N. Metz, G. Resch, F. Steinparzer
BMW
München
Germany

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CHARACTERIZATION OF PARTICULATE MATTER EMISSION OF MODERN DIESEL PASSENGER CARS

Dr. Ing. Norbert Metz [1], Dipl.Ing. Gerhard Resch [2], Ing. Fritz Steinparzer [2]

[1] = BMW AG München, Energy and Environment, norbert.metz@bmw.de, Fax:089-38245760

[2] = BMW Motoren GmbH, Steyr, Diesel Engine Development, gresch@bmw.co.at, Fax: 07252 888 717

ABSTRACT

Particulate matter emissions of a swirl chamber engine are compared with a high pressure injection and a common-rail engine in respect to mass, size distribution, chemical composition and morphology. Due to remarkable emission reduction especially in the last decade a careful characterization of particulate matter is necessary to respond to the new health concern of fine and ultrafine particles and to the discussion on number counting and the limitations on the gravimetrical determination of decreasing mass emission especially from modern diesel technology. The development of particulate matter mass emission as well as the development of the legislation is shown from 1980 to 2005. New diesel technology reduced the mass of PM remarkable and does not emit a larger number of particulate matter [5,6,10]. The results of this investigation show very clearly that together with the mass also the number of particulate matter is decreasing significantly. Measuring devices to characterize adequate the health effect related size classes for particulate matter are described.

1. INTRODUCTION

The successful reduction of the gaseous emissions from passenger cars [1] , [2] - both for gasoline and diesel engines - turns the attention now more to particulate matter emission not only in terms of mass but also in terms of size distribution and number. Remarkable mass reduction have been already achieved for passenger cars [3], but new discussions on the effects of ultrafine particles [4] create the need to clarify the matter. Therefore BMW has conducted an extensive investigation with 3 different combustion respective injection concepts to proof which changes in respect to mass, the size and size distribution as well as the chemical composition are caused by modern diesel technologies. The clarification is necessary because due to its better fuel economy the modern direct injecting diesel engine increases its market share and contributes therefore distinctively to the reduction of CO_2, the main greenhouse gas. To fulfill the ambitious goal of the European car industry to reduce the CO_2 -emissions in 2008 by about 25% , given as a self commitment to the European Commission [7], the contribution of diesel passenger cars is a forcing ingredient. High pressure injection engines reduce without any doubt besides gaseous also particulate matter emissions [5], [6].

2. DIESEL PASSENGER CAR PARTICULATE MATTER MASS DEVELOPMENT

Emission standards for diesel passenger cars have been remarkable tightened the last few years [8]. The PM emission standard from USA of 0.6 g/mile (ca. 0.37 g/km), valid from 1982 to 1987 was also in this time period for Europe the target, because most car manufacturers sold their diesel cars in the States as well. In relation to this standard the European EURO 3 emission standard of 0.05 g/km, obligatory in 2000 leads to a reduction of almost 90 %. The further reduction of the European PM emission standard [9] in 2005 to 0.025 g/km leads to a reduction to almost 1/20 in 18 years (1987 to 2005) [10]. Figure 1 shows the development of PM standards and PM emissions of diesel passenger cars.

