rush hours it reached 85% and 87% with total amount of  $6 \times 10^3$  and  $5 \times 10^3$  passenger cars per hour, respectively. The threshold level of  $3 \times 10^3$  cars per hour was chosen to differentiate between rush and background hours. Therefore, the resulting period of rush and background hours was set to 6:00–22:00 and 0:00–6:00 + 22:00–24:00, respectively.

In the case of crossroads, the traffic intensity survey carried out during the period from 6:00 to 20:45, showed that the main road of the crossroads was passed by  $1.68 \times 10^4$  cars (see Fig. 4) with 87.5% of passenger cars, 7.8% of light trucks + busses and 4.7% of heavy trucks. The higher percentage of passenger cars shows that the area of Suchdol is affected by heavy trucks less than the freeway. The threshold level of  $10^3$  cars per hour was chosen in this case to distinguish between rush

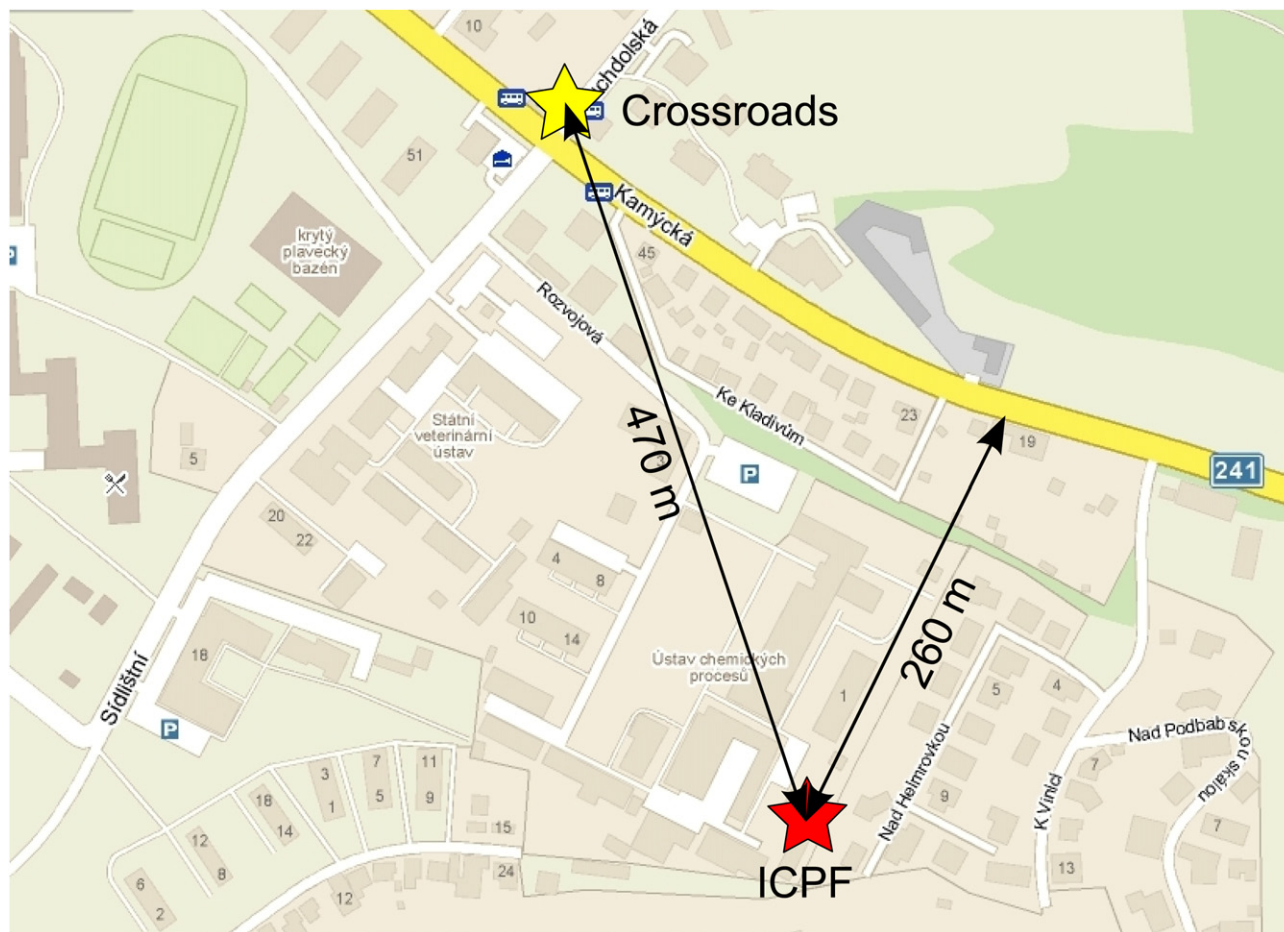


Fig. 3. Crossroads and ICPF measurement sites.

and background hours. The resulting period for rush and background hours was adjusted to 6:45–9:00 + 16:00–19:45 and 0:00–6:45 + 9:00–16:00 + 19:45–24:00, respectively.

