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Roadside measurements of particulate matter size distribution

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Abstract

Roadside measurements were performed in order to document the size distribution of particulate matter (PM) under dilution conditions similar to those found in real world. These activities covered measurements at engine test beds, at different locations in a road tunnel as well as in an urban environment. In order to get a clear picture of the evolution of the PM in different size classes, the in-tunnel locations ranged from curb-side to different locations inside the exhaust air system. Additional measurements were performed in the ambient air at curb-side at a street crossing as well as in urban background.

At those times when heavy traffic occurs, tunnel measurements show size distributions similar to those derived from engine/vehicle measurements. During times with little traffic the size distributions are closer to those recorded in ambient air. As soon as the traffic load increases the size distribution changes, due to rapid coagulation of the smallest particles with the accumulation mode. As the travel time of the particles through the tunnel and up the stacks is very long "aging" effects could be observed. Thus, these spectra are quite different from ambient measurements in urban air, especially in the region above 30–40 nm.

Keywords: Roadside measurement; Particle-size distribution; Particulate matter; Particulate emissions; Tunnel measurement

1. Introduction

An increasing number of studies indicate that particulate matter (PM) air pollution can have a severe effect on human health. These findings have led to discussions on the introduction of standards for size-specific particulate mass concentrations. Especially (PM) smaller than 10 μm (PM 10—coarse particles) or less than 2.5 μm (PM 2.5—fine particles) has to be considered. There are strong indications that the number of particles may be more relevant for human health effects than the particle mass (Donaldson et al., 1998). In ambient air the maximum of the number size distribution is typically found in the range between 10 and 100 nm, depending on the measurement location and time of day. However, these particles contribute very little to the total suspended mass and thus are not taken into account by emission or air quality standards related to particle mass only, even though they may have severe health effects.

This article will concentrate on PM emissions from on-road vehicles. Although the technology of measuring ultra-fine particles has improved within the last few years, there are very little data available on production

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rate, airborne concentrations and the time development of the particle-size distribution. On top of that, up to now no standardized method for PM size distribution measurements for vehicle exhaust emissions has been developed. It is well known that sampling conditions strongly influence the shape of the size distribution and the occurrence of nucleation mode PM (Kittelson, 1999).

In order to gain an insight into the genesis of PM size distribution, aerosol measurements were performed at engine test beds, at different locations in road tunnels and at different ambient locations. The main focus of these measurements was the characterization of the particle-size distribution of vehicle emissions and the time development of this particle-size distribution in different environments.

2. Instrumentation

2.1. Engine measurements

The measurements at the engine test bed were performed using a measurement protocol developed under the framework of the EU-FP5 research program PARTICULATES (http://vergina.eng.auth.gr/mech/lat/particulates/). The measurements were performed on the basis of applying a constant dilution ratio at all measurement points (engine loads) during stationary and non-stationary tests. A scanning mobility particle sizer (SMPS 3080, CNC 3010, TSI) and a dual dynamic particle mobility sizer (DDMPS, Reischl) were used.

2.2. Tunnel and ambient air measurements

The ambient PM size distribution measurements were undertaken, employing SMPS instruments. In the tunnel measurements, four SMPS systems (TSI with a DMA 3071A, CNC 3022A; DMA 3080 CNC 3010) were used. The flow rate was set to 0.3 l/min, the up scan time was 220 s and the size range was 10–700 nm. Calibration of all SMPS was performed before and after the field measurements. All spectra were corrected by taking one SMPS as the reference and applying correction factors for the other SMPS based on this calibration. No correction for diffusion losses in the sampling line and the DMA was applied.

The measurements in the urban atmosphere were performed with one single SMPS (TSI with a DMA 3080 and a CNC 3010); the size range was set to 7–300 nm.

3. Engine measurements

Particle formation is a function of engine load, engine technology, fuel quality and unfortunately also of sampling conditions. In the framework of the above-mentioned PARTICULATES project, attempts to define sampling conditions were undertaken with the aim of measuring real-world size distributions at the test facilities. Therefore, extensive investigations were made in various labs in Europe in order to come up with a uniform measurement procedure. If one takes as example a widely used heavy duty diesel engine (310 kW, EURO 3, 12000 cm^3) measured at our lab (Institute for Internal Combustion Engines and Thermodynamics, Graz University of Technology) according to the uniform procedure, size distributions similar to those shown in Fig. 1 are found. In the case of low engine loads (idling and 10% load) a clear peak in the nucleation mode range (<30 nm) is visible, followed by a decrease of number concentrations in the ranges of bigger size classes. The shapes of the curves change with higher engine loads. Already at 25% engine load a second peak in the soot mode range (80–100 nm) appears, becoming dominant at 50% engine load. As the measurement procedure requires a constant dilution ratio of 1:11, the dilution air flow was permanently adjusted to the engine exhaust air flow in order to minimize any influence of varying dilution ratios.

