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Winter and summer time size distributions and densities of traffic-related aerosol particles at a busy highway in Helsinki

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Abstract

The number concentration and size distribution of traffic related particles were measured at road-side in Helsinki. Two winter campaigns took place in 10–26 February 2003 and 28 January–12 February 2004 and two summer campaigns in 12–27 August 2003 and 6–20 August 2004. The measurements were performed simultaneously at distances of 9 m and 65 m from the highway. This study concentrates on data that were measured when the wind direction was from the road to the measurement site. The total concentration in winter time was 2–3 times higher than in summer time and it was dominated by nucleation mode particles. The particles smaller than 63 nm (at aerodynamic size) constitute ~90% of all particles in winter time and ~80% of particles in summer time. The particle total concentration increases with increasing traffic rate. The dependence of particles smaller than 63 nm on traffic rate is stronger than for particles larger than 63 nm both during summer and winter. The particle distribution at the roadside consists of two distinguishable modes. The GMD of nucleation mode (Mode 1) was 20.3 nm at summer and 18.9 nm at winter. The GMD of the larger mode (Mode 2) was 72.0 nm at summer and 75.1 nm at winter. The GMD values of the modes do not depend on traffic rate. The average density value for Mode 1 particles was 1.0 g/cm^3 both in summer and winter time, while the average density value for Mode 2 was 1.5±0.1 g/cm^3 and 1.8±0.3 g/cm^3 for summer and winter time, respectively.

1. Introduction

Traffic is one of the most significant sources of fine particles in urban environment. This has been shown earlier in many studies (Wåhlin et al., 2001; Molnár et al., 2002; Burón et al., 2004; Janhäll et al., 2004; Kittelson et al., 2004; Kristensson et al., 2004; Weijers et al., 2004). Recently, also the (number) concentration of particles measured at urban background was related to traffic flow rate (Van Dingenen et al., 2004; Hussein et al.,

2004) .

Based on results obtained in laboratory studies, the fine particles in vehicle exhaust are distributed into two modes. The nucleation mode particles are small (number based geometric mean diameter ~5–30 nm) liquid particles consisting mainly of hydrocarbons, water and sulphates (Kittelson, 1998; Khalek et al., 2000). The nucleation mode particles form during dilution of the exhaust in the atmosphere (Abdul-Khalek, 1999; Kittelson et al., 2000). The larger particle mode (number based geometric mean diameter 40–100 nm) in the vehicle exhaust consists of soot particles and volatile materials condensed on them (Kittelson et al., 2000; Harris and Maricq, 2001). These soot particles are agglomerates formed in the engine during the combustion process. The soot particles are emitted mainly form diesel vehicles, whereas gasoline engines emit less soot (Harris and Maricq, 2001). The nucleation mode particles are related both in diesel and gasoline vehicles, and their emissions depend more on the external conditions such as dilution etc.

Due to the worldwide increase in traffic intensity, the traffic related pollutant problem is relevant and it is hard to solve, even if the developing vehicle technology gradually reduces emissions. In addition, new emission technologies can bring along new problems. For example, the diesel particle filters that are used for removing soot particles from the exhaust gas, have been reported to enhance formation of nucleation mode particles (Vaaraslahti et al., 2004).

The size and concentrations of traffic related particles at road-sides have been widely studied during recent years (Morawska et al., 1999; Hitchins et al., 2000; Wåhlin et al., 2001; Molnár et al., 2002; Sturm et al., 2003; Ketzel et al., 2004; Kristensson et al., 2004; Janhäll et al., 2004). In this paper, the concentration of different sized particles measured at road side is connected to traffic rate. Also the differences between the emissions during winter and summer time are studied. Specially, the characteristics of modes appearing in measured particle size distributions at the road-side are investigated. In addition, the average density of particles in modes will be determined. There are very limited amount of information concerning the particle density at urban measurement sights. Stein et al. (1994) measured the density of atmospheric particles in size range 0.1–0.25 μm. The measured densities varied from 1.60 to 1.79 g/cm^3. McMurry et al. (2002) used DMA-APM technique to analyze the density of urban aerosol particles of size 0.107 μm and 0.309 μm. They found that 0.107 μm particles had densities between 1.35 and 1.7 g/cm^3. For 0.309 μm particles they found two ranges of density values: low values between 0.35 and 0.65 g/cm^3 and higher values approximately 1.6 g/cm^3. According to authors' knowledge, density values for urban aerosol particles in the size range of nucleation mode have not been reported before.

2. Description of measurement campaign

The measurement site was located in Helsinki, Herttoniemi, about 6 km to east from the city centre. The investigated highway (Itäväylä) is one of the main roads in Helsinki area and its direction is northeast from the city centre (Fig. 1). The highway consists of 3 lanes in both directions. The measurements were part of the Finnish project LIPIKA ("Correlation between fine particle emissions of traffic and laboratory tests of vehicles") and they were performed during 4 campaigns. Two winter campaigns took place in 10–26 February 2003 and 28 January–12 February 2004, and two summer campaigns in 12–27 August 2003 and 6–20 August 2004. The measurements were performed simultaneously at distances of 9 m and 65 m from the highway. The sampling heights were 5.7 m above the ground level.

Next to the measurement site (35 m to north from 65 m cabin) a factory was located. Its emissions were clearly distinguishable from traffic emissions (strong concentration peak at 30 nm). Based on the measured wind direction, the data with wind direction directly from the factory towards the measurement site was omitted.

2.1. Instrumentation

The total number concentration at the road-side was measured with a condensation particle counter (CPC, TSI, model 3025). The detection limit of CPC 3025 is 3 nm. Due to the high particle concentrations especially during the rush hours, a passive diluter with dilution ratio ~1:4–1:6 were used in CPC measurements. The average and maximum and minimum values for particle number concentration, measured by CPC, was recorded every 5th second. During the campaigns at 2004 the Scanning Mobility Particle Sizer designed for nano particles (nano-SMPS: nano-DMA, TSI+CPC 3025) was used in 9 m cabin (measurement range 3 nm–57 nm). Scanning Mobility Particle Sizer (referred further as “long-SMPS”) with DMA model 3071 and CPC 3025 was used in 65 m cabin during all campaigns. The “long-SMPS“ measurement covered the size range of 5 nm–160 nm. Electrical Low Pressure Impactor (ELPI) with filter stage was used in both cabins, covering the particle size range of 7 nm–6.6 μm. The results concerning particle concentrations, shown later, were measured at 9 m cabin. The size distribution data measured in 65 m cabin were used only, when the modes in size distribution and particle density were investigated.

