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AEROSOL EMISSION IN A ROAD TUNNEL

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Abstract—Continuous measurements of aerosol emissions were performed within the scope of emission measurements in the Gubrist tunnel, a 3250 m long freeway tunnel near Zürich, Switzerland, from 20 September to 26 September 1993. The particles in the respirable size range ($d < 3\ \mu m$) were found to be mainly tail pipe emissions with very small amount of tire wear and road dust. The calculated PM_3 emission factor for diesel engines was about 310 mg km^{-1}, where the main part (63%) of the diesel vehicles were heavy-duty vehicles. Thirty-one percent of the PM_3 emissions from diesel vehicles were black carbon and 0.86% particle bound PAH. Due to the high fraction emitted by diesel engines the contribution of gasoline engines could not be evaluated by the statistical model. During their residence time in the tunnel the particles undergo significant changes, resulting in a more compact structure. It is concluded that this is mainly due to adsorption of volatile material from the gas phase to the particle surface. Copyright © 1996 Elsevier Science Ltd

Key word index: Vehicle emissions, diesel, gasoline, particulate emissions, emission factors, tunnel, field experiment.

1. INTRODUCTION

Motor vehicles, in particular diesel powered vehicles, are known to be important sources of particulate matter. The mutagenic and carcinogenic potential of diesel exhaust particles has been demonstrated in several studies (McClellan, 1987; Heinrich, 1989; Mauderly *et al.*, 1993). Therefore it is of great importance for human health to measure not only the gaseous but also the particulate emissions of motor vehicles. Field measurements are an addition to test bench studies. The determination of the emission factors (EF) of specific engines cannot describe the input of a fleet of vehicles with different vehicle ages, states of repair, etc. that exist on the road. Moreover, chassis dynamometer studies cannot simulate aging effects or the mixing of the exhaust gases of different vehicle types.

The freeway tunnel emission measurements presented in this paper were part of the EUROTRAC subproject GENEMIS. Besides the evaluation of emission factors the aim of the measurements was to study changes of the characteristics of combustion aerosols during their residence time in the tunnel. Typical aging effects of combustion aerosols in the atmosphere are adsorption or desorption of semi-volatile material on or from the particle surface (Burtscher *et al.*, 1993a). Other possible aging effects are restructuring and aggregation processes as well as degradation of particle bound volatile material as for example polycyclic aromatic hydrocarbons (PAH) (Nielson, 1988; Kamens *et al.*, 1988). These processes change the physical and chemical properties of the particles, which again have an influence on the lifetime of the particles in the atmosphere.

2. EXPERIMENTAL

Description of the Gubrist tunnel

Figure 1 shows the setup of the two measuring stations in the Gubrist freeway tunnel near Zürich, Switzerland. The tunnel is divided in two separate tubes, with only one direction of the traffic flow in each tube. The speed limit is 100 km h^{-1}. During the measurement period the ventilation of the tunnel was switched off. Because of the one-way traffic, the wind field was produced by car movements only.

During the day, at times of heavier traffic, the air velocity was high (9 m s^{-1}) and the air residence time was about 6 min. In contrast, at night, with low traffic density, the air residence time was about 20 min (air velocity 2.7 m s^{-1}). Measurements were performed simultaneously at the entrance and exit of the slightly ascending (1.3%) 3250 m long southern tube.

The first measuring station was located about 100 m after the tunnel entrance, where freshly emitted particles were measured. The fraction of ambient aerosols is considered to be small due to the following reason: The black carbon mass concentration 100 m after the tunnel entrance was 14.1 $\mu g\ m^{-3}$ on workdays (see below), which is by far higher than

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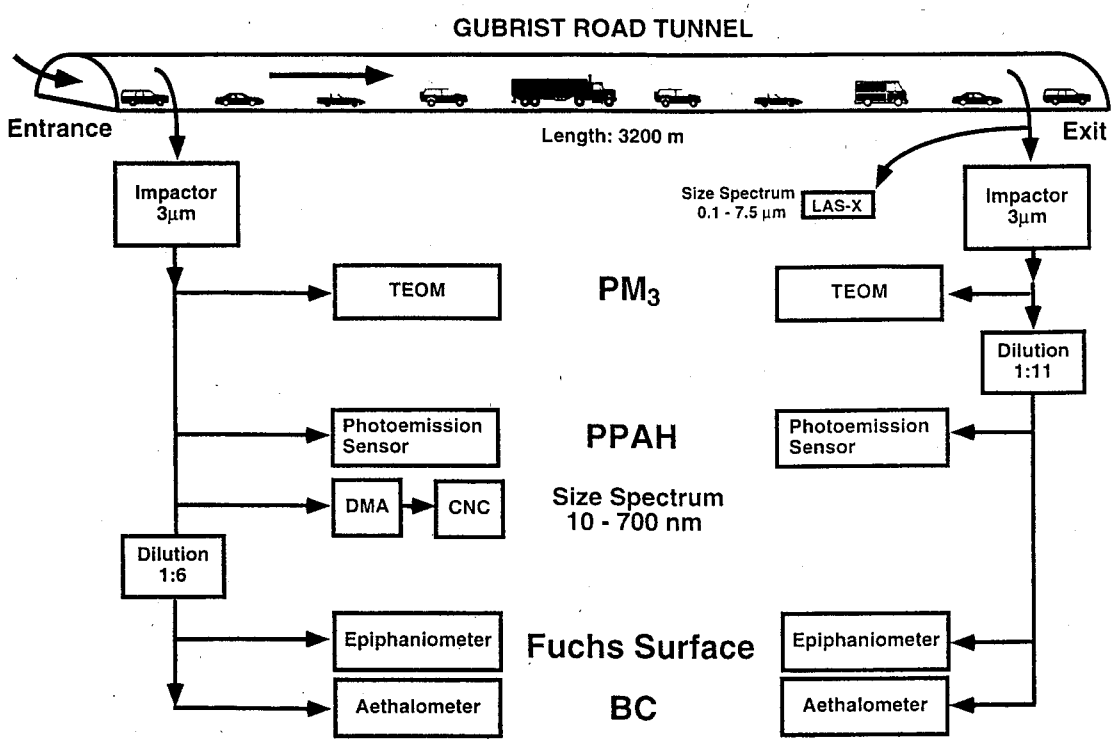