### 3.2. Meteorological conditions

The freeway measurement campaign was conducted during the beginning of autumn. The prevailing wind direction during the measurement was south-western, almost parallel to the freeway, with the average speed of  $1.4 \text{ m s}^{-1}$ . The average temperature, relative humidity and atmospheric pressure reached the following values  $21.2 \text{ }^\circ\text{C}$ ,  $61.9\%$  and  $99.2 \text{ kPa}$ , respectively. The meteorological conditions measured at ICPF were: wind direction south-western, wind speed  $0.9 \text{ m s}^{-1}$ , temperature  $20.4 \text{ }^\circ\text{C}$  and relative humidity  $71.4\%$ .

In the case of the crossroads campaign, the measurement took place in the late spring and the meteorological conditions were monitored only at the campus of ICPF. Nevertheless, the meteorological conditions on the crossroads should be identical since the distance between the two measurement sites is only  $0.5 \text{ km}$ . The prevailing wind direction was north-eastern (blowing most of the time from crossroad sampling site towards the crossroad) with the average speed of  $2.3 \text{ m s}^{-1}$ . The average temperature and the relative humidity were  $14.5 \text{ }^\circ\text{C}$  and  $47.2\%$ , respectively.

### 3.3. Number size distribution

The Fig. 5 and Fig. 6 show the average number size distributions of the fine and coarse particles, respectively, for all sites during the

rush and background hours (see above the Traffic monitoring paragraph). Generally, the largest difference in measured particle number concentration is in the range of ultrafine particles, comparing the measurements close to the traffic sources (freeway and crossroads) and the ICPF measurements (background). Such observation is in agreement with the origin of these particles from combustion processes. It means that the particles originated mostly in the engines of cars passing the freeway or the crossroads.

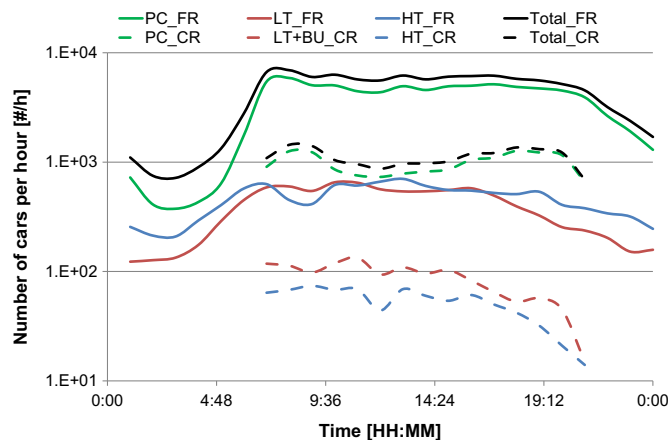


Fig. 4. Traffic intensity survey for freeway (FR) and crossroad (CR) measurement; PC – passenger car, LT – light truck, BU – bus, HT – heavy truck (freeway: 10.9.2008 8:00–11.9.2008 8:00, crossroad: 13.5.2009 6:00–20:45).

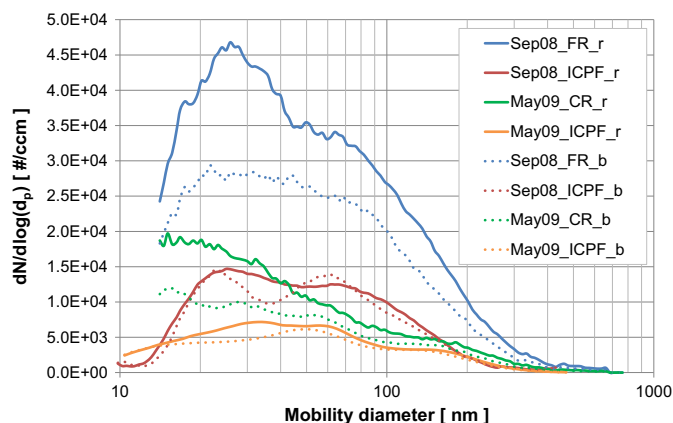


Fig. 5. Average number size distributions for fine particles during the rush (r) and background (b) hours measured by SMPS; freeway (FR), crossroad (CR).

The size distributions measured close to the freeway during rush hours (Fig. 5) and at ICPF display the bimodal size distribution with the modes located somewhere between 25–30 and between 60 and 70 nm. Such bimodal type of size distribution is often found during the measurements of direct exhaust emissions and also during measurements on the roads, measured directly behind the car or by a stationary station located nearby the road (Kittelson et al., 2006a). This kind of bimodal size distribution can result from either direct emissions of nanoparticles from the exhaust or nucleation of low-volatile vapours after their cooling (the first mode) followed by the coagulation of these particles (second mode) and/or condensation of low-volatile vapours on the primary particles. The coagulation can be found more often in case of diesel emissions, especially due to the very high concentration of particles (~10<sup>8</sup> particles cm<sup>-3</sup>), that favours fast coagulation. Nevertheless, the gasoline engines can produce the same bimodal type of size distribution. The similar bimodal pattern found at the ICPF site can be explained by prevailing SW wind that brought particles emitted by traffic from quite heavy populated SW region of Prague. The data from the freeway during background hours did not exhibit the bimodal pattern; probably even more than two modes were hidden under the wide main peak of the size distribution. This different pattern might be caused by different ratios between cars, and light and heavy trucks during background hours.