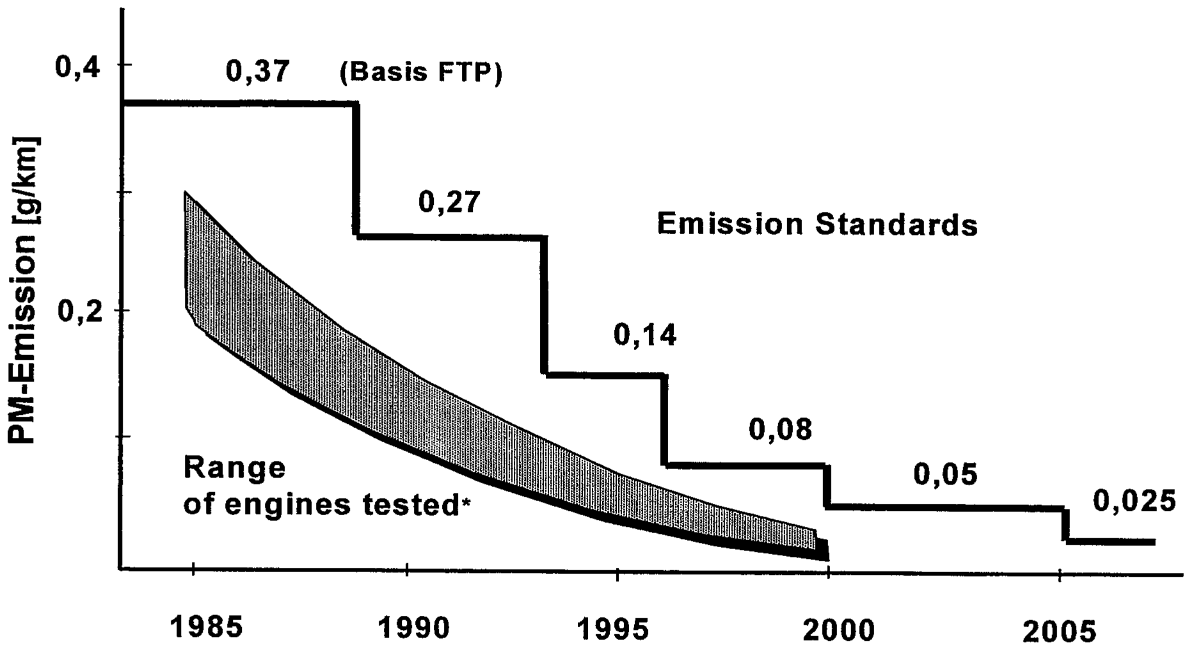


Figure 1: Development of PM standards and PM emissions of diesel passenger cars

With these improvements the German Environment Protection Agency UBA calculated the development of PM emissions of road transport - the increase in number and mileage of vehicles taken into account - up to 2010 [11]. The result is shown in figure 2. It is remarkable how successful the efforts of the automotive industry and the legislators have been. This success will also result in a better air quality in relation to TSP, PM 10, PM2.5 and soot.