Fig. 1. Sketch of the Gubrist freeway tunnel with the experimental setup of the two measuring stations at the tunnel entrance and exit.

the value expected for a relatively clean suburban area. The second station was located 100 m before the tunnel exit, where a mixture of fresh and aged particles was present. At both stations the air was sampled 50 cm below the top of the tube. Further details are described in Staehelin *et al.* (1995).

Aerosol measurements

Figure 1 shows that for most instruments only particles in the respirable size range were sampled. An impactor removed all particles with an aerodynamic diameter larger than 3 μm. The particle mass concentration (PM_3) was measured with two tapered element oscillating microbalances (TEOM, Patashnick and Rupprecht, 1986) operating at 50°C and a flow rate of 3 $\ell\,min^{-1}$. These instruments collect particles on a filter which is placed on a resonating tube. A change in resonant frequency is measured when particles are collected on the filter. Similar to the PM_{10} standard, PM_3 stands for particulate matter of aerodynamic diameter 3 μm or less. At the tunnel exit it was necessary to dilute the air for most instruments (see Fig. 1). This was done by mixing one part of aerosol with ten parts of filtered air. Photoelectric charging of particles was used to monitor the concentration of particle bound polycyclic aromatic hydrocarbons (PPAH) (Burtscher and Siegmann, 1993; Hart *et al.*, 1993). Measurements were performed with two photoelectric aerosol sensors working with low-pressure mercury lamps ($\lambda = 183$ nm). These sensors were calibrated every second day with a commercially available photoelectric aerosol sensor (LQ1, U. Matter AG). From time to time size-spectra were measured. At the tunnel exit an optical particle counter (LAS-X, Particle Measurement Systems) measured the particle size distribution between 0.1 and 7.5 μm. The LAS-X was the only instrument which measured the aerosol before the impactor. At the tunnel entrance we used an SMPS system (differential mobility analyzer (DMA) combined with a condensation nucleus counter (CNC)). The DMA (TSI 3071A) was operated with a total flow rate of 3.3 $\ell\,min^{-1}$ and the data acquisition was performed using the SMPS software. For the other measurements at the tunnel entrance the air was diluted by a factor of 6 by applying the same dilution method as described above. At both stations epiphaniometers (Gäggeler *et al.*, 1989) measured the Fuchs surface (FS) of the aerosol. The Fuchs surface represents the total particle surface area a diffusing molecule "sees"; for more details see Pandis *et al.* (1991). With aethalometers (AE9 and AE10, Magee Scientific) the black carbon mass concentration (BC) was determined (Hansen *et al.*, 1984). These instruments measure the change of the attenuation (ATN) of visible light through a fibrous filter. The black carbon content S_{BC} of the filter and the ATN are related by the linear relationship $S_{BC} = ATN \cdot \sigma^{-1}$, where σ is the absorption coefficient. Liousse *et al.* (1993) showed that σ depends on the atmospheric environment which is studied (e.g. age and origin of the particles). Despite this difficulty an absorption coefficient of $\sigma = 19\ m^2\,g^{-1}$ (factory calibration) was applied, which was thought to be reasonable for this application. The time resolution of the obtained data was between 2 and 10 min depending on the apparatus.

To compare the instruments at the entrance and exit with each other, additional measurements in a car-parking garage were performed. Three couples of identical instruments (TEOM, aethalometer and epiphaniometer) were connected to the same sampling line. A correlation of half-hour values for a period of 48 h showed a very good agreement between the epiphaniometers ($R^2 = 0.99$), while the correlations between the aethalometers ($R^2 = 0.87$) and the TEOMs ($R^2 = 0.78$) were slightly lower. The slopes of the aethalometer and TEOM regression lines were between 1 and 1.1. Therefore no corrections were made in the factory calibration. The epiphaniometer data were used to calibrate these instruments