A different, multi-modal pattern of the number size distribution was observed during the crossroad measurement in May, most probably as a result of combination of the traffic and background

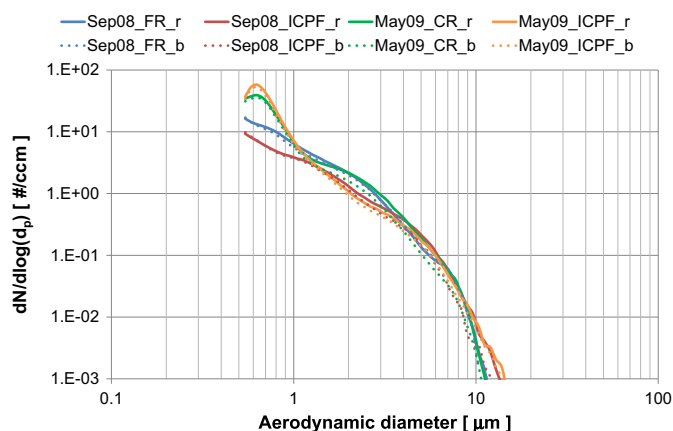


Fig. 6. Average number size distributions for coarse particles during the rush (r) and background (b) hours measured by APS; freeway (FR), crossroad (CR).

aerosol. The distributions measured at the crossroads site exhibit a maximum below 20 nm, illustrating a prevailing contribution of pipe tail emissions even though the aerosol was sampled upwind (but very close to) the crossroad. In case of the ICPF site in May, the first mode includes mostly the primary emissions from traffic. The second mode can be the result of coagulation of primary emissions and the last mode (the largest particles) covers the secondary aerosol from regional background and long range transport. The differences in concentrations measured at ICPF during two different campaigns (September and May) can be assigned to day to day variability and especially to the influence of prevailing wind direction. The difference between the concentrations measured during the rush and the background hours was much larger for the traffic site than for the background site. It again confirms that traffic was a dominant factor contributing to number concentrations of ultrafine particles during the campaigns.

The number size distributions measured using the APS spectrometers (see Fig. 6) differ mainly in particles between 0.5 and 1 μm where the regional background aerosol is a main source. The difference reflects day to day variability of background aerosol, in this case driven probably by different air mass trajectories during measurements in September and in May, as will be shown later. The coarse particles (>1 μm) show only small differences that can be explained by the measurement uncertainties of both APS instruments. If we compare the traffic (freeway and crossroads) and background (both ICPF) measurements it can be seen (Fig. 6) that the concentrations of particles about 2.5 μm in diameter are a bit elevated at both “traffic” sites, pointing to particles emitted by abrasion of tyres, brakes and the road surface. This explanation will be further supported by elemental size distributions in the following chapters.

Daily pattern of the number concentration of the ultrafine (<100 nm) and fine particles (100 nm–1 μm) shows (see Fig. 7) that the fluctuations of values measured close to the freeway or crossroads are much higher than in the case of measurement at the background ICPF station. The fluctuations are given primarily by the vicinity of the measurement place to the source of particles – the cars. In the case of freeway measurement campaign, the number concentrations measured close to the freeway reach much higher values than those measured in ICPF (see Fig. 7), but the overall daily trend follows the morning and afternoon traffic rush hours. However, the rush hours are observed at ICPF with a little delay. The maximum values measured next to the freeway rose up to 10<sup>5</sup> particles cm<sup>-3</sup>, which is more than three times higher than in the case of ICPF background site. The highest contribution to the total number concentration of particles <10 μm has the size bin

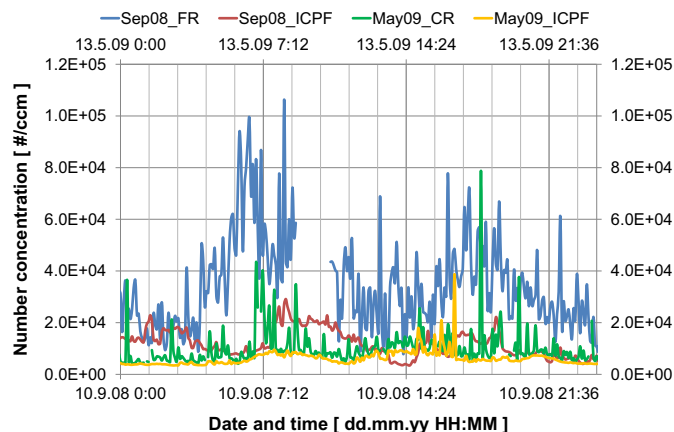


Fig. 7. The daily pattern for the ultrafine and fine mode particles (17.3–1000 nm).