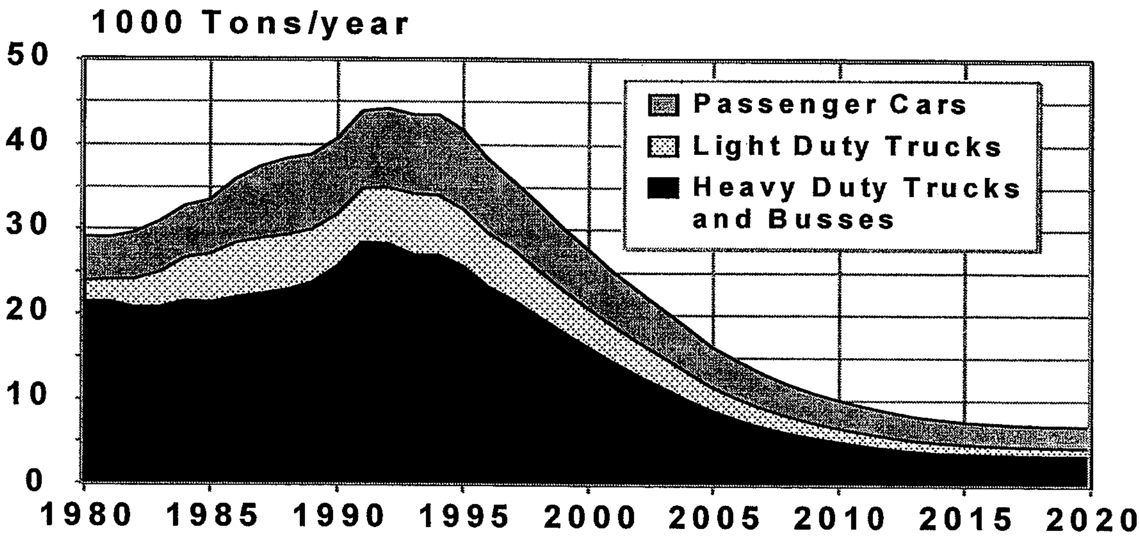


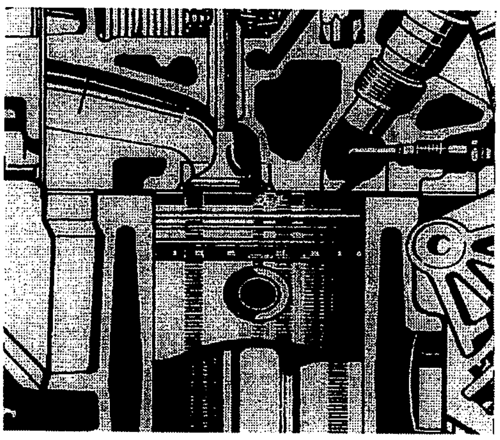
Figure 2: PM emission-development of diesel passenger cars, light and heavy duty vehicles

3. EXPERIMENTAL APPROACH

3.1 Test vehicles

The aim of the investigation was to determine if and when how the characteristics of particles changes dependent from the combustion concept, (IDI with low pressure injection and DI with high pressure injection) and from the injection concept (injection distribution pump and common-rail system).

3 BMW passenger cars 525tds, 320d and 530d presently on the market are used. All 3 passenger cars represent in their segment a fully developed engine. The 525tds sedan is used as a basis for the technology used up to now. This car is characterized through indirect injection into a swirl chamber before the main combustion chamber and a low pressure injection system with a maximal pressure of ca. 500 bar, see figure 3 has a displacement volume of 2.5 liter, a inertia weight of 1560 kg. and a power of 105 kW. Its emissions are below EU 2.



- **Swirl chamber**
- **combustion chamber divided**
- **two valves**
- **low pressure injection with distribustion injection pump**
- **Turbo charged with charge air cooler**

Figure 3: Characteristics of the IDI engine tested

The other 2 direct injection concepts - 320d and 530d - are selected to show if different injection methods will lead to different results. Both engines belongs to the same engine family with the same basic design. The 320d car- characterized through a displacement volume of 2 liter, a inertia weight of 1560 kg and a power of 100 kW, emissions below EU 3 - has a distribution injection pump while 530d - characterized through a displacement volume of 3 liter, a inertia weight of 1760 kg. and a power of 155 kW, emissions below EU 2 - has a common rail system. Both injection systems are high pressure systems, but with fundamental different injection characteristics, see figure 4.