In addition to stationary measurement sites, the mobile measurement unit, “Sniffer” (Pirjola et al., 2004), was used in background concentration measurements. The urban background concentrations were measured at Saunalahti, about 600 m northwest from the measurement site. The background particle concentrations were measured by SMPS and ELPI.

2.2. Traffic count

The traffic count was performed at Itäväylä by Helsinki City and Finnish Road Administration by automatic traffic measurement system about 3 km from measurement site towards the city center. The long term and continuous traffic rate measurement provides hourly averages. The traffic to both directions at the highway is calculated separately. In addition, the number of heavy-duty vehicles was determined by the measurement system.

2.3. Meteorological conditions

The meteorological data (wind speed, wind direction, temperature and relative humidity) were measured at 9 m cabin with Vaisala weather station. The maximum, minimum and average values for temperature and relative humidity values are shown in Table 1. Also the average values for the day time (i.e. 06:00–20:00) are shown in Table 1.

The data was classified according to wind direction similarly with the method by Pirjola et al. (2005). This paper concentrates on results of wind sector S1, which consists of wind directions 255°–345° (wind blowing from the road to the measurement cabins). The wind sector is marked in Fig. 1 with dashed lines.

3. Method to define particle density

The method used in this study for determination of particle density, is based on parallel method described by Ristimäki et al. (2002) and Virtanen et al. (2004). The parallel method is based on distribution measurement performed by ELPI and SMPS and further on the relationship between particle aerodynamic size, mobility size and effective density. The basic idea is to minimize the difference of the measured ELPI currents and currents simulated by using SMPS number distribution and ELPI response functions (i.e. the charger efficiency and impactor kernel functions). The minimization can be made by altering the particle density. It should be noted here, that in parallel method, the primary output data of ELPI, i.e. the measured current signals are used. The ELPI number distribution is not calculated from measured currents at any point, thus the problems caused by using the impactor stage cut point concept or inversion can be avoided. Instead, the charging and collection of SMPS number distribution in ELPI is simulated by using the ELPI charger efficiency curve and impactor kernel functions, determined for the specific individual impactor used. The particle collection in impactor depends on particle density. Thus, by comparing the simulated currents and measured ELPI currents the density of particles can be found. Ristimäki et al. (2002) described the method for unimodal distribution that consists of constant density particles. Virtanen et al. (2004) extended the method to be suitable for fractal-like aggregates, whose density decreases with increasing particle size.

To be able to apply the parallel method in road-side particle studies the method was modified to be suitable for multi-modal distributions. In this modification, the lognormal distributions are fitted into the measured SMPS data. These fitted lognormal distributions are then used in ELPI current simulation instead of measured SMPS distributions. The fitting is done by assuming 3 modes in SMPS measurement range (5 nm–160 nm). To limit the degrees of freedom in density search procedure, the constant density of each mode is assumed. Thus as a result, the average density of each mode is found.

The method was tested in laboratory by using two test oils: Fomblin and di-octyl sebacate (DOS). The density of Fomblin is 1.9 g/cm^3 and the density of DOS is 0.91 g/cm^3. Bimodal distributions with one mode consisting of Fomblin and the other of DOS, were generated in laboratory by using the tube furnace for Fomblin and nebulizer with condensation-evaporation cycle for DOS. The geometric mean diameters of DOS distributions were varied between 40–50 nm and of Fomblin distributions between 90–150 nm. The resulting densities for DOS and Fomblin were 0.8±0.08 g/cm^3 and 1.8±0.26 g/cm^3, respectively.

4. Results

4.1. Correlation of particle emissions and traffic rate

All results concerning traffic rate and particle concentration etc. presented in this section were obtained at wind directions S1 during the weekdays (Monday–Friday).

In comparison to Finnish average values, the traffic rate at Itäväylä is high. The day time (06:00–20:00) average traffic rate was 3290 vehicles/hour during the summer campaigns and 2910 vehicles/hour during the winter campaigns. The average values for traffic rate measured during the campaigns are presented in Fig. 2a. The traffic rate is peaking during the morning and evening rush hours i.e. 06:00–10:00 and 15:00–18:00. During the rush hours, the traffic rate reaches ~4000 vehicles/hour. There is no remarkable difference between the traffic rates during the winter and summer campaigns or between the morning and evening rush hours.

In Fig. 2b the particle concentrations measured with CPC at 9 m distance from road are shown. The dashed lines represent the average day time background concentration values for winter and summer time (light gray line for summer time and dark gray line for winter time). No continuous background data was achievable. Thus, the average value of background concentration is calculated from occasional measurement periods made by the mobile laboratory. The road side concentrations follow the same temporal pattern as the traffic rate. Figure 2b shows, that at winter time the concentration peaking is a bit stronger during morning rush hours than during the evening rush hours. Morawska et al. (1999), Williams et al. (2000), Molnár et al. (2002), Wehner et al. (2002), Charron and Harrison (2003), and Janhäll et al. (2004) observed stronger concentration peak during the morning rush hour. Wehner et al. associated the higher morning concentrations with the higher truck traffic rate. Molnár et al. attributed their results to higher wind speeds and more effective vertical mixing during the afternoons. In the case of Itäväylä, there is no difference in portion of heavy duty traffic during the morning and evening rush hours. During the summer campaigns the wind speeds were lower in the morning, but at winter there were no remarkable temporal differences in wind speeds. We assume that in our case, the effect can be explained mostly by vertical mixing between high concentration air at ground level and upper air having lower particle concentration. Because of meteorological reasons, the mixing height is systematically larger in the afternoon than in the morning hours. Therefore, when mixing with larger amounts of cleaner air from up above, this directly results in lower particle concentrations for the afternoon rush hour.

The minimum concentration values take place at 03:00. Hussein et al. (2004) reported the same hours for the minimum and maximum concentrations measured at the urban background stations located at Kumpula and Siltavuori in Helsinki. They measured particle concentrations and distributions. The reported morning rush hour concentrations were approximately 25 000 #/cm^3 and 17 500 #/cm^3 during the summer and winter seasons correspondingly. The average values for morning rush hour concentrations at road-side of Itäväylä are approximately 100 000 #/cm^3 in winter time and 70 000 #/cm^3 in summer time. Thus, the particle population at road-side and also the distribution characteristics measured at road-side differ considerably from those reported by Hussein et al.