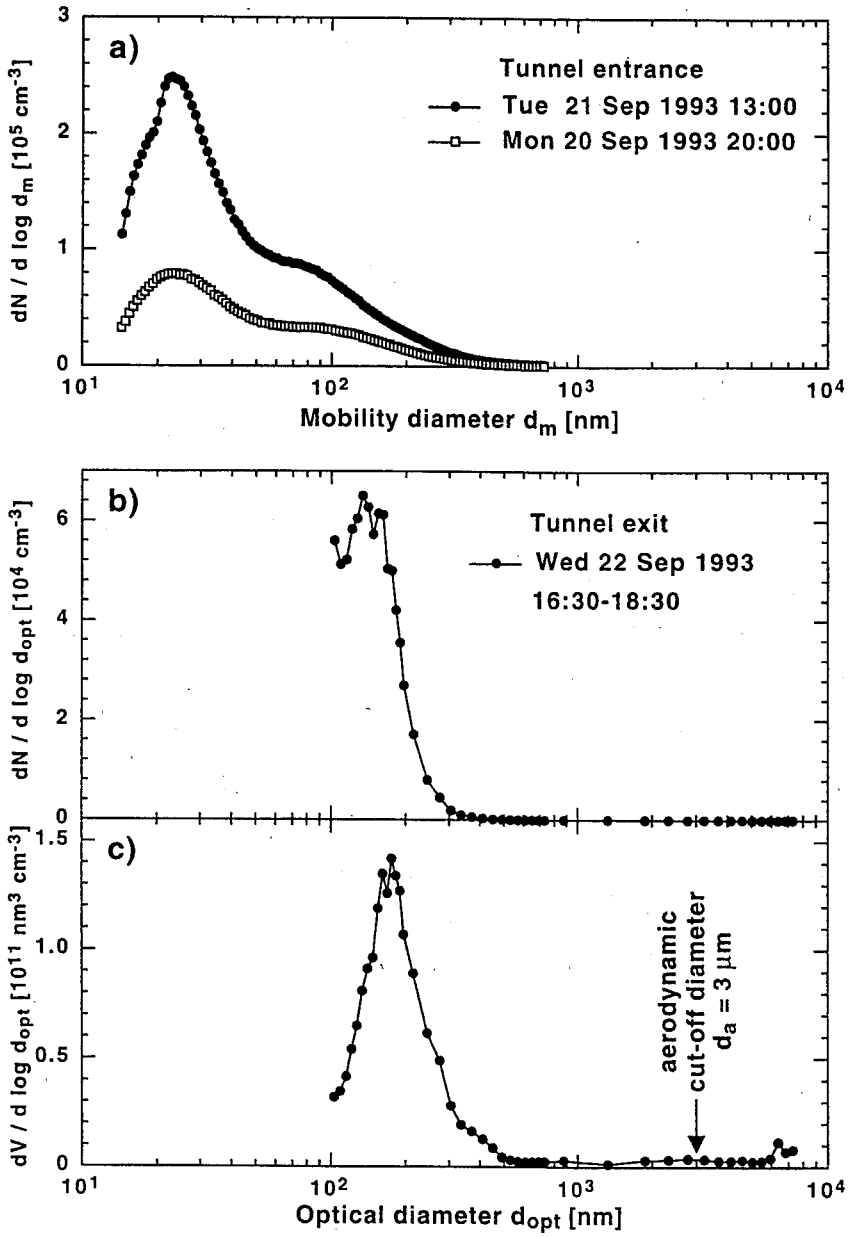

Fig. 2. Number size distribution at the tunnel entrance measured with a DMA (a) and at the exit measured with an optical particle counter (b). Also shown is the volume size distribution (c) calculated from (b) by assuming that the particles were spheres.

medium load (Burtscher *et al.*, 1993b). These values were measured for freshly emitted particles from single sources. The BC to PM_3 ratio determined in this study for diesel engine particles is lower (31%, see below). This difference might be due to adsorption of semi-volatile material from the gasoline engine particles onto the diesel particles during their residence time in the tunnel.

Regarding the average emission on workdays (Table 1), the photoemission as well as the epiphaniometer signal increased only by a factor of 2 from the tunnel entrance to the tunnel exit. A possible explanation is again adsorption of semi-volatile material on the particles, which results in a more spherical structure. Here, adsorption of water onto the particle surfaces can be excluded because the relative humidity (RH) of the air in the tunnel was below 85% during the whole measurement period. At this RH, freshly emitted combustion aerosols do not adsorb water (Weingartner *et al.*, 1995). Possibly this adsorption process is accompanied by a restructuration of the agglomerated particles. This adsorption and restructuration behavior is also illustrated in Fig. 5, which shows the correlations of the epiphaniometer and the

