ACEA Programme on the Emissions of Fine Particulates from Passenger Cars (2)

Part1: Particle Characterisation of a Wide Range of Engine Technologies**Martin Mohr, Urs Lehmann**

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ABSTRACT

The results of an investigation on PM emissions over an extended range of engine technologies are reported in this paper. These included modern Diesel vehicles, a particle trap equipped Diesel vehicle, a port-injected gasoline vehicle and direct injection gasoline vehicles. Various particulate measurement instruments were employed simultaneously, including SMPS, CPC, ELPI, for number and size and the standard gravimetric filter method for mass. Soxhlet extraction was applied to determine the chemical composition. The particle emissions showed a wide range in concentration for the different vehicles. The particle concentration of the port-injected gasoline vehicle and the trap-equipped Diesel vehicle were observed at background level. The direct injection gasoline vehicles showed a large variation in particle concentration depending on concept and operation mode but in all tests higher particle emissions in number and mass were observed than the conventional gasoline vehicle, but substantially below the conventional Diesel vehicles.

INTRODUCTION

Automotive exhaust emissions have been regulated for many years. Enforced by gradual tightening of the limit values for specified gaseous components, and particulates for Diesel engines, on one side and the awareness of the limited fossil energy resources and the enlarged atmospheric green house effect on the other side, there is an ongoing development of new technologies of engines, exhaust gas aftertreatment systems, fuel and lubricant oil. Nowadays, engine development and exhaust gas aftertreatment systems equally have to aim at high fuel efficiency and low emission levels.

In Europe there is a trend to Diesel engines in passenger cars due to advantages in fuel efficiency

compared to spark ignition engines. This advantage is confronted with higher specific emissions in nitrogen oxides and especially particulate matter. A very efficient method to reduce the particle emissions is the use of particle trap in the exhaust gas line, which is applied for example by PSA Peugeot Citroën [1]. On the other side, many efforts have been made to improve the fuel efficiency of spark ignition engines. Beside the variable valve train technology, one of the most promising approaches is the direct injection combustion technology [2] to reduce throttle losses. As many DI technologies are based on the new concept of stratified mixing the question arises about its emission levels, especially for particulate matter.

We present results of the 2nd ACEA programme on fine particulate emissions from passenger cars [3]. It is designed to continue from the 1st ACEA particle programme [4]. The objectives of the 2nd programme were subdivided in two parts: Part 1 was to investigate the particulate emission of an extended range of engine technologies of current mass production passenger cars, including modern Diesel vehicles, a particle trap equipped Diesel vehicle, port-injected gasoline vehicles and direct injection gasoline vehicles. The particles were characterised in number, size and composition and related to the regulated mass measurement method. The results of this part are presented in this paper. The objectives of Part 2 was to investigate the influence of fuel sulphur content and dilution systems on the particle emissions. These results are discussed in [5].

EXPERIMENTAL**INSTRUMENTATION**Gravimetric determination of Particulate Matter (PM)

Particulate Matter (PM) of the exhaust gas was determined gravimetrically according to the

regulations [70/220/EEC]. They were weighed on a microbalance (Mettler Toledo – MT5) having a readability of 1 µg and a measurement uncertainty of 2.6 µg for the applied mass range. For some filters, the loading was very low (down to 10 µg), resulting in a lower precision of these tests. This especially applies for the conventional gasoline vehicle and the Diesel vehicle with the trap.

Chemical composition of Particulate Matter (PM)

The chemical composition of the particulate matter was analysed by extraction in a Soxhlet apparatus. By two successive extractions with dichloromethane and a mixture of 2-propanol/H₂O (3:2), the loading of the gravimetric filter samples could be divided into a soluble organic fraction (SOF), a soluble inorganic fraction (SINOF) and non-soluble fraction (residue). By ion chromatography analysis of the soluble inorganic fraction, the amount of sulphate ions is determined.