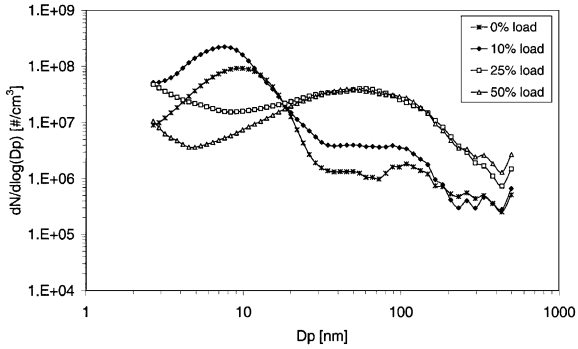

Fig. 1. PM size distribution at different engine loads (EURO 3 HDV engine).

4. Road tunnel measurements

Tunnel measurements have the advantage that the road tunnel can act as a large dilution tunnel with well-known boundary conditions, as air flow and traffic volume are well known. In addition, a broad variety of emission sources contribute to the pollution and thus provide a real-world mixture of different vehicle sources. The suitability of road tunnels for determining real-world emissions for road vehicles has been proven in multiple studies (e.g., Gertler et al., 2001; John et al., 1999; Sturm et al., 2001). These studies were mostly concerned with emissions from gaseous pollutants. Only

a few also considered PM emissions (Weingartner et al., 1997; Ulevicius and Mordas, 2002; Abu-Allaban et al., 2002; Sternbeck et al., 2002). The disadvantage of road tunnel measurements is that clear attribution to a specific source is not possible, since tailpipe emissions are not the only PM emission sources in a road tunnel atmosphere.

4.1. Measurement location

Measurements of the PM size distribution in the range below 700 nm were performed in a road tunnel in Austria, (Plabutsch tunnel) for a period of 10 days. The tunnel has a length of 10 km and is operated in counter flow in a single bore. A transverse ventilation system is installed to provide the necessary fresh air supply in the tunnel. The ventilation sections have a length of 2 km each. Two vertical stacks are used for discharging the exhaust air into the atmosphere and providing fresh air. The stacks have a height of 90 and 240 m. This complex ventilation system allows the evolution of the aerosol size distribution over the time period between the emission event and when the aerosol leaves the stack to be investigated.

A measurement program was set up which was based on size-distribution measurements at the following locations (Fig. 2):

- Measurement of the fresh (intake) air (site 1).
- Curb-side measurement inside the tunnel (site 2).
- Measurement inside the ventilation duct, at the foot of the 240 m high stack (site 3).
- Measurement at the top of the 240 m high stack, ventilation outlet (site 4).

Site 1 covers the situation for fresh air. The fresh air inlet is at an altitude of 640 m above sea level in a forest on top of a hill. The intake air exhibits typical background concentration levels. However, the shape of the size distribution might be influenced by the PM from the exhaust air stack located some 50 m above the inlet. In other words, the PM size distribution of the fresh air is similar to that of the exhaust air, but the number concentrations are much smaller.

Site 2 (Fig. 2) is located directly beside the curb inside the tunnel. As the tunnel ventilation is fully transverse, fresh air enters the traffic area at the top left corner through the false ceiling. In the traffic area it is well mixed with the exhaust due to the traffic-induced turbulence. Eventually it leaves the traffic area through openings in the false ceiling and through the exhaust air duct. Hence, the measurement covers almost only "fresh" PM emission from traffic as the longitudinal air velocity in the tunnel is relatively small (especially in the case where the counter flow restricts air flow in one direction of the tunnel).