**Table 1**

The average number concentrations in various size bins based on aerodynamic diameter (Sep08 – 10.9.08 7:55–11.9.08 7:55, May09 – 13.5.09 6:00–21:00).

Size bin	Sep08_FR	Sep08_ICPF	May09_CR	May09_ICPF
17.3–25 nm	5.37E+03	1.29E+03	2.81E+03	1.14E+03
25–100 nm	2.14E+04	6.17E+03	6.84E+03	4.73E+03
100 nm–1 µm	9.11E+03	2.35E+03	2.00E+03	1.36E+03
1 µm–2.5 µm	1.10E+00	7.10E–01	1.08E+00	8.30E–01
2.5 µm–10 µm	1.56E–01	1.25E–01	2.15E–01	1.55E–01

between 25 and 100 nm (see Table 1), which agrees with the sizes of particles produced by car engines.

In the case of crossroad measurement campaign small maxima can be found during the morning hours (see Fig. 7), corresponding to people leaving their homes for work. The values of number concentrations in the late morning and afternoon hours are almost constant with slight increase in the late afternoon and evening, when the people were returning home from their work. The number concentration in the size bin between 25 and 100 nm (see Table 1) is not that high here as in the case of freeway measurement. However, the measured number concentrations were strongly influenced by unfavourable prevailing wind direction as was explained above.

The comparison of one day measurement of coarse particles (1–10 µm) from both campaigns show similar daily pattern with a moderate time delay in the morning hours of September ICPF measurement (see Fig. 8). We may only speculate that this delay is related to the origin of these particles from local (broader Prague area) traffic, resuspended during rush hours. The greater fluctuation of the data measured close to the freeway or near the crossroads (compared to the values measured at ICPF) points to particles emitted from the brake and tyres abrasion as well as to particles resuspended from the road surface.

The average values of concentrations in various size bins can be seen in Table 1. The average values are calculated for the period of 1st impactor sampling during both campaigns. The approximately three times lower number concentrations in the size bins 25–100 nm and 100 nm–1 µm in the case of crossroad measurement reflect the lower traffic density in this locality. The comparison of total averaged PM<sub>2.5</sub> number and mass concentrations between places with different traffic influence (intensity) can be seen from Table 2.

### 3.4. Mass size distribution

The freeway campaign included two BLPs equipped with two different substrates – polycarbonate coated with Apiezon L (PC)

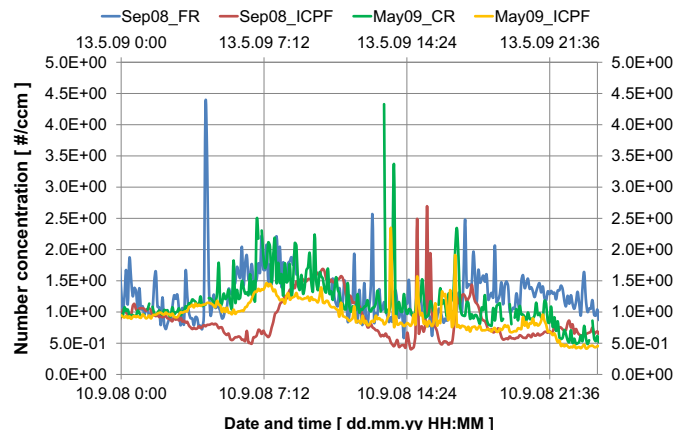


Fig. 8. The daily pattern for the coarse mode particles (1–10 µm).

**Table 2**

The average total number concentrations of particles < 2.5 µm and PM<sub>2.5</sub> mass concentrations and OC, EC and TC (Sep08 – 10.9.08 7:55–11.9.08 7:55, May09 – 13.5.09 6:00–21:00).

Method	Quantity	Sep08_FR	May09_CR	May09_ICPF
SMPS + APS	Number conc. [# ccm <sup>-1</sup> ]	3.59E+04	1.16E+04	7.23E+03
Impactor	Mass conc. [µg m <sup>-3</sup> ]	2.36E+01	1.53E+01	1.31E+01
RBS	TC [µg m <sup>-3</sup> ]	1.48E+01	–	–
Online EC/OC	OC [µg m <sup>-3</sup> ]	–	2.45E+00	2.34E+00
	EC [µg m <sup>-3</sup> ]	–	1.47E+00	8.40E–01
	TC [µg m <sup>-3</sup> ]	–	3.92E+00	3.18E+00
LVS (QBQ)	OC [µg m <sup>-3</sup> ]	–	1.68E+00	2.13E+00
	EC [µg m <sup>-3</sup> ]	–	1.99E+00	1.08E+00
	TC [µg m <sup>-3</sup> ]	–	3.68E+00	3.21E+00

and aluminium (Al). In the case of crossroads campaign, two BLPs filled with Apiezon L coated PC foils were used – one in mobile container on the crossroads and the second one in the monitoring station at ICPF. The comparison of mass size distribution from the 3 BLPs (all filled with PC foils) measuring at different places can be seen on Fig. 9.