Scanning Mobility Particle Sizer (SMPS)

Two SMPS instruments (TSI Incorporated, USA) were operated during constant operation modes of the vehicles to obtain high-resolved number size distributions for a size range between 11 and 514 nm. Data of particle emissions of a transient cycle with a time resolution on one-second basis were obtained for the total number, using the CPC (TSI 3022A) alone or for an individual narrow size channel (20nm) by setting the DMA voltage on fixed value. The particle size measured by SMPS is an electrical

mobility-equivalent diameter. Further details of the method are described at [6,7].

Electrical Low Pressure Impactor (ELPI)

The Electrical Low Pressure Impactor (Dekati LTD, Finland) consists of 12 stages, which cover a size range from 32 nm to about 7 µm (50%-cut diameters). In this study the size range below 1 µm was evaluated only as the sampling system was not optimised for larger particles. With a read-out rate of 1 second, the ELPI is able to follow a transient driving cycle. Further details of the instrument are described in [8]. The particle size measured by ELPI is an aerodynamic-equivalent diameter, which is defined for unit density. Particle size and, as a consequence, the concentration is affected by the particle density. For this reason, absolute number and size measured by SMPS/CPC and ELPI are not directly comparable.

Thermodesorber (TD)

A thermodesorber was used to distinguish between solid and volatile particles. The thermodesorber consists of two parts: a temperature controlled heated tube where the volatile material evaporates and a cooled chamber filled with active charcoal to adsorb the re-condensates. Further details of the instruments are described in [9].

In this study the thermodesorber was controlled to a temperature of 350°C. The residence time in the heated tube was about 1.8 s. All SMPS/CPC measurements were carried out simultaneously with and without TD. The losses of solid particles of about 25% were corrected in the results.

Figure 1: Schematic of the experimental set-up

vehicle code	1	2	3	4	5	6	7
Inertia Mass [kg]	1470	1590	1590	1020	1250	1360	1020
Transm (M5, A5)	M5	M5	M5	M5	M5	M5	A5
Displacement [ccm]	1910	1896	2179	1242	1998	1834	1390
No of Cylinders	4	4	4	4	4	4	4
Valves per Cylinder	2	2	4	4	4	4	4
Max Power [kW]	81	74	98	55	103	92	77
Max Torque [Nm]	275	240	314	110	200	174	130
Compression ratio	18.45	19.5	18	10	10	12.5	12
Combustion Type	Diesel DI	Diesel DI	Diesel DI	SI	SI	SI	SI
Injection Type	C-Rail	Mech	C-Rail	MPI	Direct Stoic	Direct Lean	Direct Lean
EGR Type	Electr Closed Loop	Electr Closed Loop	Electr Closed Loop	none	Electr Closed Loop	Electr Closed Loop	Electr Closed Loop
Emis Control Type	Oxy	Oxy	Oxy, Trap	3W	3W	3W / NOx Storage Cat	3W / NOx Storage Cat

Table 1: Technical data of the vehicles

Dilution systems

All samples for the particle mass measurements were taken from the CVS dilution tunnels according to the legislation. The exhaust of the Diesel and gasoline vehicles was led into two separate tunnels. A minimum dilution ratio of 10 was used for all operation conditions. For particle number measurements, a separate probe was installed close to the probe for the gravimetric filter sampling. In order to achieve a higher dilution ratio, an additional ejector-type diluter unit with a constant dilution factor of about 10 was installed downstream of the CVS for particle number measurements. Details of the working principle of the ejector-type diluter are given at [10].

COMPLETE EXPERIMENTAL SET-UP

Figure 1 shows the complete experimental set-up. The aerosol sample flow was distributed to the two CPC's, the three SMPS (DMAs) and to an ELPI. The total number of emitted particles was measured simultaneously with CPC2 and CPC4, CPC2 was equipped with a thermodesorber. For size related measurements two DMA+CPC combinations were used (DMA1+CPC1, DMA3 + CPC3). DMA1+CPC1 were installed parallel to CPC2 downstream the TD. The DMA5 + CPC5 was used with fixed size channel during transient tests only.

TEST VEHICLES

Seven vehicles were selected for this measurement programme covering a wide spectrum of engine technologies in current mass production. The vehicles 1 to 3 were Diesel passenger cars, one of them was equipped with a particulate trap with periodic regeneration. Vehicles 4 to 7 were gasoline passenger cars, one of them was naturally aspirated with MPI. The other three vehicles were equipped with direct injection system, two of them suited for lean combustion. The details are given in Table 1.