Site 3 is quite different as it is located inside the exhaust air duct. Here, all the air from a 2 km long ventilation section comes together before it enters the vertical stack. At this point the individual particles have already experienced different traveling times. Those coming from the far end of the exhaust air duct have been traveling for almost 2 km while those from the near end have just entered the duct from the traffic area below. The travel times (and thus the residence times) of the various particles vary between 200 s (far end) and a few seconds (near end) in case of full ventilation power. During times with little traffic and hence reduced ventilation rate (e.g., during night) the travel times are

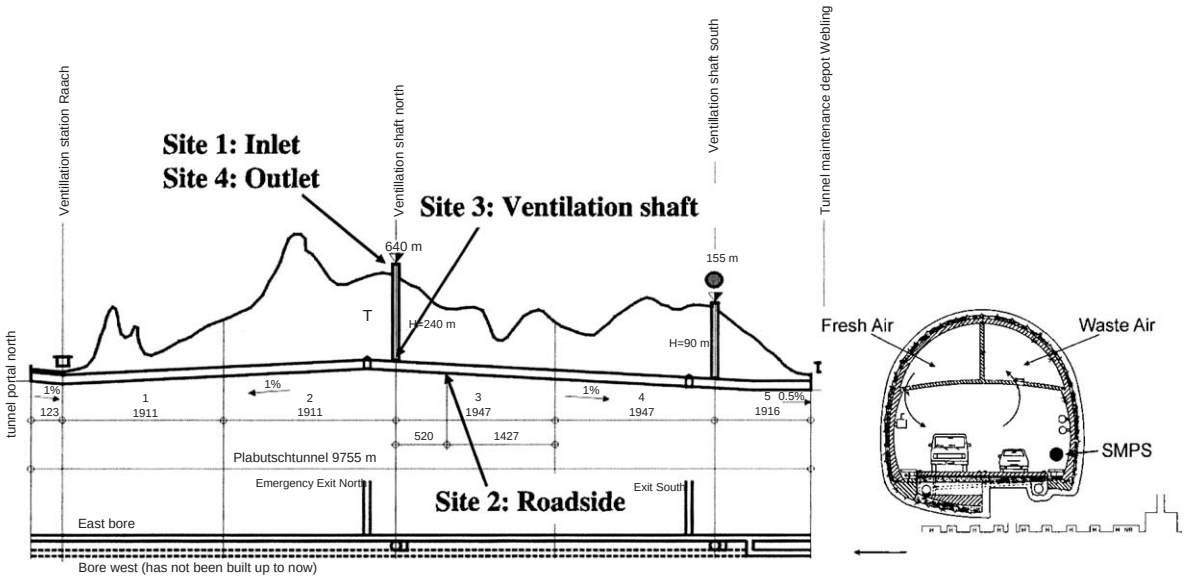

Fig. 2. Measurement locations, Plabutschtunnel.

much longer. This has a clear influence on the size distribution.

Site 4 is at the top of the exhaust air stack. Compared with site 3, no further dilution or mixing with other air takes place. The travel time between sites 3 and 4 is some 40 s during peak hours (full ventilation rate) and longer during off-peak hours.

4.2. Traffic variability

As the tunnel serves as bypass for the city of Graz, it has a very distinct traffic variation during the course of the day. On an average working day within the field campaign some 24 070 vehicles passed through the tunnel, and during the weekend the traffic volume went down to 16 930 vehicles/day. The heavy duty vehicle (HDV) share was 23% on a working day and 7.7% on a weekend day. Fig. 3 shows the traffic variations for the northbound and southbound directions. It should be noted that Sunday, 4th November was the end of a short holiday period (1st to 4th November) and a lot of transit traffic from the Balkan region to Germany passed through the tunnel. This can be seen in the very high amount of northbound Sunday traffic, while the southbound traffic was almost the same as on the Sunday the week after. Friday southbound traffic traditionally has a peak (also due to the transit of guest workers back to the Balkan region).

Passenger car (PC) traffic has a clear morning and evening peak, whereas HDV traffic has a strong increase during the morning, a flat distribution during the day and a decrease in the afternoon and evening hours. The reason for HDV peaks on Sunday evening is the lifting of the weekend HDV traffic ban at 22:00, which results in the departure of transit HDV parked in the region of Graz.

4.3. Size distributions

Averaging the measurement information over a longer period results, in general, in data with high confidence levels, as all the information available is considered. On the other hand, the time dependency of the collected information is lost.