The winter concentrations are approximately double compared with the summer concentrations. This can be seen in Fig. 3, where the particle concentrations are presented as a function of traffic rate. The day time background concentrations are marked with dashed lines. The averaged night time background concentrations, calculated by using occasional measurement periods, are ~6000 #/cm^3 and 10 000 #/cm^3 for summer and winter time, respectively. All measured data points shown in Fig. 3, with traffic rate <1000 vehicles/hour, are measured during night time. From Fig. 3, it can be seen that both the summer and winter time total concentrations increase with increasing traffic rate. A best fit to the data points was found to obey the form $y{\sim}x^a$, where y is particle concentration and x is traffic rate. The exponent "a" has value 0.62 and 0.69 for summer and winter time results, respectively (Table 2.). There is no physical explanation to the form of the function that gives the best fit.

To find out the relationship between different particle size fractions and traffic rate the particles are separated into two size classes based on cut-point of the 2nd impactor stage of ELPI. The cut point of the 2nd stage is 63 nm. The concentration of the particles in the size fraction of 63 nm–6.6 μm is calculated from ELPI distribution by integrating the particle concentrations measured on stages 2–11. This size range is referred further as "d_p >63 nm". The ELPI filter stage sometimes overestimates the number concentration of the smallest particles (7–30 nm). Due to this, the concentration of particles smaller than 63 nm is calculated by subtracting the concentration of

63 nm–6.6 μm particles from the total number concentration measured by CPC. The CPC detection limit (~3 nm) gives the lowest size limit of this smaller size fraction. This size fraction is referred further as "d_p<63 nm"

In Figs. 4a and b, the concentrations of two particle size classes are presented as a function of traffic rate. In Fig. 4a, the summer and winter concentrations for d_p<63 nm particles are presented. These particles are mostly nucleation mode particles formed during the dilution process in the exhaust plume. In addition, part of the traffic related soot particles belongs to this size fraction. Also a part of the Aitken mode, which is characteristic for urban background, is in this size range (Hussein et al., 2004). In Fig. 4b the concentration of particles larger than 63 nm is presented. These larger particles contain the traffic related soot particles with volatile material condensed on them. In addition, this size range contains Aitken mode particles and aged, not traffic related accumulation mode particles (Longley et al., 2004; Molnár et al., 2002). The concentration of aged accumulation mode particles can be remarkable especially when the traffic rate is low.

For the both size fractions the dependence of concentration (y) on traffic rate (x) is of form $y \sim x^a$ as it was for the total concentration. The "a" and R^2 values are presented in Table 2. The exponent "a" has the same values for total concentration and d_p<63 nm particle concentration. This is because the concentration of d_p<63 nm particles dominates the total particle concentration: the d_p<63 nm particles constitute ~90% of particles at winter time and ~80% of particles at summer time. The exponents "a" for smaller size fraction data (d_p<63 nm) differ in summer and winter time. This is due to the difference in relationship between background concentrations and roadside concentrations. In summer time, the background particle contribution to the concentration of d_p<63 nm particles at the roadside is approximately 2 times higher than in winter time. Naturally the total concentration follows the same pattern. On the other hand, there is no seasonal difference in exponent "a" of larger size fraction (d_p>63 nm) data. This is due to the similar contribution of background concentration to total concentration in this size fraction during both seasons.

Table 2 shows, that also the exponent of the lines fitted into different size fraction data differs both in summer and winter time. The reason is the same as it is in the case of seasonal difference explained above: the fraction of background concentration of d_p>63 nm particles is larger than of d_p<63 nm particles.

Figures 4a and b indicate that the winter concentration of d_p<63 nm particles is approximately double compared to the summer concentration while there is no clear seasonal difference in the concentration of d_p>63 nm particles. The difference in seasonal behavior of these two size fractions is related to their formation mechanisms. The soot particles are formed during the combustion. According the laboratory studies by Ristimäki et al. (2005), the temperature of engine intake air does not affect remarkably the soot particle concentration even if the intake air temperature is below 0°C as it was during the winter campaigns. Thus, the soot formation seems to be insensitive to the temperature of surrounding air. On the other hand, the formation of nucleation mode particles is sensitive to dilution conditions, such as temperature and relative humidity of surrounding air. According to studies of Kittelson et al. (2000), the low ambient temperatures favor nucleation at vehicle exhaust. In laboratory studies of Ristimäki et al. (2005), the effect of low dilution temperature on nucleation mode particle concentration depends on vehicle technology. In vehicle test-cycle measurements the low dilution temperatures enhanced nucleation mode formation modestly at most cases.

The nano-SMPS data measured at 9 m could not be used in number concentration comparison for size fractions, because the nano-SMPS concentrations were significantly lower than the CPC concentrations. The average day time concentrations calculated from nano-SMPS were only 40% of CPC concentrations at winter and 15% of CPC concentrations at summer. This large difference can not be explained by narrow measurement range of nano-SMPS (3–60 nm). From the distributions measured with “long-SMPS” (measurement range 5–160 nm) at 65 m it can be seen, that the nucleation mode particles smaller than 60 nm dominate the distributions both during the winter and summer time. Calculated from “long-SMPS” measurements at 65 m, the fraction of particles smaller than 60 nm is approximately 80% and 90% during the summer and winter time, respectively. The nano-SMPS data was still used to study the dependence of nucleation mode (d_p=3–30 nm) particle concentration on vehicle density. The concentration of particles in size range 3–30 nm calculated from nano-SMPS data was normalized in respect with the calculated maximum concentration. The results are shown in Fig. 5., where it can be seen, that the concentration of nucleation mode particles increases with increasing traffic rate. Due to the low number of data points, no line was fitted to data points. But the concentration dependence of particles in size range 3–30 nm on traffic rate is similar to that of total concentration shown in Fig. 3. Thus, it is evident that the nucleation mode particles at the road-side of Itäväylä are strictly related to the traffic rate. This is contradictory to the study of Charron and Harrison (2003). According to them, the concentration of nucleation mode particles measured at Marylebone Road at London, UK was not related to the traffic intensity.

4.2. Characteristics of road-side distributions

In Fig. 6, the typical measured SMPS size distributions for winter and summer time are shown. The distributions are measured with SMPS equipped with “long- DMA”, which was located at 65 m far from road. It should be noted here, that all results presented in this chapter are calculated from the data measured at 65 m distance from the road due to the wider measurement range of SMPS used in 65 m compared to the nano-SMPS at 9 m. Figure 6 shows, that both distributions are dominated by small, probably nucleation originated particles with peak size approximately at 20 nm. Similar observation is reported also by Wehner and Wiedensohler (2003) and Ketzel et al. (2004). On the right side of the distribution, the other mode can be seen. The mode is more distinguishable in the summer than in the winter.