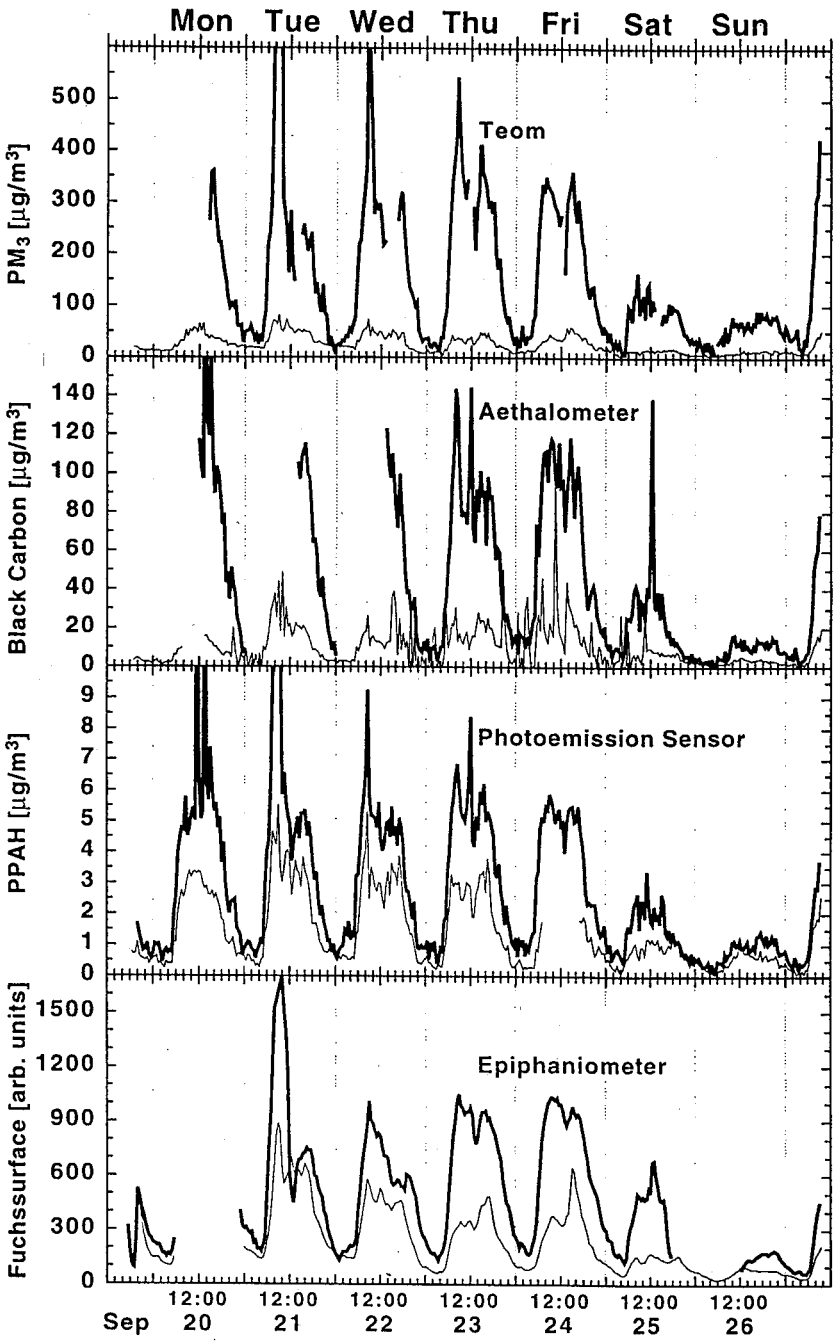

Fig. 3. The signals of the TEOM, the aethalometer, the photoemission sensor and the epiphaniometer at the tunnel entrance (thin line) and at the tunnel exit (thick line).

Table 1. Average emission of PM_3, BC, PPAH and Fuchs surface (FS) during a workday and the weekend

	Workday (23 September)			Saturday (25 September)			Sunday (26 September)		
	Entrance	Exit	Factor	Entrance	Exit	Factor	Entrance	Exit	Factor
PM_3 ($\mu g/m^3$)	25.0	201.6	8.07	12.8	70.9	5.55	10.9	52.7	4.86
Black carbon ($\mu g/m^3$)	14.1	55.7	3.96	5.5	19.7	3.56	2.9	8.7	2.96
PPAH ($\mu g/m^3$)	1.73	3.47	2.01	0.75	1.41	1.89	0.50	0.81	1.62
FS (cps)	240.6	606.0	2.52	119.5	293.1	2.45	66.3	98.9	1.49

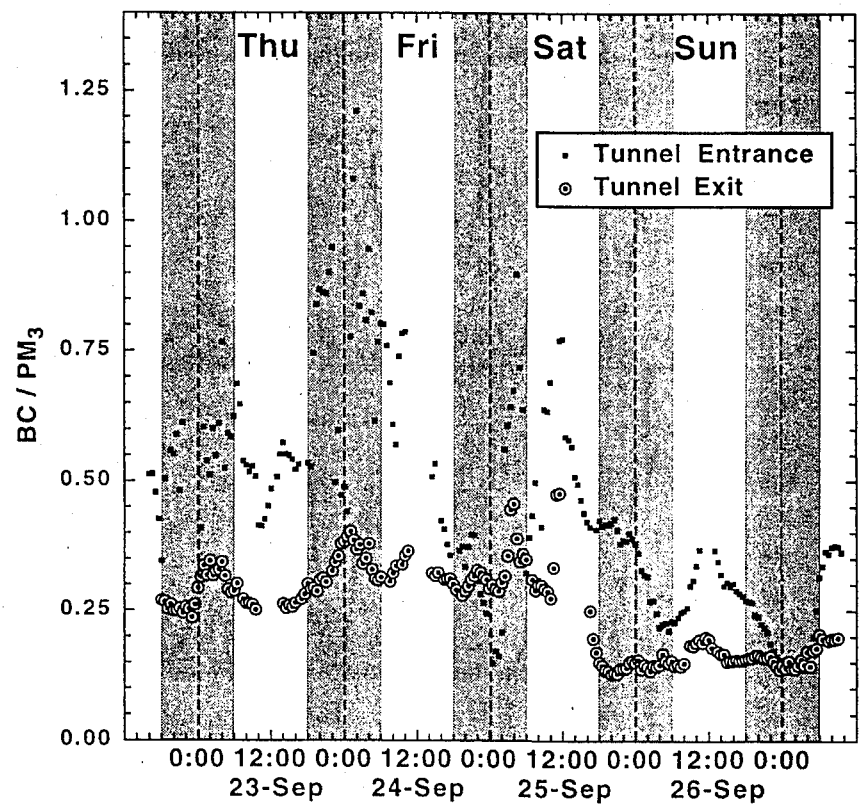

Fig. 4. Temporal variation of the black carbon (BC) to PM_3 concentration ratios at the tunnel entrance and exit.