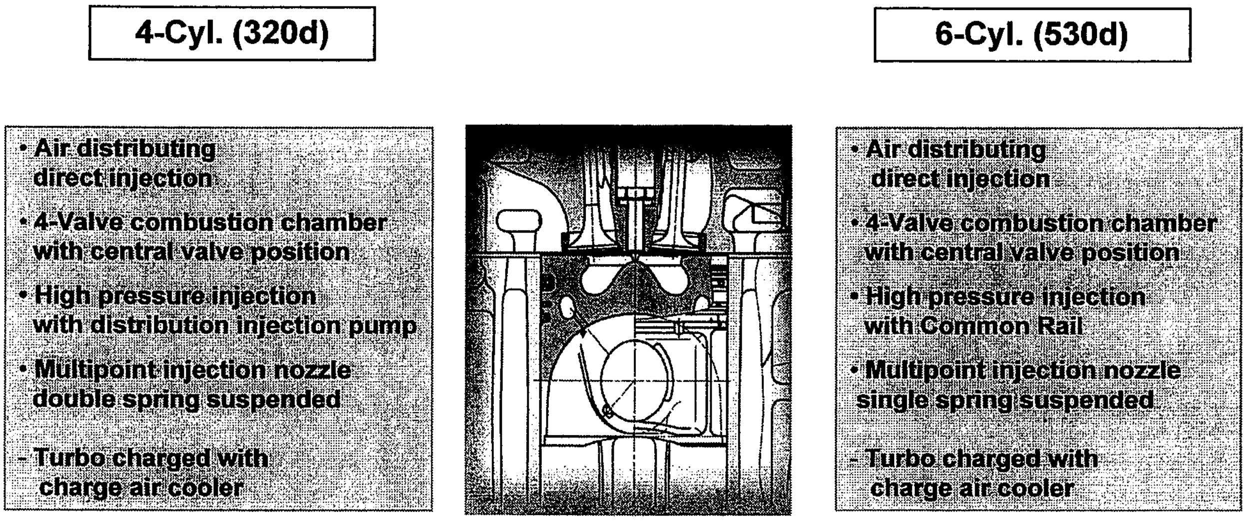


Figure 4: Characteristics of the DI engines tested

3.2 Measuring program

To ensure a stable aerosol formation - details are described in the following chapter -, the measurements have been conducted on an engine test bed, where vehicle relevant operation points were chosen. The operation point 50 km/h was chosen to come close to the driving in cities and the operation point 120 km/h - the maximum speed of the European test cycle used for certification - to be close to the driving on a European highway.

3.3 Size distribution

The PM size determines how deep particles penetrate into the respiratory system, on which site they will be deposited, which amount of material will be exhaled again, what kind of interactions will take place, by which means PM will be removed and how long the clearance time will be [12].

The typical size of a soot particle - the maximum of the number distribution - is around 100 nm. A particle with this aerodynamic diameter consists of 15 to 25 primary particles with a diameter of 15 to 40 nm. From the combustion chamber to the exhaust pipe the primary particles agglomerate to aggregate particles typically with the size range of 100 nm. The shape differs from chains to more bulky agglomerates [13].

To determine the size distribution of the polydisperse diesel aerosol it is necessary to sample the diesel particles in different size categories as a more or less monodisperse fraction. Different measuring concepts are used with different physical mechanism. Therefore the sizes depend on the measuring principle. Some examples of the measuring procedures are given in the following chapters.