To find out the characteristics of particle size distributions multi-lognormal fitting for measured distributions were done. The lognormal distribution fitting was done in the similar way as in the case of particle density definition procedure described above. The measurement range of SMPS was 5–160 nm. At most of the cases, only 2 lognormal size modes were found in the distribution, marked as “Mode 1” and “Mode 2” in Table 3, where the average values of day time (i.e. 06:00–20:00) results are shown. Only in few cases, a third mode existed with size >110 nm. These results differ from the results reported by Hussein et al. (2004). They found mostly 3 modes in Helsinki: average geometric mean diameter (GMD) of nucleation mode was at ~10–15 nm, Aitken mode at 40–50 nm and accumulation mode at ~150 nm. Their measurement sites were at Kumpula and Siltavuori, which represent rather the background sites than road-side sites. Thus the concentration values were significantly smaller (about 4 times lower) than those reported in this study and the traffic related particle emissions were mixed into more aged urban background. At the road-side, the distribution is dominated by fresh traffic related particles i.e. fresh nucleation mode particles and soot particles. Thus, Aitken and accumulation modes, which are characteristics for urban aerosol population, are not well distinguishable in our distributions.

As shown in Table 3, the average geometric mean diameter (GMD) of traffic related nucleation mode (Mode 1) is 20.3±2.7 nm during the summer and 18.9±1.8 nm during the winter. Imhof et al. (2005) measured particle size distributions in two different road tunnels in Graz and Liverpool. They found nucleation mode with GMDs around 15–20 nm in Graz and 25 nm in Liverpool and geometric standard deviations (GSD) approximately 1.8 and 1.5, respectively. Ketzel et al. (2004) found out that the traffic related distributions peaked at 22 nm in the center of Copenhagen. In addition, Wehner and Wiedensohler (2003) reported the peak in the urban number size distribution around 20 nm. They also found the additional mode peaking at 10–15 nm during the summer time. This mode was related to the new particle formation event which correlated with the amount of global radiation. During our measurement campaign we observed only two similar formation event days during summer campaign 2004. This data was omitted from results. The found values for the Mode 1 correspond to values for nucleation mode particles of single vehicle emissions. The GMD values for nucleation mode emitted by single vehicle is usually around 10–20 nm (e.g. Kittelson et al., 2004).

The GMD of Mode 2 is 72.0±14.3 nm and 75.1±14.6 nm in summer and winter time respectively. Imhof et al. (2005) measured the road tunnel soot distributions peaking at around 80–100 nm (GSD~1.85) in tunnel in Graz. On the other hand, they also reported that in tunnel in Liverpool, the soot mode was not clearly distinguishable and its GMD was around 45 nm (GSD~2.22). Rose et al. (2005) studied the soot particle distribution in street canyon in Leipzig. They observed the soot distribution peaking at 65 nm and 70 nm during summer and winter. They also found out that the soot particles consists of 50–60% of concentration of 80 nm particles at the road side. In urban background the corresponding percentile is 20–25%. In addition, the GMD values for soot mode of singe vehicle emissions are reported to be typically 50–90 nm (e.g. Harris and Maricq, 2001). Thus according to earlier studies, it can be assumed that the GMD of Mode 2 is determined by the traffic related soot particles.

In Fig. 7, the GMD values of fitted distributions are shown as a function of traffic rate. The GMD of nucleation mode and accumulation mode seems to be rather independent on traffic rate. At traffic rate values <500 vehicles/hour (i.e. night time measurements), the GMD of both modes seems to increase. This is caused by diminishing portion of traffic related particles in particle population. At this case the urban background i.e. Aitken and accumulation modes become dominant modes in measured distribution. In fact, the fitted GMD values at low traffic rates are 30–40 nm and 80–140 nm which corresponds GMDs of Aitken and Accumulation mode measured both at urban and rural background stations (e.g. Tunved et al., 2003; Hussein et al., 2004).

4.3. Particle density

In Table 3, also the resulted density values corresponding to these two modes are shown. The density of “Mode 1”, i.e. nucleation mode (GMD 10–25 nm), is 1.04±0.14 g/cm^3 and 0.96±0.07 g/cm^3 during the summer and winter time, respectively. There is no published data for density of traffic related nucleation mode particles. Sakurai et al. (2003a) analyzed the composition of nucleation particles emitted from heavy duty diesel engine without any aftertreatment systems. They found out that the particles consist of organic compounds with carbon number 24–32. Sakurai et al. (2003b) estimated density 0.8 g/cm^3 for these compounds. In addition, the nucleation mode formation is connected to the sulphate formation, especially when oxidation catalyst is used (Lepperhof, 2001; Maricq et al., 2002; Vaaraslahti et al., 2004). According to studies of Vogt et al. (2003) and Gieshaskiel et al. (2005) the sulphuric acid – water nucleation seems to have important role in nucleation mode formation. This means that the traffic related nucleation mode particles may include also water and sulphuric acid with densities 1 g/cm^3 and 1.8 g/cm^3, respectively.

The found effective density value for "Mode 2" (GMD 60–80 nm) was 1.45±0.10 g/cm^3 and 1.87±0.30 g/cm^3 for summer and winter time, respectively. The results are in good agreement with the study of McMurry et al. (2002). They found that the density of ~0.1 μm urban aerosol particles measured in Atlanta, USA, varied between ~1.4–1.7 g/cm^3. The density values found in this and previous studies are high compared to the reported values for soot particle densities. The material density of soot is close to value 2 g/cm^3 but soot particles are agglomerates having lower effective density values due to the porosity of particles. According to laboratory studies of Park et al. (2003), Virtanen et al. (2004) and Maricq et al. (2004) the density of diesel soot particles in the soot mode is close to 1 g/cm^3 in the case of particles having size ~60 nm and below ~0.6 g/cm^3 for the ~100 nm sized particles. If the voids in the agglomerated particles get filled with condensable materials while the particle mobility size remains unchanged, the particle density grows. Still it is unlikely that the condensation of volatiles could increase the density of soot particles in "Mode 2" up to 1.4–1.8 g/cm^3. More probable is that "Mode 2" consists of soot particles and urban background particles of other materials. The found density for "Mode 2" is the average density for the externaly mixed aerosol. In fact the relatively low increase of concentration of particles larger than 63 nm with increasing traffic rate (see Fig. 4 and Table 2) and the results of Rose et al. (2005) discussed above supports the assumption, that a significant portion of particles in "Mode 2" are not fresh vehicle emitted particles.