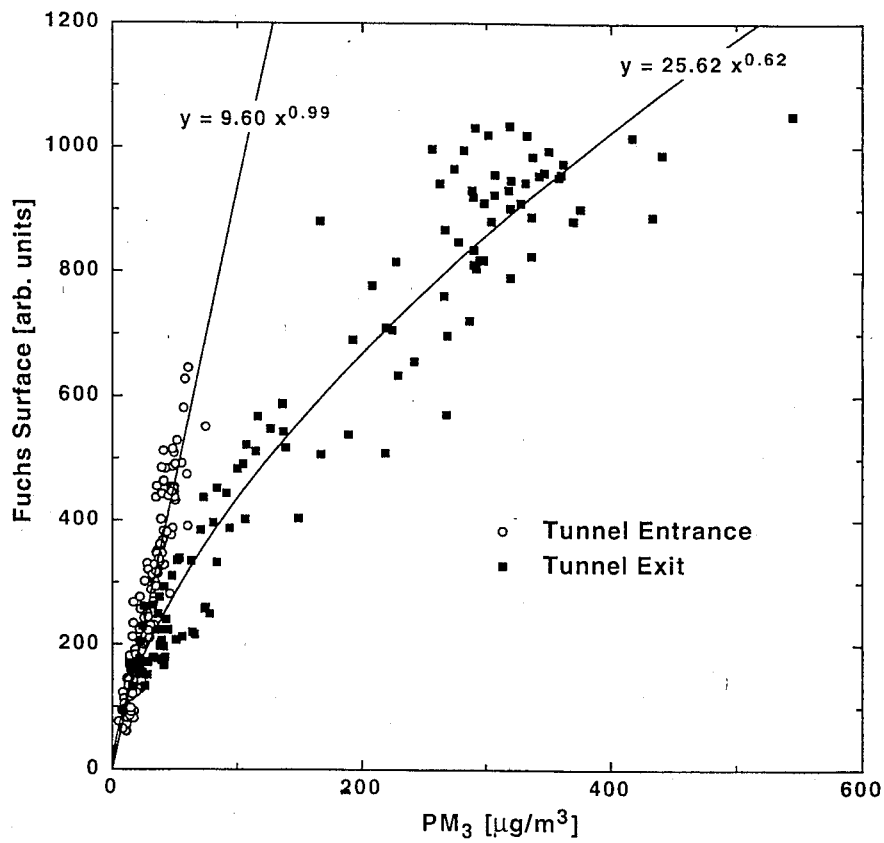

Fig. 5. Fuchs surface vs PM_3 concentration for the tunnel entrance and exit during workdays (22–24 September). The different slopes indicate more compact particles at the tunnel exit compared to the entrance.

TEOM signals for both stations during workdays (22–24 September). Apparently, the surface to volume ratio of the particles decreases during the residence time of the particles in the tunnel. This means that the particles become more compact. In Fig. 5 this process is illustrated by a different slope of the fitted curves. A similar behavior is found for the correlation between the photoemission signal and the TEOM signal.

Evaluation of emission factors

The total emission factors (EF) were calculated from the difference in concentration between exit and entrance according to

$$EF = \frac{(c_{\text{exit}} - c_{\text{entrance}}) \cdot v_{\text{air}} \cdot t \cdot A}{L \cdot N} \quad (1)$$

where c denotes the concentration, v_{air} the velocity of the air, A the tunnel cross section, L the tunnel length and N the number of vehicles which passed the tunnel in the time interval t. By linear regression the emission factors for certain vehicle categories and speed classes were determined using 10 min averages. As an example, Fig. 6 shows a scatter plot of the PM_3 emission factor (calculated by equation (1)) vs the fraction of diesel vehicles (i.e. the sum of the heavy duty vehicles plus diesel-powered delivery vans and diesel passenger cars).

First, a regression model using the two classes light duty vehicles (LDV, i.e. passenger cars and delivery vans) and heavy duty vehicles (HDV),

$$EF_i = EF_{\text{LDV}} \cdot p_{\text{LDV},i} + EF_{\text{HDV}} \cdot p_{\text{HDV},i} + \varepsilon_i \quad (2)$$

was fitted. The emission factors EF_i are the average emission per vehicle and kilometer in the time interval i, the two p_i's denote the fraction of vehicles of the two classes, and ε_i the random errors. The model allows to estimate the emission factors EF_{LDV} and EF_{HDV} for the two vehicle classes.

Since the residuals were unsymmetrically distributed and showed unequal variances, the estimation was in fact based on

$$\sqrt{EF_i} = \sqrt{EF_{\text{LDV}} \cdot p_{\text{LDV},i} + EF_{\text{HDV}} \cdot p_{\text{HDV},i}} + \varepsilon_i \quad (3)$$

which describes the same physical relationship as in equation (2). For more details about the estimation method, which also takes care of the serial correlation of the random errors, see Staehelin *et al.* (1996).