Fig. 4 shows the median and the upper and lower quartile of the size distribution for the four sites, averaged over the whole sampling period. The curve for site 1 (inlet) peaks at 100 nm, the number concentration ($\#/cm^3$) is relatively low, as it is a rural background location. This distribution is most possibly influenced by

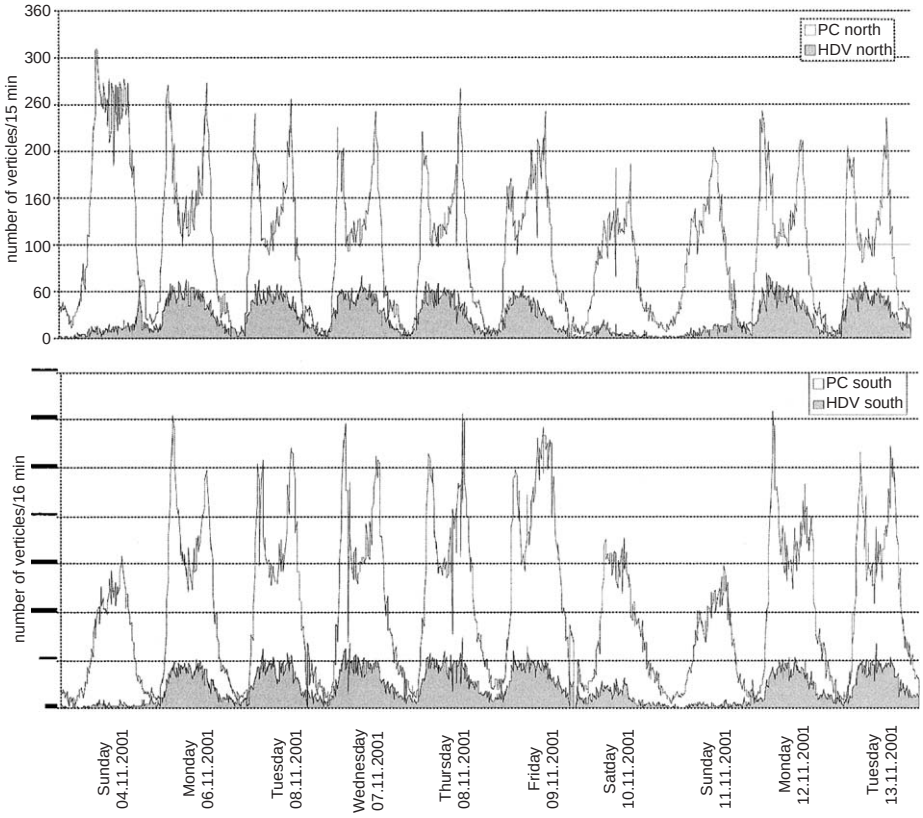

Fig. 3. Traffic frequency during the tunnel measurements.

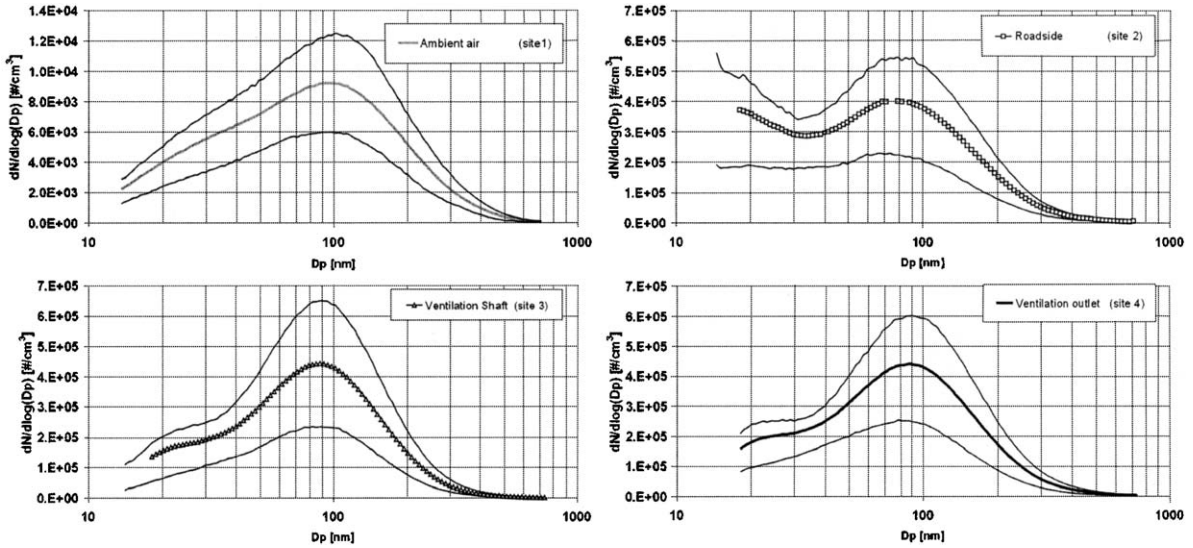

Fig. 4. Comparison of the spectra at the different tunnel measurement locations. The centre lines represents mean values, while the other two lines give 25 and 75 percentile values.