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5. Conclusions

The number concentration and size distribution of traffic related particles were measured at road-side in Helsinki in winter and summer time. The measurements were performed simultaneously at distances of 9 m and 65 m from the highway. The total concentration was 2–3 times higher in the winter time than in the summer time and it was dominated by nucleation mode particles. The $dp<63$ nm particles (at aerodynamic size) constitute ~90% of particles in winter time and ~80% of particles in summer time. The total number concentration of particles increases with increasing traffic rate The dependence of $dp<63$ nm particles on traffic rate is stronger than for $dp>63$ nm particles both during the summer and winter time.

The particle size distribution at the roadside consists of two distinguishable size modes. The GMD of nucleation mode (Mode 1) was 20.3 nm in summer and 18.9 nm in winter. The GMD of the larger mode (Mode 2) was 72.0 nm in summer and 75.1 nm in winter. These values correspond to values for nucleation mode and soot mode particles of single vehicle emissions. The average density value for Mode 1 particles was 1.0 g/cm^3 both in summer and winter time while the average density value for Mode 2 was 1.5±0.1 g/cm^3 and 1.8±0.3 g/cm^3 for summer and winter time, respectively. According to authors' knowledge density values for urban, traffic related nucleation mode particles have not been previously published.

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References

Abdul-Khalek, I. S., Kittelson, D. B., and Brear, F.: The influence of dilution conditions on diesel exhaust particle size distribution measurements, SAE technical paper series, no. 1999-01-1142, 1999.

Burón, J. M., López, J. M., Aparicio, F., Martín, M. A., and García, F.: Estimation of road transportation emissions in Spain from 1988-1999 using COPERT III program, Atmos. Environ., 38, 715–724, 2004.

Charron, A. and Harrison, R. M.: Primary particle formation from vehicle emissions during exhaust dilution in the roadside atmosphere, Atmos. Environ., 37, 4109–4119, 2003.

Gieshaskiel, B., Ntziachristos, L., Samaras, Z., Scheer, V., Casati, R., and Vogt, R.: Formation potential of vehicle exhaust nucleation mode particles on-road and in the laboratory, Atmos. Environ., 39, 3191–3198, 2005.

Harris, S. J. and Maricq, M. M.: Signature size distribution for diesel and gasoline engine exhaust particulate matter, J. Aerosol Sci., 32, 749–764, 2001.

Hitchins, J., Morawska, L., Wolff, R., and Gilbert, D.: Concentrations of submicrometre particles from vehicle emissions near a major road, Atmos. Environ., 34, 51–59, 2000.

Hussein, T., Puustinen, A., Aalto, P. P., Mäkelä, J. M., Hämeri, K., and Kulmala, M.: Urban aerosol number size distributions, Atmos. Chem. Phys., 4, 391–411, 2004, SRef-ID: 1680-7324/acp/2004-4-391.

Imhof, D., Weingartner, E., Prévöt, A. S. H., Ordóñez, C., Kurtenbach, R., Wiesen, P., Rodler, J., Sturm, P., McCrae, I., Sjödin, Å., and Baltensberger, U.: Aerosol and NO_x emission factors and submicron particle number size distributions in two road tunnels with different traffic regimes, Atmos. Chem. Phys. Discuss., 5, 5127–5166, 2005, SRef-ID: 1680-7375/acpd/2005-5-5127.

Janhäll, S., Jonsson, Å. M., Molnár, P., Svensson, E., and Hallquist, M.: Size resolved traffic emission factors of submicrometer particles, Atmos. Environ., 38, 4331–4340, 2004.

Ketzel, M., Wåhlin, P., Kristensson, A., Swietlicki, E., Berkowicz, R., Nielsen, O. J., and Palmgren, F.: Particle size distribution and particle mass measurements at urban, near-city and rural level in the Copenhagen area and Southern Sweden, Atmos. Chem. Phys., 4, 281–292, 2004, SRef-ID: 1680-7324/acp/2004-4-281.

Khalek, I. A., Kittelson, D. B., and Brear, F.: Nanoparticle growth during dilution and cooling of diesel exhaust: experimental investigation and theoretical assessment, SAE technical paper series, no. 2000-01-0515, 2000.

Kittelson, D. B.: Engines and nanoparticles: a review, J. Aerosol Sci., 29, 575–588, 1998.

Kittelson, D., Johnson, J., Watts, W., Wei, Q., Drayton, M., Paulsen, D., and Bukowiecki, N.: Diesel aerosol sampling in the atmosphere, SAE technical paper series, no. 2000-01-2212, 2000.

Kittelson, D. B., Watts, W. F., and Johnson, J. P.: Nanoparticle emissions on Minnesota highways, Atmos. Environ., 38, 9–19, 2004.

Kristensson, A., Johansson, C., Westerholm, R., Swietlicki., E., Gidhagen, L., Wideqvist, U., and Vesely, V.: Real-world traffic emissions of gases and particles measured in a road tunnel in Stockholm, Sweden, Atmos. Environ., 38, 657–673, 2004.

Lepperhof, G.: Influences on the particle size distribution of diesel particulate emissions, Topics in Catalysis, vols. 16/17, nos. 1–4, 2001., 2001.

Longley, I. D., Gallagher, M. W., Dorsey, J. R., and Flynn, M.: A case-study of fine particle concentrations and fluxes measured ina busy street canyon in Manchester, UK, Atmos. Environ., 38, 3595–3603, 2004.

Maricq, M., Chase, R., Xu, N., and Laing, P.: The effects of catalytic converter and fuel sulfur level on motor vehicle particle emissions: Light duty vehicle emissions, Environ. Sci. Technol., 36, 283–289, 2002.

Maricq, M. and Xu, N.: The effective density and fractal dimension of soot particles from premixed flames and motor vehicle exhaust, J. Aerosol Sci., 35, 1251–1274, 2004.

McMurry, P., Wang, X., Park, K., and Ehara, K.: The relationship between mass and mobility for atmospheric particles: A new technique for measuring particle density, Aerosol Sci. Technol., 36, 227–238, 2002.

Molnár, P., Janhäll, S., and Hallquist, M.: Roadside measurements of fine and ultrafine particles at a major road north of Gothenburg, Atmos. Environ., 36, 4115–4123, 2002.

Morawska, L., Thomas, S., Gilbert, D., Greenaway, C., and Rijnders, E.: A study of the gorisontal and vertical profile of submicrometer particles in relation to a busy road, Atmos. Environ., 33, 1261–1274, 1999.

Park, K., Cao, F., Kittelson, D., and McMurry, P.: Relationship between aprticle mass and mobility for diesel exhaust particles, Environ. Sci. Technol., 37, 577–583, 2003.