The residuals of this model for Sunday observations were mostly negative. This undesirable effect can be attributed to the van fraction in the LDV class, which was much lower (2%) on Sunday than on

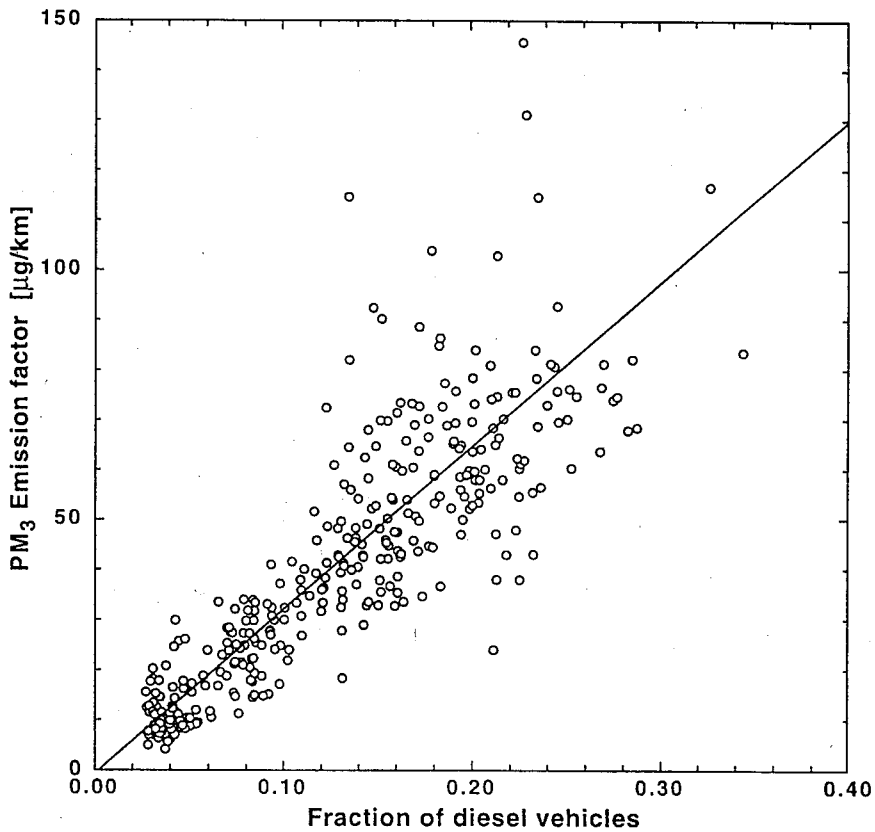

Fig. 6. PM_3 emission factor (calculated with equation (1)) vs fraction of diesel vehicles.

Table 2. Emission factors and error estimates calculated by using either light duty vehicles (LDV) and heavy duty vehicles (HDV) (equation (3)) or gasoline and diesel vehicles as vehicle categories (equation (4)) (the LDV category contained 5.7% diesel vehicles on workdays)

	LDV	HDV	Gasoline	Diesel	Units
PM_3	8.53 ± 0.47	383.5 ± 10.7	-0.84 ± 1.36	313.0 ± 12.1	$mg\,km^{-1}$
BC	1.60 ± 0.21	122.7 ± 5.7	-1.77 ± 0.58	98.0 ± 6.8	$mg\,km^{-1}$
PPAH	0.120 ± 0.009	3.01 ± 0.17	0.010 ± 0.036	2.70 ± 0.30	$mg\,km^{-1}$
FS			18.7 ± 25.8	411 ± 141	arbitary units

workdays (11%). We therefore turned to using the two classes of gasoline and diesel-powered vehicles,

$$\sqrt{EF_i} = \sqrt{EF_{\text{gasoline}} \cdot p_{\text{gasoline},i} + EF_{\text{diesel}} \cdot p_{\text{diesel},i}} + \varepsilon_i. \quad (4)$$

This model fitted the data well. Using speed as an additional explanatory variable brought no significant improvement.

The results (Table 2) clearly show that the particulate emissions are almost completely produced by diesel vehicles. These are mainly trucks, because the fraction of diesel vehicles with a weight < 3.5 t is 37%. The values for the LDV category cannot be compared with chassis dynamometer studies of gasoline engines because the LDV category contains 5.7% diesel vehicles on workdays. Negative PM_3 and BC emission factors were calculated for gasoline cars. Particulate emissions in the tunnel are dominated by diesel vehicles, which results in rather large errors of the emission factors for gasoline engines. While the emission factor for PM_3 for gasoline engines is not statistically significant, the result for BC can be explained by differences in composition of the car park between workdays and weekend.

In Table 3 we compare the PM_3 emission factors obtained in this study for gasoline and diesel vehicles with values determined by other authors in tunnel measurements and at engine test benches. The fraction of black carbon (BC) and PPAH in the aerosol are also compared with literature data. The PM_3 emission factors measured in this study for gasoline and diesel engines are lower than the ones reported by Pierson and Brachaczek (1983) and Williams *et al.* (1989a, b). There are several possible reasons for this difference. First, their data are total suspended particulate matter (TSP) and not PM_3 emission factors. Second, the data by Williams *et al.* (1989a, b) were from test engines running over an urban test cycle (ADR 36 and ADR 37) with varying speed or load. Third, the average mass of the diesel trucks in the study of Pierson and Brachaczek (1983) was about 30 t, whereas in Switzerland the allowed upper limit of the truck weight is 28 t. Moreover, progress in engine technology will also contribute to lower emission factors.