		DIESEL	GASOLINE
		D210F50	CEC-RF-02-99
Density @ 15°C	kg/m ³	822.1	750.5
Sulphur	mg/kg	< 10	< 10
Aromatics	% wt	20.8	37.5
Saturates	% wt	79.2	
Benzene	% wt		0.16

Table 2: Fuel specification data

FUELS AND LUBRICANTS

All tests were run with fuels with less than 10 ppm sulphur and the same commercially available high-quality lubricant (Shell Helix Plus). All Diesel and all gasoline vehicles were run with fuel of the same batch. The specification of the fuels and lubricant oil is given in Table 2 and Table 4, respectively.

Density @ 15°C	kg/m ³	854.6
Sulphur	mg/kg	4760
Viscosity at 40°C	mm ² /s	81.34
Viscosity at 100°C	mm ² /s	14.19
Sodium	mg/kg	7
Magnesium	mg/kg	1100
Phosphor	mg/kg	860
Zinc	mg/kg	1000
Chlorine	% wt	0.9

Table 4: Specification data of the lubricant oil

Vehicles	2 Diesel vehicles, DI 1 Diesel vehicle, DI, with particle trap 1 port-injected gasoline vehicle 1 direct injection gasoline vehicle, stoichiometric 2 direct injection gasoline vehicle, lean
Fuels	Diesel (S< 10 ppm), Gasoline (S<10 ppm)
Cycles	Transient: NEDC2000 Steady state: 120 km/h (5 th gear), 100 km/h (5 th gear), 50 km/h (4 ^h gear)
Measurement instruments	CPC DMA+CPC (SMPS) ELPI Gravimetric analysis
Chemical analytic technique	Soxhlet extraction
Sampling systems	Combination of CVS and ejector dilution unit

TEST PROTOCOL

The seven vehicles were driven over the transient cycle (NEDC2000) starting the engine at cold conditions and in different steady state modes (120km/h, 100 km/h, 50 km/h). All tests were run three times. The measurement period lasted for one week for each vehicle. The measurement conditions were always the same during the whole programme. To ensure the comparability between the tests an extended cross-validation of the measurement instruments and additional runs were carried out weekly with a reference vehicle (vehicle 1) during the whole 2 month period of the measurement programme part 1. The results showed a decrease of about 20% for all particle number instruments, but rather stable results for the particle mass. No reliable explanation can be given for this finding, but it is assumed that the drift is caused by the dilution system (CVS tunnel followed by an ejector-type diluter). More details are described in [5].

All cars were conditioned in exactly the same way. The conditioning for the transient tests were carried out according to the regulations. For the steady state tests, the vehicle was warmed up for 10 min at 80 km/h (5. gear), followed by the test sequence 120 km/h, 100 km/h, 50 km/h. At each speed 10 min were taken for conditioning to ensure stable conditions for the measurements. This was checked by monitoring the lubricant temperature and the exhaust gas temperature. For the measurement programme special attention was paid to the quality control including the vehicles, the fuel, the chassis dynamometer and measurement instruments. Table 3 gives an overview of the test programme.

RESULTS AND DISCUSSION

The results for particle number measurements were corrected for the dilution, the background, diffusion losses in the sampling lines according to [11] and for losses inside the thermodesorber. All concentrations are normalised to standard ambient conditions (273.15 K, 101.3 kPa). The detection limit was approximately 10⁹ particles per km determined by measurements of background concentration and diluted exhaust flow rates. Some results of vehicle tests were very close to this detection limit.

The range bars shown in the bar-diagrams indicate the maximum and minimum measured value out of 3 repetitions.

For vehicles with regeneration systems (particulate trap, NOx storage trap) the average particulate emission was calculated according to intervals of operation with and without regeneration.