Pirjola, L., Parviainen, H., Hussein, T., Valli, A., Hämeri, K., Aalto, P., Virtanen, A., Keskinen, J., Pakkanen, T. A., Mäkelä, T., and Hillamo, R. E.: "Sniffer" – a novel tool for chasing vehicles and measuring traffic pollutants, Atmos. Environ., 38, 3625–3635, 2004.

Pirjola, L., Paasonen, P., Pfeiffer, D., Hussein, T., Hämeri, K., Koskentalo, T., Virtanen, A., Rönkkö, T., Keskinen, J., and Pakkanen, T.: Dispersion of particles and trace gases nearby a city highway: mobile laboratory measurements in Finland, Atmos. Environ., in press, 2005.

Ristimäki, J., Virtanen, A., Marjamäki, M., Rostedt, A., and Keskinen J.: On-line measurement of size distribution and effective density of submicron aerosol particles, J. Aerosol Sci., 33, 1541–1557, 2002.

Ristimäki, J., Keskinen, J., Virtanen A., Maricq, M., and Aakko, P.: Cold temperature PM emissions measurement: Method evaluation and application to light duty vehicles, Environ. Sci. Technol., 39, 9424–9430, 2005.

Rose, D., Wehner, B., Ketzel, M., Engler, C., Voigtländer, J., Tuch, T., and Wiedensohler, A.: Atmospheric number size distributions of soot particles and estimation of emission factors, Atmos. Chem. Phys. Discuss., 5, 10 125–10 154, 2005, SRef-ID: 1680-7375/acpd/2005-5-10125.

Sakurai., H., Tobias, H., Park, K., Zarling, D., Doherty, K. S., Kittelson, D. B., McMurry, P. H., and Ziemann, P.: On-line determination of diesel nanoparticle composition and volatility, Atmos. Environ., 37, 1199–1210, 2003a.

Sakurai, H., Park, K., McMurry, P., Zarling, D. D., Kittelson, D. B., and Ziemann, P.: Size-Dependent mixing characteristics of volatile and nonvolatile components in diesel exhaust aerosols, Environ. Sci. Technol., 37, 5487–5495, 2003b.

Stein, S. W., Turpin, B. J., Cai, X. P., Huang, P. F., and McMurry, P. H.: Measurements of relative humidity-dependent bounce and density for atmospheric particles using dantem-DMA-impactor technique, Atmos. Environ., 28, 1739–1746, 1994.

Sturm, P. J., Baltensberger, U., Bacher, M., Lechner, B., Hausberger, S., Heiden, B., Imhof, D., Weingartner, E., Prevot, A. S. H., Kurtenbach, R., and Wiesen, P.: Roadside measurements of particulate matter size distribution, Atmos. Environ., 37, 5273–5281, 2003.

Tunved, P., Hansson, H.-C., Kulmala, M., Aalto, P., Viisanen, Y., Karlsson, H., Kristensson, A., Swietlicki, E., Dal Maso, M., and Komppula, M.: One year boundary layer aerosol size distribution data from five nordic background stations, Atmos. Chem. Phys., 3, 2183–2205, 2003, SRef-ID: 1680-7324/acp/2003-3-2183.

Weijers, E. P., Khlystov, A. Y., Kos, G. P. A., and Erisman, J. W.: Variability of particulate matter concentrations along roads and motorways determined by a moving measurement unit, Atmos. Environ., 38, 2993–3002, 2004.

Wehner, B., Birmili, W., Gnauk, T., and Wiedensohler, A.: Particle number size distribution in a street canyon and their transformation into the urban air background: measurements and a simple model study, Atmos. Environ., 36, 2215–2223, 2002.

Wehner, B. and Wiedensohler, A.: Long term measurements of submicron urban aerosols:

statistical analysis for correlations with meteorological conditions and trace gases, Atmos. Chem. Phys., 3, 867–879, 2003, SRef-ID: 1680-7324/acp/2003-3-867.

Williams, P. I., Gallagher, M. W., Choularton, T. W., Coe, H., Bower, K. N., and McFiggans, G.: Aerosol development and interaction in an urban plume, Aerosol Sci. Technol., 32, 120–126, 2000.

Wåhlin, P., Palmgren, F., and Van Dingenen, R.: Experimental studies of ultrafine particles in streets and the relationship to traffic, Atmos. Environ., 35, 63–69, 2001.

Vaaraslahti, K., Virtanen, A., Ristimäki, J., and Keskinen, J.: Nucleation mode formation in heavy duty diesel exhaust with and without particulate filter, Environ. Sci. Technol., 38, 4884–4890, 2004.

Van Dingenen, R., Raes, F., Putaud, J.-P., Baltensberger, U., Charron, A., Facchini, M.-C., Decesari, S., Fuzzi, S., Gehrig, R., Hansson, H.-C., Harrison, R. M., Hüglin, R., Jones, A. M., Laj, P., Lorbeer, G., Maenhaut, W., Palmgren, F., Querol, X., Rodriquez, S., Schneider, J., ten Brink, H., Tunved, P., Torseth, K., Wehner, B., Weingartner, E., Wiedensohler, A., and Wåhlin, P.: A European aerosol phenomenology – 1: physical characteristics of particulate matter at kerbside, urban, rural and background areas in Europe, Atmos. Envrion., 28, 2561–2577, 2004.

Virtanen, A., Ristimäki, J., and Keskinen, J.: New method to define the effective density and fractal dimension of agglomerate particles, Aerosol Sci. Technol., 38, 437–446, 2004.

Virtanen, A., Ritimäki, J., Vaaraslahti, K., and Keskinen, J.: Effect of engine load on diesel soot particles, Environ. Sci. Technol., 38, 2551–2556, 2004.

Vogt, R., Sheer, V., Casati, R., and Benter, T.: On-road measurement of particle emission in the exhaust plume of a diesel passenger car, Environ. Sci. Technol., 37, 4070–4076, 2003.

Table 1. The maximum, minimum and average values of temperature and relative humidity during the winter and summer campaigns.

	T (˚C)		average		*RH* (%)		average	
	max	min	all	day	max	min	all	day
Summer 03	22.0	7.0	15.7	16.4	98	43	79.0	76.1
Summer 04	22.0	7.0	15.4	16.0	98	42	78.7	74.7
Winter 03	5.1	−15.2	−3.3	−2.8	98	51	85.6	84.0
Winter 04	3	−17.8	−4.9	−4.9	98	55	86.9	86.3

Table 2. The "a" values of the function $y{\sim}x^a$, where y is particle concentration and x is traffic flow. The "a" and R^2 values for total concentration and concentration of two size fractions for summer and winter time data are presented in the table.