The first mode of the size distribution measured next to the freeway, covering “ultrafines” (here <200 nm) represents particles coming from direct exhaust emissions and particles formed by coagulation of the former particles or condensation of low-volatile compounds on their surface. The second mode – fine particles (0.2–1 µm) represents mainly the accumulation mode originating mainly in regional background. The coarse particle mode (1–10 µm) includes particles emitted from the tyres, brakes and road surface abrasion as well as from the resuspension of particles from the road surface. Furthermore, it can also contain the particles from regional resuspension and local sources. Similar results, with similar modal structure, were obtained during measurements in other cities. The modal structure in our case matches the results obtained, using a 6 stage ELPI impactor (30–1000 nm), in Hongkong (Yao et al., 2007). Tri-modal size distribution, measured using 13 stage ELPI impactor (30–10,000 nm), was observed in the city of Rome, in distance about 50 m from the road, during the measurement on the parking place (Canepari et al., 2008). In this case the modes were in size range of 100–200 nm and about 5 µm.

The other two mass size distributions represent the crossroad campaign, including the crossroad measurement and the background measurement in the campus of ICPF. Both size distributions show a bimodal structure with modes around 300 nm and 5 µm. The first mode is typical for ICPF background station (observed during many previous years, e.g. Schwarz et al., 2005) and comprises mainly secondary aerosol particles from regional

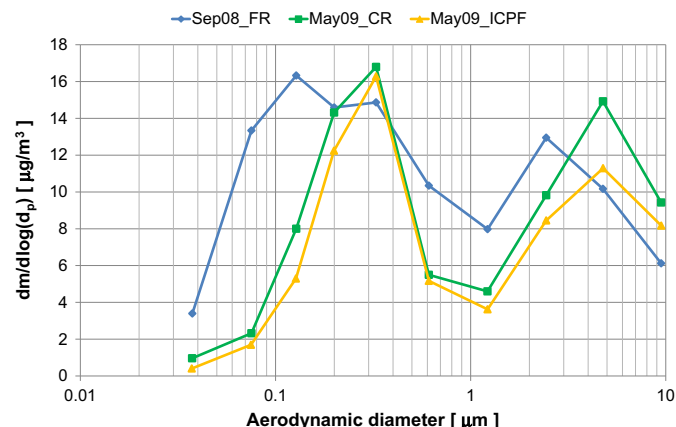


Fig. 9. The mass size distribution from BLPs.

background (ammonium sulphate and nitrate). The second mode corresponds to the resuspended aerosol particles and particles formed by mechanical friction including mainly particles of traffic origin. The comparison of crossroad and ICPF background measurement reveals little differences in fine and also coarse mode caused by the traffic emissions, in the case of fine particles the exhaust emissions and in the case of coarse particles the particles emitted by abrasion and resuspension. The figure also shows a distinct shift of the crossroad size distribution towards smaller particles, pointing again to the effect of exhaust emissions.

The differences in modal structure of submicron particles measured close to the freeway and at the crossroad can be attributed to the much higher concentrations of fresh ultrafine particles from traffic at the freeway site that were able to form separate mode even in mass size distribution. In the case of coarse mode particles, the higher concentrations for crossroads campaign (even for the background ICPF samples) could have been caused by construction works taking place close to both measurement sites. Also, the differences of total PM<sub>2.5</sub> averaged mass concentrations between places having different traffic intensity can be seen from Table 2. The total PM<sub>2.5</sub> mass concentrations decrease with decreasing traffic intensity in the following sequence: freeway → crossroads → ICPF background. However, it should be noted that only measurements done the same day may be directly compared.

### 3.5. Size resolved ionic composition

The size resolved concentrations of water soluble ions obtained during the freeway campaign can be seen in Fig. 10. The results show that the accumulation mode (in this case particles about 0.3 μm in diameter) contains sulphate, nitrate and ammonium, coming predominantly from the regional background and they are almost solely of secondary origin (Viana et al., 2008). The coarse fraction of nitrate ions originates most probably from resuspension, but also other processes may play their role here. The Na<sup>+</sup> ions might appear as a result of long range transport of the sea salts, where the NaCl is transformed in the atmosphere by nitric acid to form sodium nitrate during the transport (Finlayson-Pitts and Pitts, 2000).

In the case of the crossroad campaign, the analysis of the main ions shows that the main portion of the first mode of the size distribution is composed mainly of ammonium sulphate and nitrate (see Fig. 11), in other words the substance typical for the inorganic part of secondary aerosol. The similar concentrations of these ions next to the crossroad and at the background ICPF station denote their origin in regional secondary aerosol. The coarse mode is dominated by the nitrate anion. Regarding the minor ions (not