3.3.1 Impactors

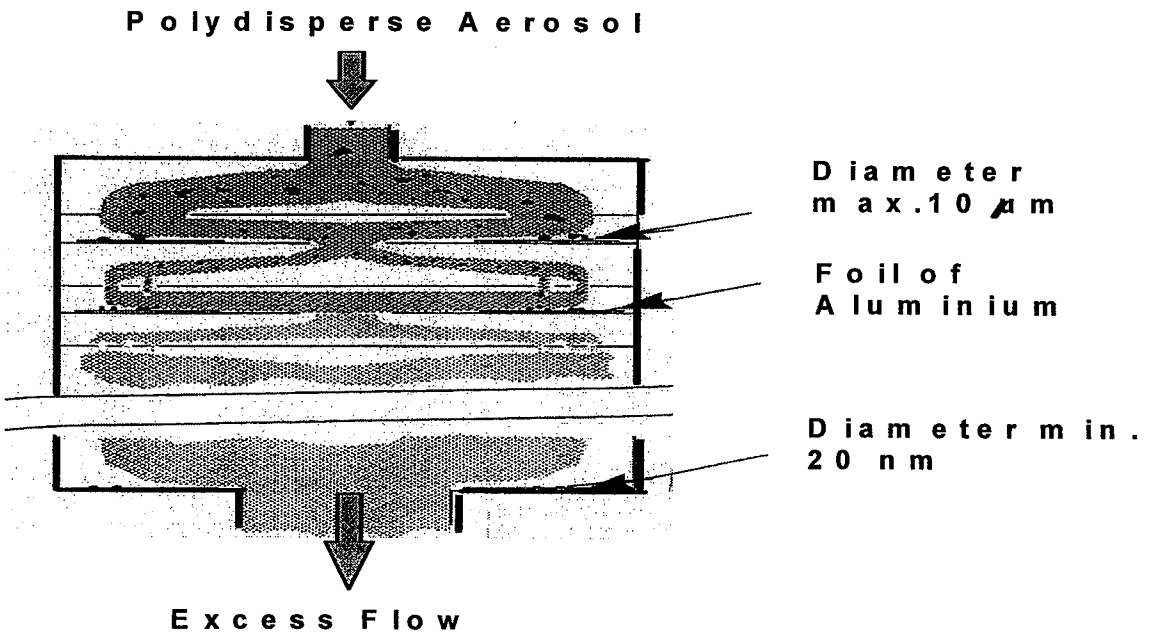


Figure 5: Impactor measuring principle

Figure 5 shows the impactor principle. The classification of the polydisperse aerosol in classes of monodisperse aerosols is caused through by a direction change of the gas stream, where larger and heavier particles can not follow the stream lines. These particles are deposited on a thin foil which can be weighted afterwards. Impactors have up to 11 stages starting with the largest size fraction, each following stage sampling with half the diameter range of the preceeding stage. The total range is 10 µm to 20 nm. This procedure classifies the aerosol by the so called aerodynamic diameter [14]. One disadvantage of the instrument is the long sampling time necessary to get enough material on the stages to ensure qualified results. Only an integral result at a stationary engine operation point or of a whole driving cycle can be obtained. An other disadvantage is the complex handling which allows only research laboratory measurements and not continuos measurements as usual for the gaseous components. The electric-low-pressure-impactor (ELPI) is derived from the cascade impactor and the complex handling with all the foils of the different cascade stages can be avoided, however even more complex calibration work is necessary to obtain meaningful results.