$y{\sim}x^a$		a	R^2
Total concentration	summer	0.62	0.70
	winter	0.69	0.81
3 nm≤dp<63 nm	summer	0.60	0.63
	winter	0.71	0.83
63 nm≤dp<6.6 μm	summer	0.49	0.60
	winter	0.50	0.60

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Table 3. The day time (06:00–20:00) summer and winter average values of distribution characteristics (geometric mean diameter GMD, geometric standard deviation GSD and number concentration N) and average density (ρ) of particles in the modes.

	Mode 1				Mode 2			
	GMD *(nm)*	GSD	N *(#/cm^3)*	ρ (g/cm^3)	GMD *(nm)*	GSD	N *(#/cm^3)*	ρ (g/cm^3)
Summer								
Average	20.3	1.7	18 960	1.0	72.0	1.8	13 750	1.5
Stdev (%)	13.5	6.2	77.3	13.3	19.8	9.6	56.9	6.6
Winter								
Average	18.9	1.7	61 310	1.0	75.1	1.6	6810	1.8
Stdev (%)	9.4	4.9	44.7	7.2	19.5	12.6	72.6	16.2

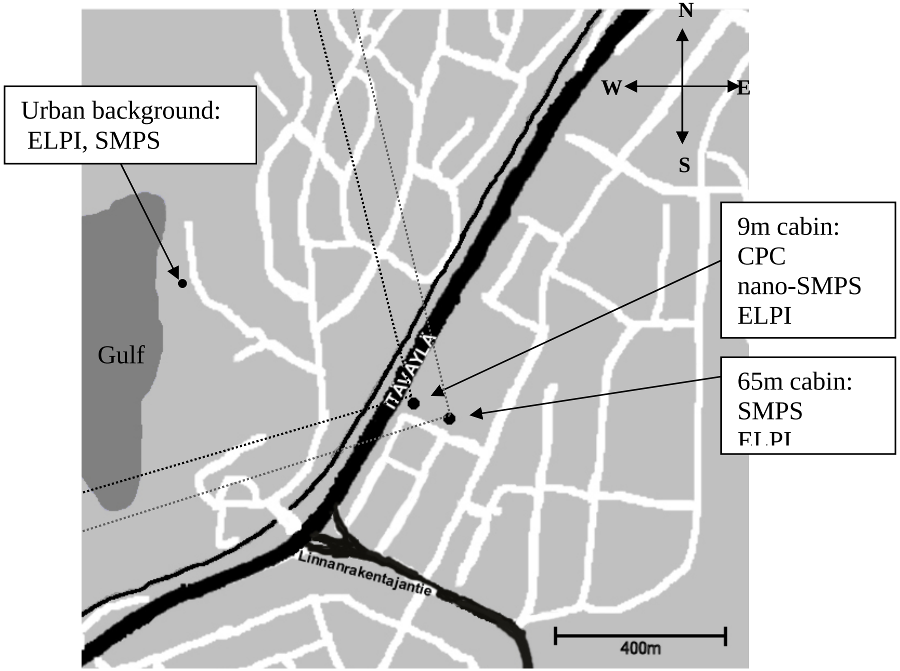

Fig. 1. Map of the measurement site. Studied wind sectors (255°–345°) marked with dashed lines.

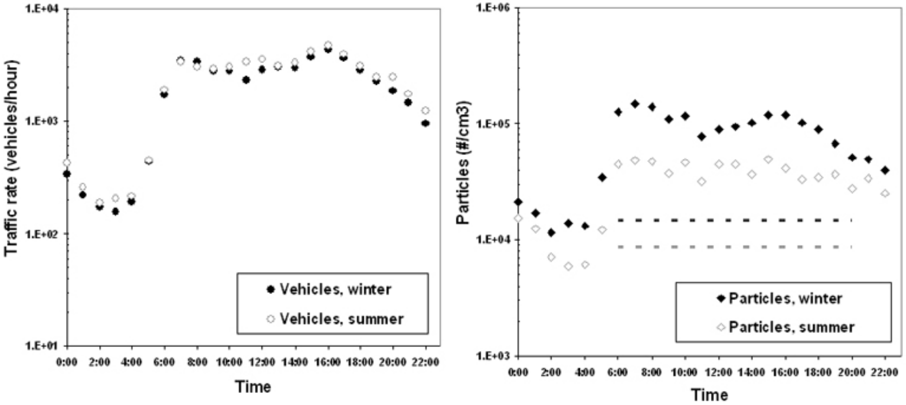

Fig. 2. **(a)** The traffic rate and **(b)** the measured particle concentration at summer and winter time. Dashed lines in (b) represent the day time averages for urban background (black line for winter and light line for summer).

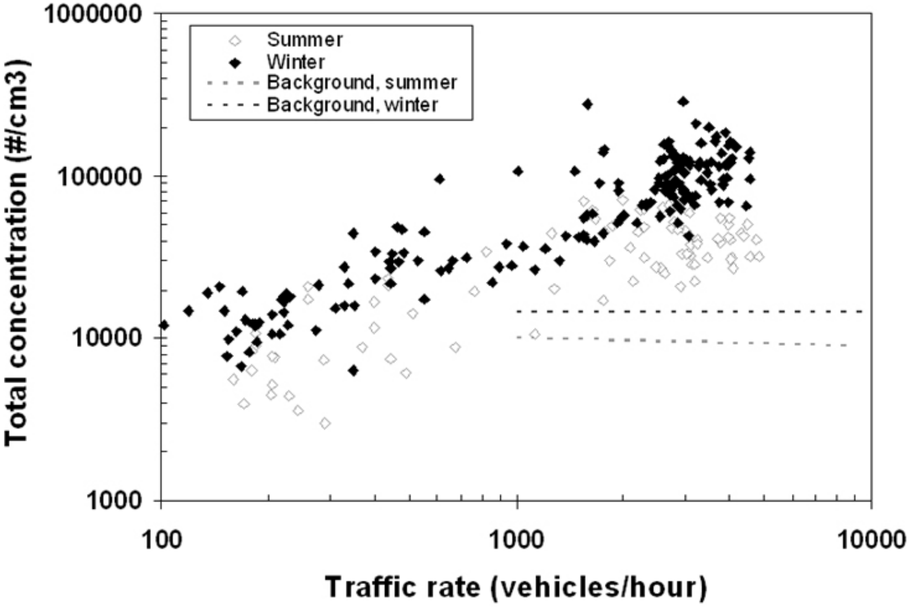

Fig. 3. The particle concentrations as a function of traffic rate.