the air from the ventilation exhaust air outlet which shows a similar size distribution but—of course—much higher number concentrations (see results for site 4). At site 2 (roadside) a bimodal distribution having one peak in the very small fraction (<30 nm) and a second peak in the soot mode (~80 nm) is found. The averaged PM size-distribution at site 3 (foot of one of the exhaust stacks) shows again a bimodality with a major peak in the 80–100 nm range and a second much smaller one in the 30 nm range. A similar situation appears at the top of the ventilation shaft (site 4), some 240 m or at least 40 s later, although some coagulation of the ultra-fine aerosols can be expected.

However, as will be shown in the next section, the size distributions at all sites vary as a function of time, as the variation of traffic volume has a significant influence on the concentration of aerosols and the size distribution. As an example the situation concerning the time dependency of the size distributions at the curb-side location (site 2) and at the top of the exhaust stack (site 4) is shown in Fig. 5. At the curb, the big size classes dominate during the day (7:00, 13:00, 19:00) while these classes are much lower during night (little traffic), but still remarkably higher than the inlet concentrations. Bimodality is present at each time, but with a dominant soot mode during high-emission situations (daytime) and a dominant condensation particle mode in the 30 nm range during nighttime. This can be explained by lower production of ultra-fine particles during daytime, because the semi-volatile gases condense on the existing surfaces rather than forming new particles.

This clear time dependency is also shown for the stack top (site 4). Working hour traffic produces a dominant peak in the soot mode range (80 nm), while during the night a bimodal distribution with a peak in the small range and one in the soot mode appears. Thus, coagulation of particles is fast enough to decrease the concentrations of ultra-fine particles significantly between sites 2 and 4. It has to be mentioned that in the night the ventilation rate is much smaller and therefore the pollutants remain much longer in the tunnel compared to the daytime hours.

Fig. 5 also depicts the 25 and 75 percentile values for each of the curves. The curb-side measurements show in all cases a big spread in the region of the small size classes, while this is not the case in the regions above 40–50 nm. The spread in the small size class is biggest during nighttime where only a few emission events occur. The outlet site (site 4) does not show this behaviour anymore. The 25 and 75 percentile values follow much more the average line.

In a further step a principal component analysis (PCA) was performed on the data of the measurement campaign in the Plabutsch tunnel. The PCA tries to explain data as a combination of influencing factors (see Eq. (1)), i.e. the principal components (PC), and weighing factors (t), which describe the individual weight of each of these PCs.

$$F = \mathrm{PC}_1 * t_1 + \mathrm{PC}_2 * t_2 + \cdots + \mathrm{PC}_i * t_i. \quad (1)$$

The first PC (PC_1) represents a kind of averaged spectrum of the different measurement sites. The second PC (PC_2) describes the most important features of the