3.3.2 Differential Mobility Analyzer (DMA)

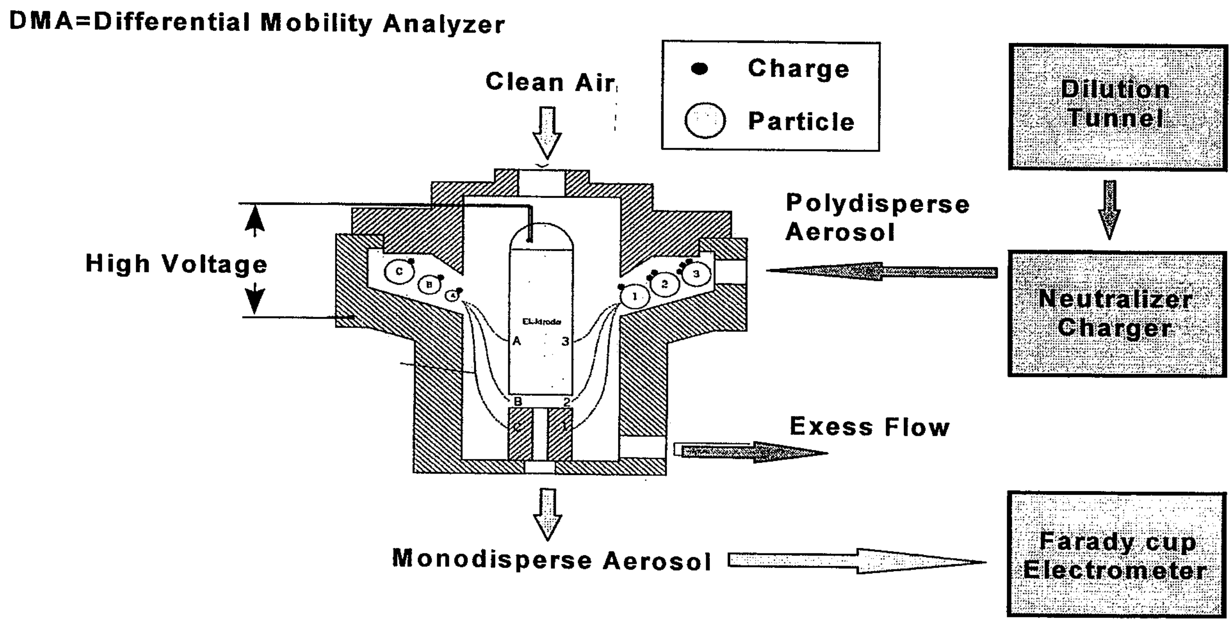


Figure 6: DMA measuring system

The principle of this instrument is different [15]. Figure 6 shows the DMA principle. The particles in the exhaust gas are charged in a bipolar ionic atmosphere before entering the DMA. In the center of the test cell is a electrode which is supplied with a variable high tension (7.5 V to 12.5 kV). Close to the outergrounded electrode a thin sheet of aerosol and clean air which fills the residue of the cylindrical channel stream downwards. On the lower end of the center electrode is a narrow ring slit, where the sample is taken. Depending on the size of the charged particles and the electric field in the channel of the DMA the particles moves radially with different drift velocities. Small particles (A) are stronger affected and collected on the center electrode, large particles (C) migrate too slowly towards the center electrode and are removed with the excess air. Only a few particles (B) fulfill the condition for entering the small annular slit. These are the particles which belong to a specific size class(nearly monodisperse aerosol) which can be further analyzed. Variation of the voltage enable the selection of different size classes. The instruments used for the investigation are varying the tension controlled. Particles with multiple charges, seen on the right side of figure 6, marked (1),(2),(3) are corrected adequately. This procedure is based on the so called electrical mobility diameter. For the determination of the number concentration of the selected size fraction the scanning mobility particle sizer (SMPS) uses a condensation nuclei counter. The electron mobility spectrometer (EMS) uses a particle size and material independent faraday cup electrometer, instead. An EMS system with two DMAs (DDMPS) developed by Prof. Georg Reischl, university of Vienna was used in the investigation discussed here. This system has the advantage of a large size range (2 to 1000 nm), a short cycle time (75 sec. per complete size distribution) and excellent resolution also for very small particles and a good reproducibility. Compared with earlier investigations this EMS is a significant progress. It must be noted that due to the complex nature the instrument is costly and need skilled expertise to be adequately operated. Since the instrument measures fast it is possible to observe real time changes of the engine and the exhausted material. It has turned out that it takes rather long time for the system to generate a stable aerosol. Especially after a load change of the engine up to 30 min. had to be waited to encounter stable aerosol conditions.

Table 1 gives an overview of commonly used instruments and shows additional an other instrument - the electrical aerosol analyzer (EAA) -, which was not used in this investigation.

Table 1: Methods to determine the size distribution from PM

Aerodynamic diameter	Electrical mobility diameter
Impactor (Low pressure Imp. Berner)	**Scanning Mobility Particle Sizer (SMPS)**
Electrical Low Pressure Imp. (ELPI)	- Charger
	- Differential Mobility Analyzer DMA
	- Condensation Particle Counter
	Electron Mobility Spectrometer EMS
	- Neutralizer
	- Charger
	- Differential Mobility Analyzer DMA
	- Faradaycup Electrometer (FE)
	Electrical Aerosol Analyzer (EAA)

3.4 Electron microscopy

The determination of the particle size with one of the instruments discussed before enables the examination of the size distribution but gives no information about the shape, the character and the construction of the particle. Therefore a transmission electroscope (TEM) [16] was used for some samples to get information to the following questions:

- Construction of the agglomerate and aggregate particles and size of the primary particles
- for different engine technologies and
- different operation points in the engine map.

These investigations were done in the „Zentrum für Elektronenmikroskopie“ Graz .