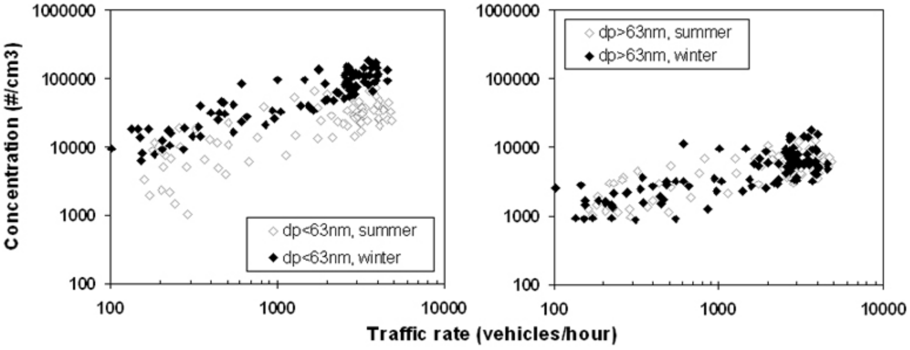

Fig. 4. Concentrations of two different size fractions as a function of traffic rate: **(a)** particles smaller than 63 nm **(b)** particles larger than 63 nm.

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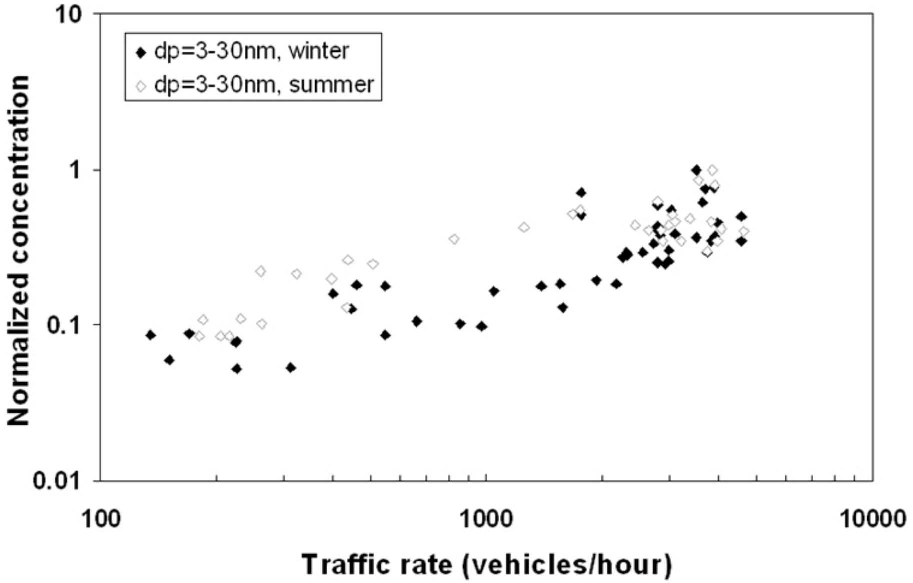

Fig. 5. The dependence of nucleation mode particle (dp=3–30 nm) concentration on traffic rate.

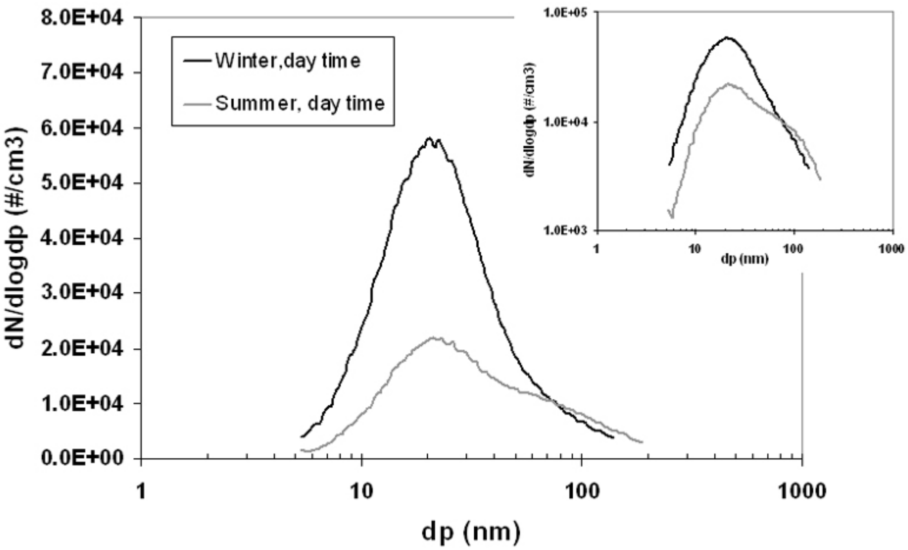

Fig. 6. Typical SMPS size distributions measured at winter (black line) and summer (light line). Distributions in log-log scale are presented in the upper corner.

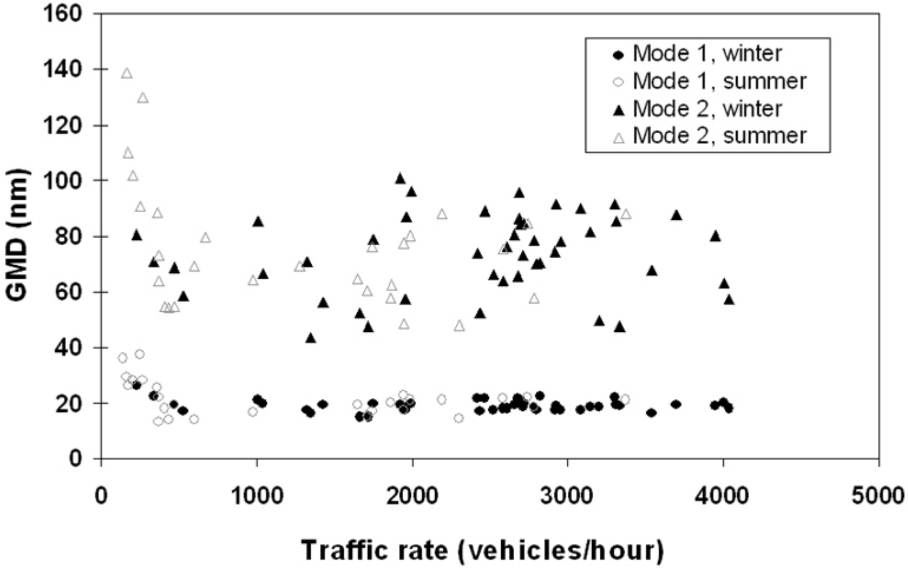

Fig. 7. Geometric mean diameters of "Mode 1" and "Mode 2" as a function of traffic rate.