TRANSIENT TESTS

Total number and mass

Figure 2 shows the total number (plain bars to the left scale) and mass (stripped bars the right scale) of the particles emitted during the NEDC2000. The concentrations for vehicle 3 and 4 are so low that they almost cannot be seen in the graphs. Lower particle number resulted in lower mass. The particulate mass measured from the trap-equipped vehicle 3 was 30 to 40 times lower than from Diesel vehicles 1 and 2, however, for number concentration the difference was 2 to 3 orders of magnitude between the vehicles with trap and without. Regarding the gasoline vehicles the conventional concept (vehicle 4) emitted significantly less particles than any of the direct injection versions (vehicles 5 to 7). It is remarkable that also the stoichiometric direct injection concept (vehicle 5) differs significantly from the conventional multi point injection concept. The comparison between measurements with and without thermodesorber (TD) show a similar result for the Diesel and the gasoline vehicles and does not indicate the presence of a significant fraction of nucleation particles for any vehicle run on low sulphur fuel.

Figure 2: Total particle number (plain bars to left scale) compared to particle mass (striped bars to right scale) of the Diesel (left graph) and the gasoline vehicles (right graph) in the transient cycle NEDC2000

The calculation of the mean particulate number and mass emission of vehicle 3 equipped with a particulate filter is based on weighted emissions measured on the NEDC2000 without and during regeneration. This is carried out according a test procedure as proposed for ECE Regulation 83.05 and takes into account the increased emissions during regeneration. The distance or number of test cycles is taken as the basis for the weighting between the two emissions values. The loading of the trap is completed on continuous NEDC2000 cycles until regeneration occurs, the number of cycles are noted for loading and then integrated into the following formula to calculate the mean mass particulate emissions.

$$\text{Mean particle emissions} = \frac{86 * M_{ri} + 1 * M_{si}}{87}$$

M_{ri} : mass, number w/o regeneration

M_{si} : mass, number with regeneration

In case of vehicle 3 the manufacturer reported a weighting of 86:1 for measurements without regeneration.

Time-resolved emissions

Figure 3 to Figure 6 show the time resolved total particle number emission for four vehicles, measured by CPC in the transient NEDC2000. The results are plotted as emission per distance, calculated for a constant speed over the cycle, i.e. the average cycle speed. Besides the different emission levels, different pattern can be observed for the different vehicle technologies: the emissions of the conventional Diesel vehicle are strongly related to the exhaust flow volume, the trap-equipped Diesel shows higher emissions in the cold start phase. A possible explanation could be the release of particles from the exhaust line walls, as measurements with TD do not show a significant difference. This is similar to the conventional gasoline vehicle, which in addition shows a slight increase for the high-speed phase as well. The direct injected gasoline vehicle shows an own distinctive pattern. Higher emissions are observed during constant phases at lower speed.

Number size distribution

With ELPI, time-resolved measurements of the particle size distribution for a transient cycle were carried out without a thermodesorber, thus volatile and solid particles were measured. The average number size distribution over the NEDC2000 cycle is shown for the Diesel and gasoline vehicles in Figure 7 and Figure 8, respectively. With exception of vehicle 5, the distributions of the gasoline vehicles peak at slightly lower size than the ones of the Diesel vehicles.

Figure 3: Time-resolved emission of total particle number during NEDC2000 cycle: Diesel vehicle (vehicle 1), measured by CPC w/o TD

Figure 5: Time-resolved emission of total particle number during NEDC2000 cycle: conventional gasoline vehicle (vehicle 4), measured by CPC w/o TD

Figure 4: Time-resolved emission of total particle number during NEDC2000 cycle: Diesel vehicle w/trap (vehicle 3), measured by CPC w/o TD

Figure 6: Time-resolved emission of total particle number during NEDC2000 cycle: direct injected gasoline vehicle (vehicle 6), measured by CPC w/o TD

Figure 7: Number size distribution for the Diesel vehicles in the transient cycle NEDC2000, measured by ELPI

Figure 8: Number size distribution for the gasoline vehicles in the transient cycle NEDC2000, measured by ELPI

Chemical Composition

The chemical composition of the particles as determined by analysis of the gravimetric filter samples for the transient cycle is shown in Figure 10. The results are normalised to 100% filter loading. The filter loading for vehicle 3 and 4 were below the detection limit for chemical analysis. Similar composition of the collected particulate for the Diesel and the direct injection gasoline engines was found. Most of the particulate matter consists of non-soluble material, which is usually soot. The organic soluble material contributes less than 20% of the mass whereas almost no water-soluble components were found. This is likely to be mainly due to the use of low sulphur fuel for the tests and that a minimum dilution ratio of 10 was applied for all operation conditions.