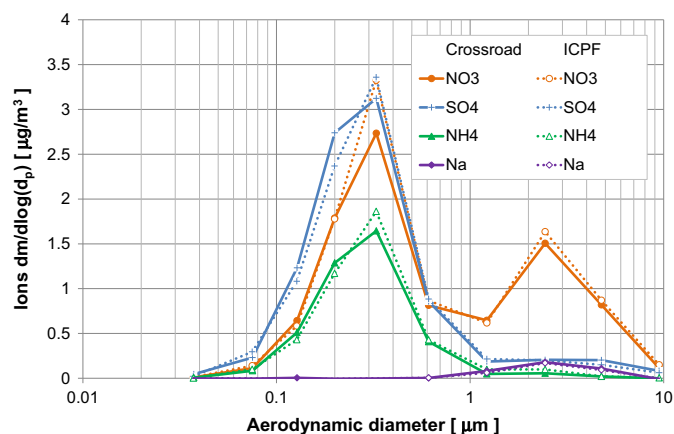


Fig. 11. The mass size distribution of main ions from BLPI measured next to the crossroads and at ICPF (May 2009).

shown because of very low concentrations), the prevailing ion in the coarse fraction of the size distribution is Na<sup>+</sup>. Furthermore, similar concentrations of Na<sup>+</sup> at both places (crossroad and ICPF) support the statement that it comes from the long range transport. In this case the particles are most probably formed by the sea salts again. This is in agreement with the position of the nitrate anion in coarse particle mode.

Comparing the two traffic measurements, freeway and crossroad, we can see much bigger concentrations of sulphate, nitrate and ammonium in the case of crossroads campaign. This was probably caused by different meteorological conditions. Moreover, meteorological conditions in May 2009 probably favoured higher accumulation of aerosol particles in the atmosphere. This fact increases the importance of the relative particulate enrichment at freeway compared to the crossroad with smaller traffic intensity. The concentrations of the other ions are very similar in both places, which again points to the background aerosol influence.

### 3.6. Size resolved elemental composition

The PC foils from impactor measurements were also analyzed using PIXE for content of several elements (see Figs. 12–15).

The analysis of samples collected during the freeway campaign reveals a group of elements with similar behaviour regarding their mass size distribution. The similar shape of the size distribution may indicate identical origin of these elements. The first and the most pronounced group is represented by Fe, Cu, Mn and also Zn having mode about 2.5 μm (see Figs. 12 and 14). These elements may originate from the brake abrasion or from the abrasion of various engine parts (Sternbeck et al., 2002; Sanders et al., 2003; Wählin et al., 2006; Hjortenkrans et al., 2007; Canepari et al., 2008; Handler et al., 2008; Johansson et al., 2008). Another important source of Zn in the coarse mode is tyre abrasion (Wählin et al., 2006; Hjortenkrans et al., 2007). Thus Zn is often used as a marker for tyre emissions (Hopke et al., 1980). Furthermore, the broad size distribution of Zn having also another ultrafine mode reflects various sources of Zn. The origin of the ultrafine Zn particles can be attributed also to the additives of the engine oils (Huang et al., 1994; Handler et al., 2008; Johansson et al., 2008). These elements are considered to be the typical group for traffic emissions (Viana et al., 2008).

The second most important group of elements include Si, Al, Ca and coarse mode of K centred at about 5 μm (see Fig. 12), representing the elements typical for the earth crust. Since these elements are considered to be the typical representatives of regional resuspension, so called “city dust” and road dust (Viana

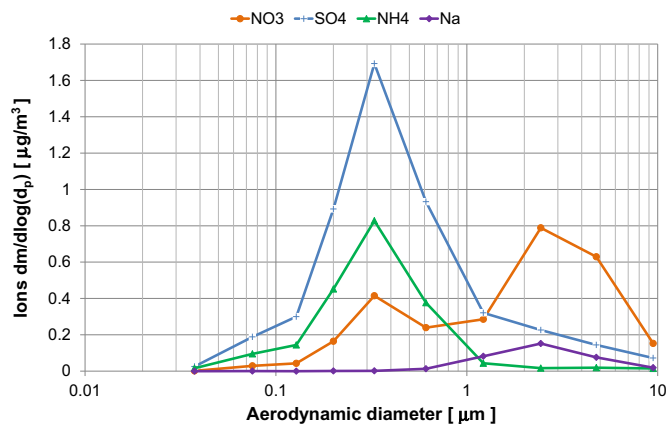


Fig. 10. The mass size distribution of main ions from BLPI measured next to the freeway (September 2008).



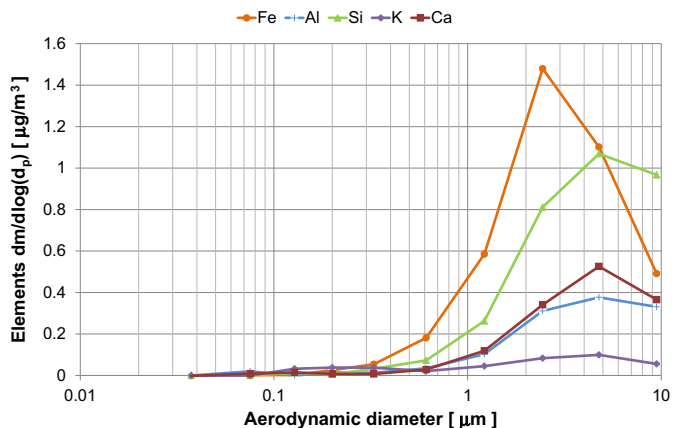


Fig. 12. The mass size distribution of main elements from BLPI measured next to the freeway (September 2008).

et al., 2008) they may origin from the resuspension of particles from the road surface as well as from the abrasion of the road.