In the case of gasoline engines, emission factors depend strongly on the lead and bromine content of the fuel. Williams *et al.* (1989a) measured exhaust emission factors on engine test benches for 22 spark ignition vehicles using leaded fuel (lead content 0.4 g Pb ℓ^{-1}). They found a relatively high TSP emission factor of 73 ± 46 mg km^{-1}, which was explained by 46% lead and bromine. This is also true for the high TSP emission factor for gasoline engines reported by Pierson and Brachaczek (1983). In 1977, when that study was performed, 52% of the engines were operated with leaded gasoline. The particulate Pb and Br emission factors of 12.4 ± 1.6 and 5.75 ± 0.45 mg km^{-1}, respectively, given by these authors mean that about 35% of the TSP emission was due to lead and bromine. During our measurements (in 1993, Switzerland) the fraction of gasoline vehicles operated with leaded gasoline was less than 20%, with a lead content of 0.15 g Pb ℓ^{-1}. Therefore, the low emission factors for gasoline engines measured in the Gubrist tunnel are certainly also due to the increased use of catalysts and the phase-out of leaded fuel.

Table 3 shows that in this study 31% of the particle mass emitted from diesel vehicles (with $d < 3$ μm) was black carbon. This value is in good agreement with the EC fraction obtained by other authors in tunnel experiments, despite the different analysis method. The EC is that fraction of total carbon which is not volatilized at 600°C in an inert atmosphere.

The amount of PPAH on diesel engine particles was investigated by several authors (see Table 3) by collecting fresh exhaust particles on filters or low-pressure cascade impactors and subsequent analysis with HPLC or GC-MS. They found a PPAH fraction between 0.04 and 0.8 wt% for diesel engines in dynamometer studies. In our field study the PPAH content of diesel particles was 0.86%. So far it is not clear why we obtained a relatively high PAH content of the particles. The higher value could be due to different engine types and driving conditions in the tunnel. Another possible explanation is, again, that semi-volatile material (in particular PAH) adsorbs from gasoline particles onto the surface of diesel particles. As already mentioned gasoline engines emit a higher fraction of semi-volatile material, in particular PPAH. Grimmer *et al.* (1991) report a PPAH fraction of 2.8 wt% for gasoline engine exhaust. Such an effect would not be seen in dynamometer studies, which emphasizes the importance of such field experiments.

Table 3. Emission factors (EF) for gasoline and diesel vehicles determined in tunnel experiments and in chassis dynamometer studies (the black carbon (BC) or elemental carbon (EC) as well as the PPAH fraction were determined for diesel trucks unless stated)

Reference	Method	Measured size fraction	EF Gasoline (mg/km)	EF Diesel (mg/km)	BC or EC fraction	PPAH fraction
Pierson and Brachaczek (1983)	Tunnel experiment	TSP	51.0 ± 30.0	870 ± 100		
Hering *et al* (1984)	Tunnel experiment	$PM_{1.3}$	8.0(estimated)	300 (estimated)	35% BC[a]	
Wittorff *et al.* (1994)	Tunnel experiment	PM_{10}	9.3 ± 37.3	416 ± 81		
Rogak *et al.* (1994)	Tunnel experiment	$PM_{2.5}$	4.3 ± 4.3	284 ± 155		
Lies *et al.* (1986)	Chassis dynamometer	TSP		174[b]		0.11%[b]
Williams *et al.* (1989a)	Chassis dynamometer	TSP[c]	73.0 ± 46.0[d]		4% EC[e]	
Williams *et al.* (1989b)	Chassis dynamometer	TSP[c]		PC: 228 ± 59[f]	66% EC	
				DV: 427 ± 190[g]	24% EC	
				HDV: 2090 ± 1750[h]	33% EC	
Metz (1993)	Chassis dynamometer	TSP	< 10[i]	300 ± 60		
Needham *et al.* (1989)	Chassis dynamometer	TSP			41% EC[j]	
Grimmer *et al.* (1991)	Chassis dynamometer	TSP				0.80%
Westerholm and Egebäck (1994)	Chassis dynamometer	TSP		545[k]		0.035%[k]
Borstel *et al.* (1995)	Chassis dynamometer	TSP				0.11% – 0.28%
This study	Tunnel experiment	PM_3	– 0.8 ± 1.4	313 ± 12	31% BC	0.86%

[a] Value for a car fleet with 3–6% diesel trucks.
[b] Mean values from diesel passenger cars without catalyst.
[c] The authors reported that the majority of the particles was < 1.5 μm.
[d] Average of 22 S.I. vehicles driven over an urban test cycle (ADR 37); leaded fuel 0.4 g Pb ℓ^{-1}.
[e] For gasoline engines operating with leaded fuel 0.4 g Pb ℓ^{-1}.
[f] Average and standard deviation of 4 passenger cars (PC); ADR37.
[g] Average and standard deviation of 15 delivery vans (DV); ADR37.
[h] Average and standard deviation of 13 heavy duty vehicles (HDV)); ADR36.
[i] High way test cycle, leaded fuel 0–0.55 g Pb ℓ^{-1}.
[j] Average of 16 heavy duty vehicles.
[k] Average of two heavy duty vehicles without any exhaust gas treatment.

However, more detailed information is necessary in order to validate this hypothesis.

The influence of tire wear on PM_3 concentration

In earlier studies, most of the material produced by tire wear was found as non-suspendable material deposited near the road (Pierson and Brachaczek, 1974). Pierson and Brachaczek (1975) reported that the fraction of the tire-related airborne particulate matter was about 2% of the TSP for a typical car mix. These particles were typically in the size range between 20 and 40 μm (Schultz *et al.*, 1993) and should therefore not contribute significantly to the submicron size range.