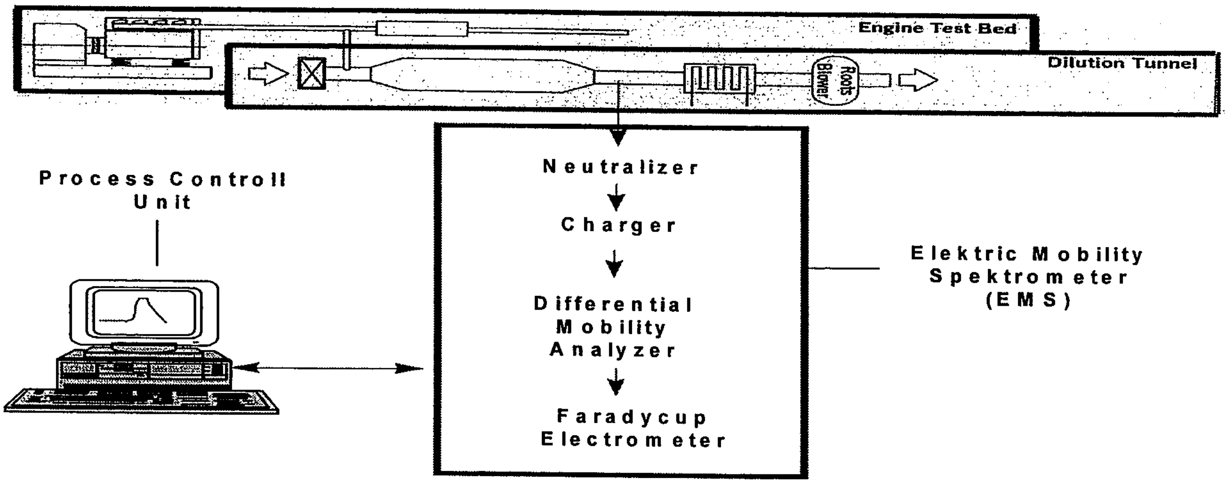


Figure 7: Experimental setup

Figure 7 shows the experimental set up [17]. A part from the exhaust gas from the is diluted in the dilution tunnel with a ratio of 10. Sampling of the probe for the EMS size distribution was done on the end of the dilution tunnel, according to the legal test procedure for certification to determine the mass of particulate matter.