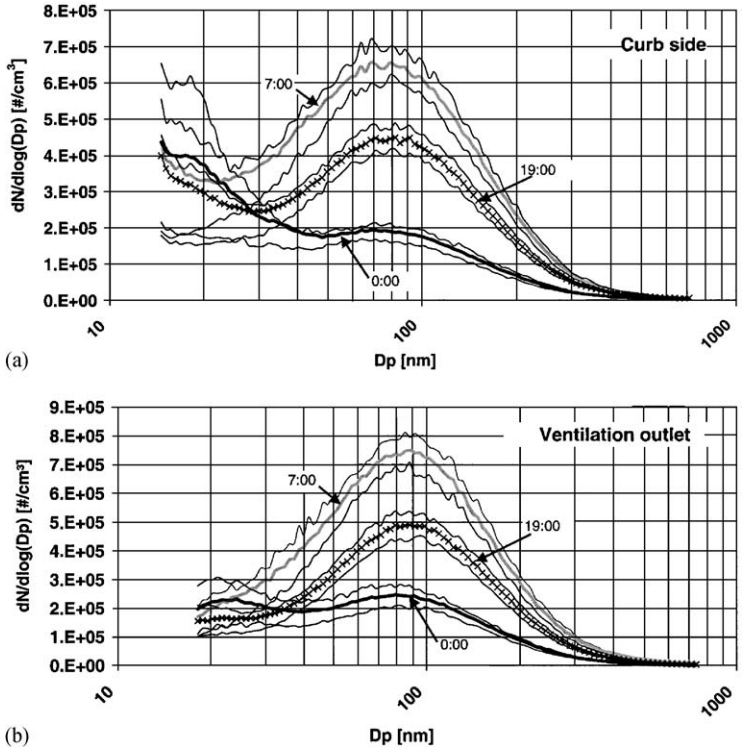

Fig. 5. Size distribution at curb-side (site 2) and ventilation stack outlet (site 4) as a function of time of day; concentrations in #/cm^3, diameter D_p in nm, mean, 25 and 75 percentile values.

measurement data, which cannot be described by the first PC. No higher-order PCs with significant input were found.

The first PCs of the measurement sites describe about 60% of the variance of the data of the different measurement sites. The shape of these first PCs is almost identical to the averaged size distributions shown in Fig. 4. The second PC is shown in Fig. 6 and describes 15% of the variance of the data. It can be seen that the spectra can be divided into two regions which are counterparts of each other. A rough interpretation of this function is: in case of an appearance of small particles the bigger ones are reduced and vice versa. Site 2 (roadside) shows a dominance of smaller PM, i.e., the size classes smaller than 35 nm have more weight compared to the spectra defined by the first PC. The situation is quite different for sites 2 and 3. For these two sites the PM size classes bigger than 50 nm are more pronounced than given with the first PC.

The second part of the PCA concerns the weighing factors. Fig. 7 shows these factors for site 4 (outlet). Factor one ($t1$) follows clearly the daily variation of the traffic—a morning and an evening peak. Factor 2 ($t2$) contains valuable information as well, a daily variation can be seen, at high concentrations (high values for $t1$) the second factor ($t2$) is positive. So, the negative peak at 24 nm in Fig. 6 is less pronounced. There is a different situation at nighttime when the factor of the second component is negative. The second PC is negative as well, which results in an increase of particles smaller than 50 nm and a decrease of particles larger than 50 nm. This clearly shows the influence of the varying parameters emission, dilution and residence time.

5. Measurements in urban atmosphere

In addition to the tunnel tests, measurements in urban atmosphere were undertaken at different locations and distances from road traffic emission sources. When moving to an urban atmosphere, the direct connection between road traffic emission source and concentration measurement is lost as dispersion takes place with an unknown dilution ratio. In addition, one has to consider that the background concentrations (from other sources) cannot be neglected and these may have an influence on chemical processes. Nevertheless, for curb-side measurements the background influences may not be too high, assuming that the wind direction is appropriate. A different situation appears as soon as the distance from the road increases. In such cases,