In the case of the crossroad campaign, the concentrations of K, Al and Si in the coarse mode (see Fig. 13) are almost the same at both sites (crossroad and ICPF). These elements are again typical for earth crust and most probably originate from regional resuspension. The higher concentrations of Ca (for particles > 5 µm) measured at the crossroad may also come from the construction works taking place close to the measurement site. The elevated concentrations of Fe measured close to the crossroad most probably originate from the traffic, namely from the brake abrasion (Hjortenkrans et al., 2007). This hypothesis is supported by the position of Cu and Mn modes (see Fig. 15). The Zn concentrations are also influenced by the traffic emissions (see Fig. 15) as was already mentioned before. The elevated concentration of Ba in the coarse mode represents the particles emitted from the brake mechanical abrasion. Possible source of Ba in fine fraction might be also the additives of engine oils.

Generally, comparing the elemental analysis from the two campaigns (see Figs. 12–15) we can see that the concentrations of elements connected with traffic emissions (e.g. Fe, Al, Si, Cu, Zn) have elevated concentrations near the freeway with higher traffic intensity comparing to the crossroad measurement. But at the same time, when we compare the concentrations of the same elements measured close to the crossroads and at ICPF background station and we find similar concentrations, we can attribute some of these

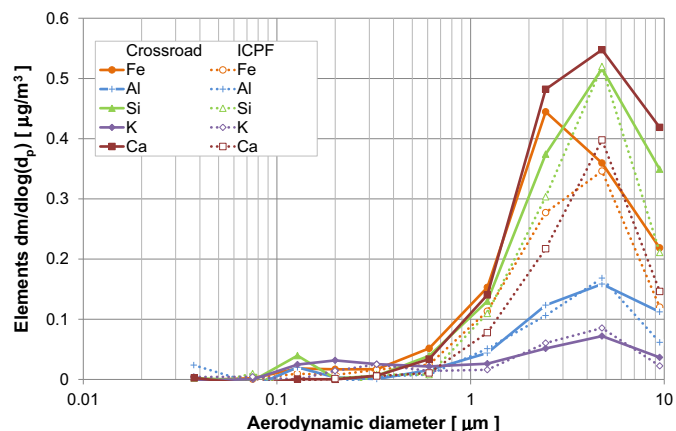


Fig. 13. The mass size distribution of main elements from BLPI measured next to the crossroads and at ICPF (May 2009).

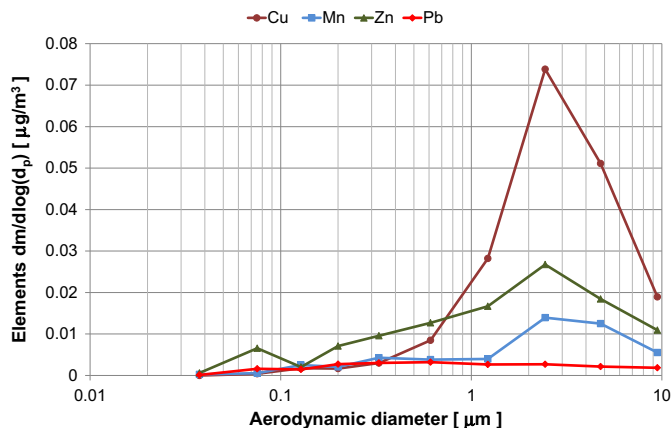


Fig. 14. The mass size distribution of minor elements from BLPI measured next to the freeway (September 2008).

elements also to the regional background. Furthermore, some of the elements (e.g. K), having similar concentrations at both traffic influenced sites and also at ICPF, indicate origin of these elements from long range transport or as a regional background aerosol.

### 3.7. EC/OC analysis

Concentrations of elemental (EC), organic (OC) and total carbon (TC) were analyzed using different methods for both traffic campaigns. In the case of freeway measurement campaign, the samples for RBS analysis were collected using the BLPI on aluminium foils. The results give us information about size resolved concentration of TC. In the case of the crossroad measurement campaign, the on-line (1 or 2 h time resolution) concentrations of elemental and organic carbon were measured using the EC/OC field analyzer, equipped with a denuder and PM2.5 sampling head, and at the same time the PM2.5 and PM10 samples were collected on quartz fibre filters using the LVS samplers. The quartz fibre filters were afterwards analyzed using the EC/OC field analyzer (off-line analysis). These results give us time resolved information about EC, OC and TC PM2.5 concentration and simultaneously the integral PM2.5 and PM10 concentrations of the same quantities.