Figure 10: Composition of particulate matter sampled during the NEDC2000 based on gravimetric filter samples, determined by Soxhlet extraction and IC (for sulphate).

Figure 9: Number emission for three steady state modes (120 km/h, 100 km/h, 50 km/h) for the gasoline (upper graph) and Diesel (lower graph) vehicles

STEADY STATE TESTS

Total particle number

Figure 9 shows the total particle number emission obtained from tests at different constant speed conditions (120 km/h, 100 km/h, 50 km/h). Shown is the total number of emitted particles measured by CPC with and without thermodesorber and ELPI.

For the conventional Diesel (v1, v2), conventional SI (v4) and the stoichiometric DI SI vehicle (v5), a simple relation between speed and emissions is observed. Vehicle 3 is close to the detection limit and therefore no effect of speed on the emissions is detectable.

For the vehicles with the stratified combustion concept (vehicle 6 & 7) the emissions depend strongly on the combustion mode. Vehicle 6 operates in a lean combustion mode at 100 km/h and 50 km/h, and vehicle 7 does so at 50 km/h. As can be seen from the figure an increase in particle emission is caused by the stratified combustion mode, compared to the stoichiometric mode. The findings for the gasoline direct injection engines are in good agreement to other studies, e.g. [12]. In order to regenerate the NO_x-storage catalyst converter, both vehicles 6 and 7 regularly switch to a stoichiometric mode for short periods. The data presented here are weighted averages of both modes. Time resolutions of the particle number are shown in Figure 14.

Figure 11: Gravimetric filter measurements of particulate mass emission (PM) for the gasoline (upper graph) and the Diesel (lower graph) vehicles in steady state modes

Particle mass

In Figure 11, the results of the particle mass measurements are presented. They are in good agreement with the number measurements and demonstrate the effect of the particle trap as well. Despite the increased emissions for the direct injected gasoline vehicles at all operation conditions, the PM emissions remain substantially below the Diesel level. The absolute values for the trap-equipped Diesel vehicle (v3) and the conventional gasoline vehicle (v4) and other low values have an increased uncertainty due to very low filter loading.

Number size distribution

The number size distributions at different steady state conditions measured by SMPS are shown in Figure 12. Both conventional Diesel vehicles (v1 and v2) show very similar distributions for each speed. The mode of the distribution is at about 70 nm and did not vary significantly for the different speeds. At a speed of 120 km/h vehicle 5 produces a large number of particles smaller than 30 nm in diameter. By using a thermodesorber in the sampling line these particles vanish (dotted line). Obviously these particles consist of volatile material, which were formed by nucleation or condensation. However, the number of particles measured downstream of the TD is still high in comparison to the other gasoline vehicles for 120 km/h. For the other investigated speeds, the ranking of the gasoline vehicles is different and no nucleation mode was observed. Increased emissions were measured in lean combustion mode (vehicle 6: 100, 50 km/h, vehicle 7: 50 km/h) compared with stoichiometric combustion (vehicle 6 and 7). However, vehicle 7 with the latest technology reveals a visible improvement in emissions for lean combustion compared to the older technology of vehicle 6. Comparing the mode of the number size distributions measured by the SMPS the mode were found to be about 20 nm smaller for gasoline vehicles in average compared to Diesel vehicles. However, for the trap-equipped diesel vehicle (v3) in general and for the gasoline vehicles at operation modes with lowest emission the distribution curves were very flat.