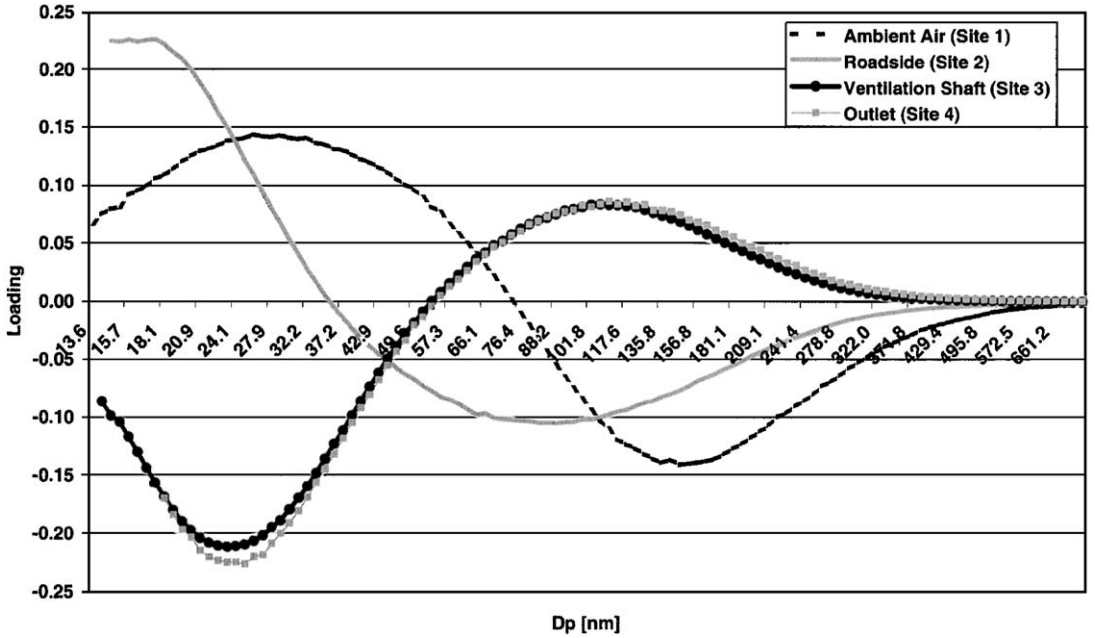

Fig. 6. Second principal components of the measurement sites.

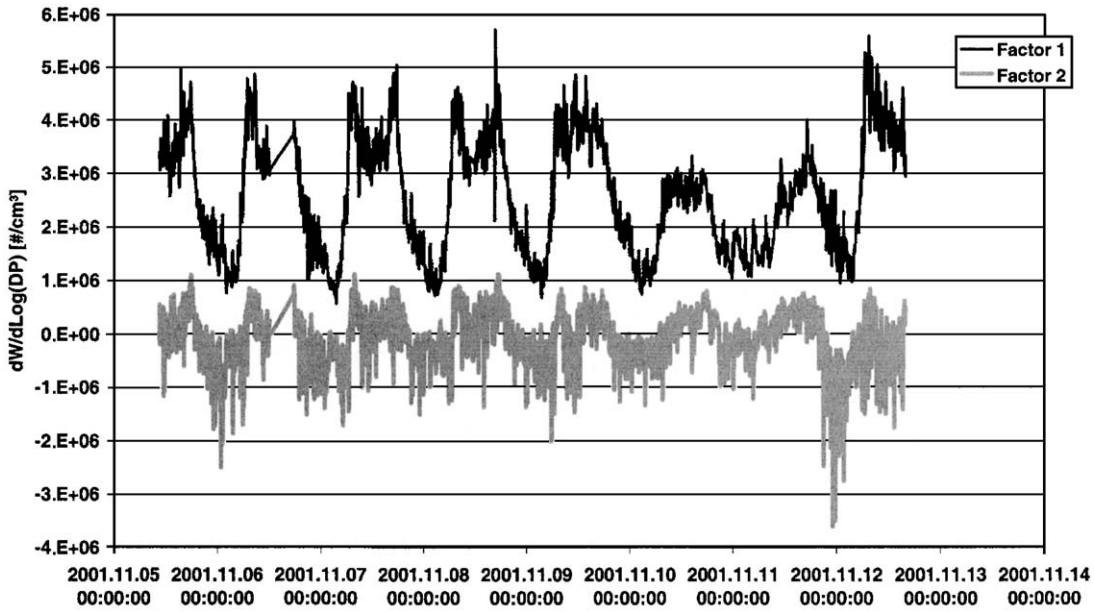

Fig. 7. First two factors of measurement site 3 (outlet).

dispersion, chemical reactions, etc., may play a dominant role.

5.1. Measurement locations

The curb-side measurements were performed at a street crossing (Don Bosco) in Graz, Austria. The measurements were performed in August 2001 and some 1500 spectra were recorded. The traffic volume was some 30 000 vehicles per day with a pronounced morning and evening rush hour peak. Although this is a typical roadside site, urban background air also has to be considered. During windward situations, where the crossing is in lee of the monitoring station, there is a relatively high PM10 load remaining.

Urban background measurements were undertaken at a residential location where no major roads exist. The nearest larger roads are some 600–800 m away. As the wind speeds were generally very low (mostly below 1 m/s) the direct influence of traffic sources was only relatively slight. The measurements were performed in September 2001 and some 2600 spectra were recorded.

5.2. Results

Data processing similar to that described in Section 4.3 was performed. The time-dependent results are given in the following section. Fig. 8 shows the size distribution for the roadside location. The distribution has a peak at around 25 nm. Compared to the tunnel measurements, much smaller concentrations in the 80–100 nm size range are visible. Fig. 8 also reveals the dependency on the traffic volume and the dispersion conditions. The maximum concentration is during the early morning peak where high emission levels meet unfavourable dispersion conditions.