It can be seen from the comparison of the values in Table 2, that the highest concentration of TC was measured near the freeway (almost 4 times higher than the crossroad concentration and 5 times higher than concentration measured at ICPF). Unfortunately,

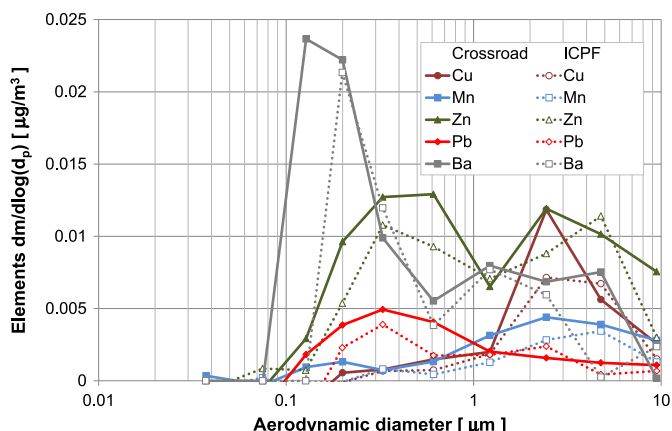


Fig. 15. The mass size distribution of minor elements from BLPI measured next to the crossroads and at ICPF (May 2009).

for this measurement we have only size resolved analysis of TC and thus we do not have any information about EC and OC concentrations, but most probably a big portion of TC is composed of OC, but the percentage of EC (from the fuel combustion) will be higher than in case of the other measurements. Such hypothesis can be confirmed by comparing the differences between EC measured near the crossroads and at ICPF (almost 2 times higher), while the OC concentrations are almost the same at both sites (coming from background sources). The mass size distribution of TC measured next to the freeway shows that large portion of the mass of ultrafine particles (here particles < 200 nm) consists of the TC (carbon and carbonaceous substances). Nevertheless, the RBS analysis of carbonaceous particles was burdened with relatively large uncertainty. The TC contained in the coarse fraction may originate mainly from the tyre abrasion, which is in agreement with position of Zn coarse mode (see Fig. 14), considered to be a marker for tyre emissions. The influence of primary biogenic particles can be also important.

#### 4. Conclusions

The measurement campaigns of traffic emissions have been conducted at two different sites within the Prague city (with different traffic intensity) together with simultaneous measurement of the same parameters at the background station located in the suburban area of Prague.

Measurements of particle number size distributions showed that the main contribution of traffic can be found in the ultrafine range (particles < 100 nm). Traffic ultrafine particles consist of two modes, one at 25–30 nm, coming from direct exhaust emissions and nucleation of low volatility vapours and second at 60–70 nm formed mainly by coagulation of the ultrafine particles. The accumulation mode particles (100–1000 nm) were mainly of the regional background aerosol origin. The coarse particles (>1 µm) concentrations were influenced mainly by the regional background aerosol, including the aerosol emitted from the construction works taking place close to the crossroad, and also the aerosol from the long range transport. Nevertheless, slightly elevated concentrations of particles with mode about 2.5 µm can be seen for both of the “traffic” related measurement campaigns, which corresponds to the particles emitted from the brake and tyre abrasion, the abrasion of the road surface and at the same time the resuspension of aerosol particles from the road surface. The comparison of particle number size distributions during rush hours showed a strong influence of prevailing wind direction on the resulting data in the case of crossroad measurement (multi-modal shape). The time series for ultrafine and fine size bins showed more fluctuations for the data measured close to the traffic pollution sources, caused by the passing cars.

The mass size distribution shows very similar results to the number size distribution. The place with the highest traffic intensity (the freeway) has the highest mass concentration of the ultrafine particles. It means that the fine mode was split into two modes in case of freeway and shifted towards smaller diameters in case of crossroad compared to the mass size distributions at the background ICPF site. Discussing the mass size distribution in contrast to number size distribution, here the first mode corresponds to the primary exhaust particles, particles formed by their coagulation and by condensation of low-volatile vapours on their surfaces. The coarse mode includes particles from the regional background as well as the particles from traffic (abrasion of tyres, brakes, etc.). The increased concentrations in the coarse modes of the crossroad campaign (crossroad and ICPF) show slightly elevated values, which may be given also by a contribution of the construction works in the vicinity of the crossroad.

The water soluble ions found in fine particle mode ( $\text{Na}^+$ ,  $\text{NH}_4^+$ ,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ ) are mainly of the regional background origin, while the nitrate found in coarse particle mode comes from long range transport. So it can be generally stated that most of the water soluble ions are not related to traffic emissions.

Concerning elemental composition, most of the major elements are located in the coarse mode. The elements emitted mainly due to the brake abrasion or the abrasion of various engine parts are represented by Fe, Cu, Mn and also Zn. Moreover, Zn can originate also from the tyre abrasion. The Si, Al, Ca and coarse mode of K (about 5 µm) are the main representatives of particles coming from the resuspension of particles from the road surface as well as from the abrasion of the road. The K particles have origin mainly in the long range transport or as a regional background aerosol. Furthermore, the higher concentrations of Ca measured during the crossroad measurement can be attributed to the construction works taking place close to the crossroads.

The PM<sub>2.5</sub> concentrations of TC have decreasing trend with decreasing traffic intensity, in the following order: freeway, crossroad, ICPF (background), which is caused mainly by decreasing of the EC content, being proved by the crossroad campaign and comparison of EC and OC values.

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