Figure 12: Number size distribution of the particles in the exhaust for three constant speeds, Diesel vehicles all 3 speeds: upper left, gasoline vehicles: upper right: 120 km/h, lower left: 100 km/h, lower right: 50 km/h

Figure 13: Total number (left) and number of 20 nm particles (right) of the Diesel vehicle 3 in the transient cycle NEDC2000 measured with and without regeneration of the particulate trap (left logarithmic scale), total PM mass is shown in both diagrams (right linear scale)

EMISSION DURING REGENERATION OF AFTERTREATMENT SYSTEMS

To investigate the effect of trap regeneration, tests without and with regeneration were carried out for vehicle 3. For tests without regeneration, the control for the trap regeneration was switched off to suppress a possible start of regeneration. For tests with regeneration, the vehicle was equipped with pre-loaded trap-systems. For this reason, three identical systems were loaded on a durability chassis dynamometer using the car tested in this programme and fuel of the same batch as for these measurements.

As Figure 13 shows the total number of particles is more than 2 orders of magnitude higher when regeneration is carried out. Most of the particles are volatile and seem to be in the range of 20 nm. Also with thermodesorber the concentration is about 10 times higher during regeneration, which could indicate a release through the trap, a formation of particles downstream of the filter or a not sufficient efficiency of the thermodesorber. Figure 15 shows the time resolved number emission of non-volatile particles during the EUDC2000 for the three runs when regeneration takes place. Obviously, the regeneration started during the extra-urban part of the cycle when the exhaust temperature is increased.

Figure 15: Effect of trap regeneration: Time-resolved emission of the total non-volatile particles for vehicle 3 for EUDC2000 cycle when regeneration of the trap took place, measured by CPC with TD

Figure 14 illustrates the two different time management of vehicle 6 and 7 for the regeneration of the NO_x-storage catalytic converter. As can be clearly seen for both vehicles, the particle emission depends significantly on whether the engine runs in stoichiometric or lean combustion mode.

Figure 14: Time-resolved particle emission of the two direct injected gasoline vehicles (v6, v7) for the three steady state modes, measured by CPC

CONCLUSION

- The particle emissions of the port-injected gasoline vehicle and the trap-equipped Diesel vehicle are at background level and are close to the detection limit in number and mass.
- The trap-equipped Diesel vehicle shows, in all operation conditions, much lower particle emission in mass and number than the two modern Euro3 Diesel vehicles w/o trap. Higher particulate emissions are measured during regeneration but these have negligible effect on the distance-weighted results.
- The direct injection gasoline vehicles show a large variation in particle concentration depending on concept and operation mode. In all tests higher particle emissions in number and mass is observed than for the conventional gasoline vehicle. Engines with stratified lean combustion concept emit in the lean mode more than in the stoichiometric mode. The emissions from the direct injection vehicle with the latest technology reveals a reduction in particle number in lean combustion mode compared with the vehicle with the older generation direct injection technology.
- Most (> 80%) of the particulate mass emitted by non-trap equipped Diesel and DI injection gasoline vehicles is found to be non-soluble for all tested operation modes when low sulphur fuel is used. The composition does not vary significantly with engine technology. For the Diesel vehicle with trap

and the port-injection gasoline vehicle the collected mass was too low to analyse the composition of the particle matter.

- The mode of the number size distribution of the particles measured as electrical mobility diameter is about 70 nm for the Diesel vehicles and about 50s nm for the gasoline vehicles.

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DEFINITIONS, ACRONYMS, ABBREVIATIONS

ACEA	Association of European Automobile Manufacturers
CPC	Condensation Particle Counter
CVS	Constant Volume Sampler
DF	Dilution factor
DI	Direct injection
DMA	Differential Mobility Analyser
d_p	Particle diameter
ELPI	Electrical Low Pressure Impactor
EMPA	Eidgenössische Materialforschungs- und Prüfungsanstalt (Swiss Federal Laboratories for Materials Research and Testing)
IDI	Indirect injection
NA	Natural aspired engine
NEDC2000	New European Driving Cycle 2000
nm	Nanometres = $1 \cdot 10^{-9}$ metres
NO_x	pollutant comprises NO and NO_2
NSD	Number size distribution
PM	Particulate matter
ppm	parts per million
Residue	Non soluble fraction
SI	Spark ignition
SINOF	Soluble inorganic fraction
SM	Single Mode Test
SMPS	Scanning Mobility Particle Sizer
SOF	Soluble Organic Fraction
TD	Thermodesorber

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