In contrast, Israël *et al.* (1994) argued recently that a considerable fraction of the fine mass concentration might result from tire wear. However, the following points indicate that the submicron particles are mainly emitted by the tail pipes.

- The PM_3 emission factor for diesel obtained in this study agrees well with chassis dynamometer studies, where only tail pipe emissions were measured.
- The size distribution of the aerosol at the tunnel exit (Fig. 2b) shows a low particle mass concentration with a light scattering diameter between 1 and 10 μm.
- Tires are mainly composed of rubber, carbon black (30%) and varying amounts of sulfur (2–5%) (Cardina, 1973; Bhakuni *et al.*, 1985). The carbon black is embedded into organic polymer compounds and therefore we expect tire wear not to be sensitive to photoelectric charging. The fact that the measured PPAH to PM_3 ratio for diesel engines is higher than chassis dynamometer measurements indicates that no large amounts of tire wear appear as submicron particles.

Therefore, we conclude that the measured small particle fraction (aerodynamic diameter < 3 μm) is mainly due to exhaust emissions with a very small amount of tire wear. This is in line with the latest findings of the Israël group, where they also reported a very low contribution of tire wear to the fine mass concentration (Rauterberg-Wulff *et al.*, 1995).

Correlation between particulate and gaseous emissions

Correlations of particulate emissions (PM_3, BC, PPAH and FS) with the continuously measured gas emissions nitrogen oxides (NO_x), carbon monoxide (CO) and total hydrocarbons (t-HC) were investigated at the tunnel entrance and exit. The measurement of these gaseous emissions is described in detail elsewhere (Staehelin *et al.*, 1995). We found a significant

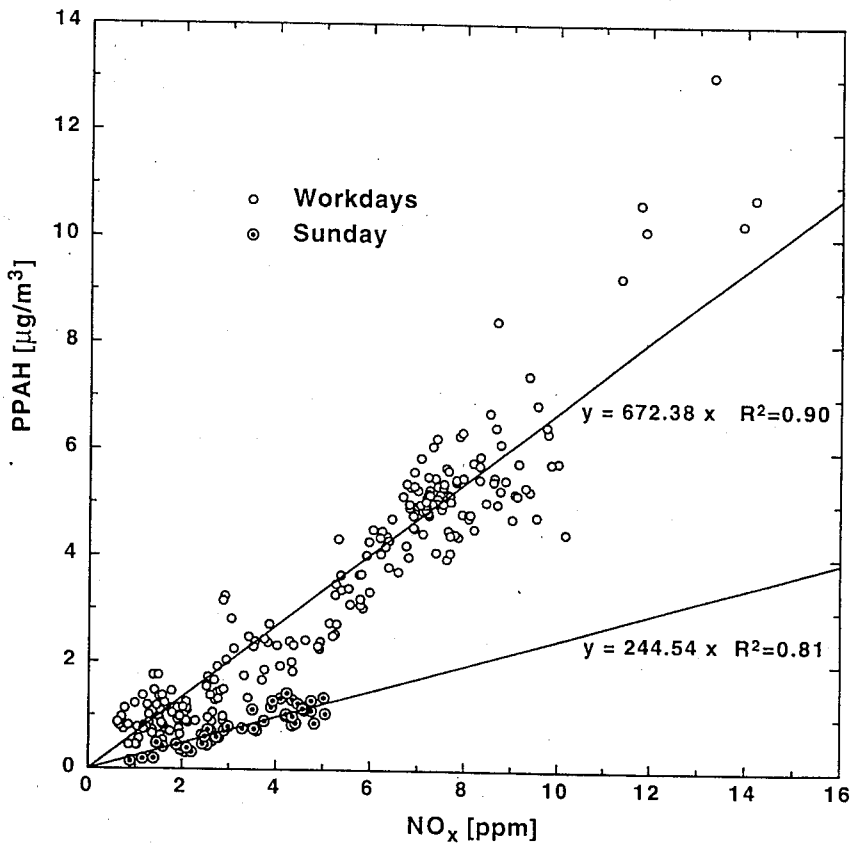

Fig. 7. PPAH concentrations vs NO_x concentrations at the tunnel exit for workdays and for Sunday.

difference of the correlations performed on workdays and on Sunday. As an example, Fig. 7 shows the correlations of the PPAH concentration with the NO_x concentration for workdays and for Sunday. Lower slopes of the regression lines, by a factor of 2 to 5, were found on Sunday for all correlations between gaseous and particulate emissions at the entrance as well as at the tunnel exit. The highest correlation coefficients at the tunnel exit were found between the NO_x concentration and the aerosol data ($0.7 < R^2 < 0.9$) for workdays and Sunday. The correlation coefficients between the CO and t-HC concentrations and the aerosol data are relatively high for Sunday ($0.6 < R^2 < 0.8$) and relatively low for the workdays ($0.3 < R^2 < 0.6$).

According to Staehelin *et al.* (1996) NO_x emissions are about 20 times higher for diesel engines than for gasoline engines. Thus, the particulate matter and the NO_x emissions have the same major source (i.e. diesel engines), which explains the good correlation. On the other hand, CO and t-HC are emitted to a significant fraction by the gasoline engines, resulting in low correlation coefficients. The different slopes between particulate and NO_x emissions are due to the fact that the ratio of the diesel to gasoline vehicles emission factors is much higher than 20 in the aerosol case. This results in lower slopes on Sunday with a low fraction of diesel vehicles.

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