The dominating part of the distribution in the urban background (see Fig. 8) is in the 30–40 nm range. The 80–100 nm range, which is present in the tunnel, is hidden. This situation prevails during almost the whole day.

6. Conclusions

The main focus was on carrying out investigations at different locations in order to provide information about the evolution of the aerosol size distribution as a function of travel time and dilution conditions. While in the engine measurements a constant dilution rate (1:11) with clean air is applied, the primary dilution process in the tunnel as well as in the ambient air involves a higher dilution ratio, but the dilution air contains aerosols with varying concentrations.

The investigations carried out so far can be summarized as follows:

- The curb-side measurements in the tunnel show an aerosol size distribution strongly varying with the traffic volume. Depending on the traffic and ventilation situation nucleation mode particles or soot mode particles dominate (Fig. 8).
- The roadside measurements at an intersection show a distribution having a peak at 25 nm, along with

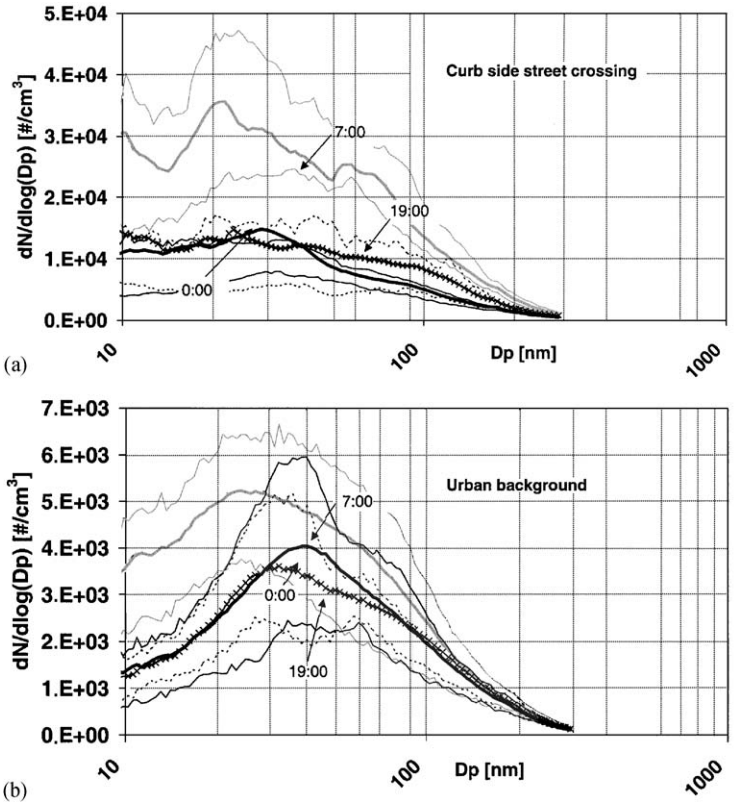

Fig. 8. Time-dependent and average size distribution at curb-side location (above) and in urban background (below), concentrations in #/cm^3, diameter D_p in nm.

smaller nucleation particles around 10 nm. The pure soot mode (80 nm range) which is present in the engine and the tunnel measurements is hardly visible in this case (Fig. 8). This is consistent with the findings in other cities (e.g., Baltensperger et al., 2002).

- Measurements in the urban background show that less particles of the nucleation mode are present in the urban background compared to the roadside, and the mode of the size distribution is shifted towards larger particles, due to coagulation and growth processes (Fig. 8).
- In street tunnels a nucleation mode was observed at the curb-side, but it is less pronounced than for urban measurements because nucleation is partly suppressed by the high existing surface area concentration. This also results in a much higher coagulation loss rate of the small particles compared to processes in the urban atmosphere.
- The PCA performed showed that most effects of the variations in the diurnal patterns of the size distributions can be captured by the first and second PCA component.

Tunnel measurements offer the possibility of investigating emissions from a huge variety of road vehicles. They are thus an optimal means for investigating real-world emission behaviour. However, in contrast to gaseous pollutants, the PM size concentration due to emissions is influenced by the size distribution that is already present in the tunnel. Thus, particle number size distribution profiles recorded in street tunnels do not necessarily represent situations occurring in ambient air.

Acknowledgements

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