4. RESULTS

4.1 Size distribution

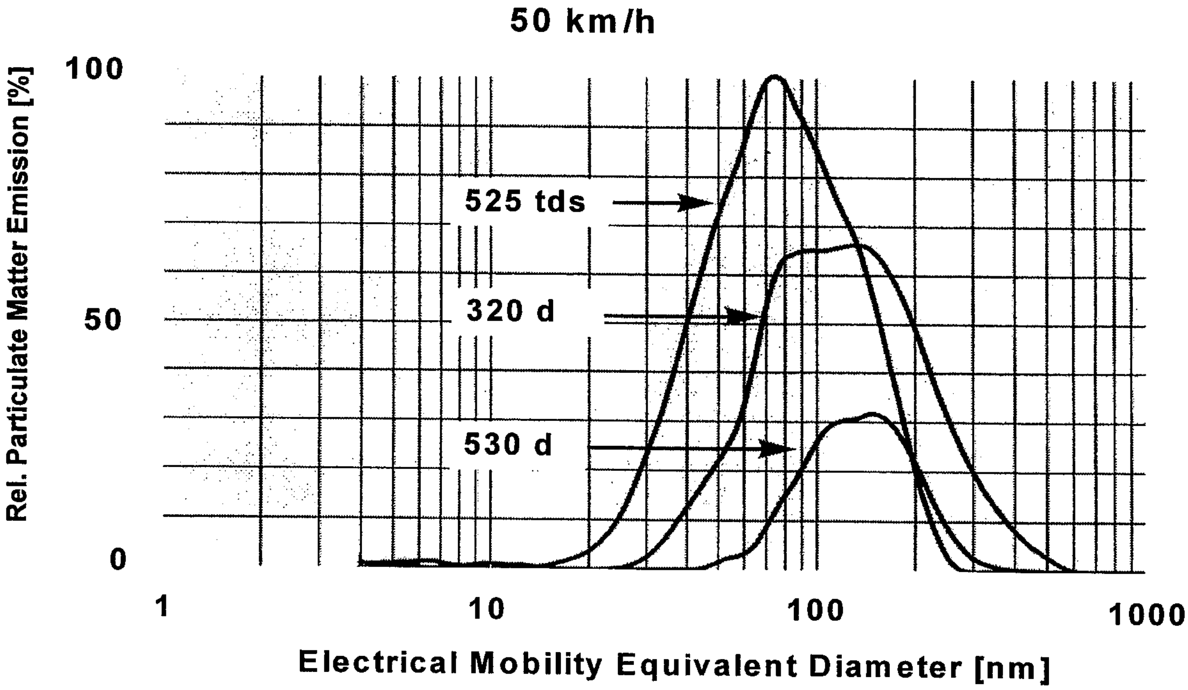


Figure 8: Particulate matter size distribution at 50 km/h

Figure 8 shows the size distribution of the 3 passenger cars at 50 km/h. On the x-axis the scale is logarithmic as usual, the y-axis is linear showing in % the change in numbers per km (100 % is the maximum number of the older swirl chamber 525 tds engine). The maximum of the number distribution for the swirl chamber engine - the old technology in this investigation - lies at 75 nm with a clear peak. The double peak with one maximum at 90 nm for the 320d - high pressure injection with a distribution pump - and the second maximum at 120 nm. For the 530 d - common rail system - the first maximum is at 110 nm and the second at 130 nm. A tendency to more agglomeration at high pressure could be an explanation, also a higher exhaust gas recirculation results in an increase in particle size. It can be seen very clearly that new technologies are not increasing the number of fine or ultrafine particles.

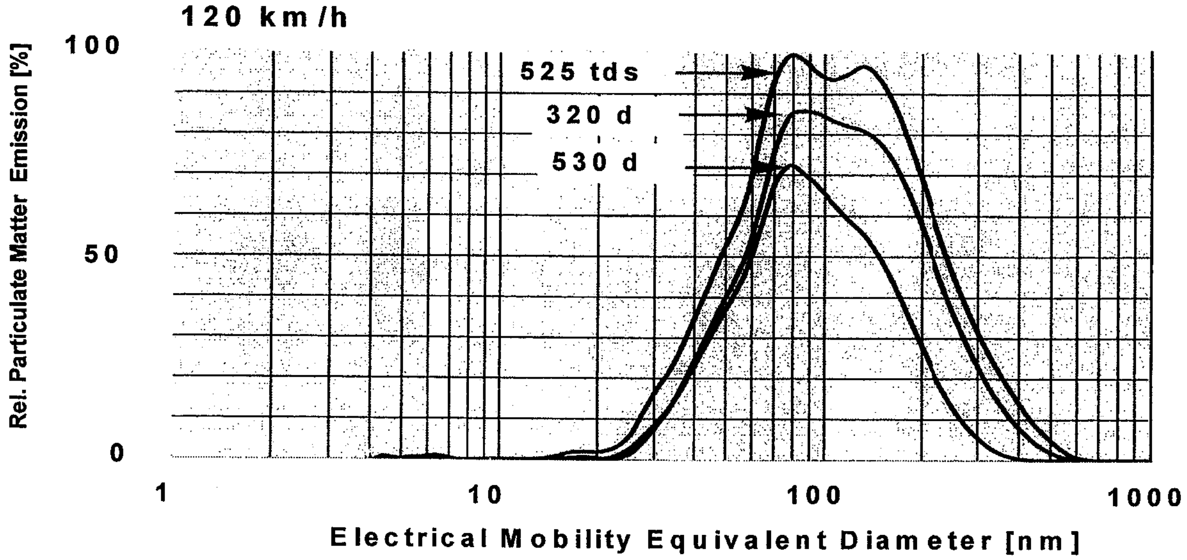


Figure 9: Particulate matter size distribution at 120 km/h

Figure 9 shows the size distribution of the 3 passenger cars at 120 km/h. The first maximum of the frequency distribution for the swirl chamber engine - (old technology) - lies at 80 nm the second peak at 140 nm. The high pressure injection concepts had also a double peak with one maximum at 90 nm for the 320d (high pressure injection with a distribution pump) and for the 530 d (common rail system) the first clear maximum is at 80 nm. Also for this driving condition it is evident that new technologies due not increase the number of fine or ultrafine particles. Table 2 summarizes the main findings of the 3 injection concepts:

Table 2: Characteristics of the size and number from 3 injection concepts

		525 tds	320 d	530 d
50 km/h				
Size range	in nm	16 - 300	28 - 600	45 -400
Maximum at	in nm	75	90, 140	110, 160
Height of the Maximums	in %	100	65	32
120 km/h		525 tds	320 d	530 d
Size range	in nm	15 - 600	23 - 550	23 - 400
Maximum at	in nm	80, 140	90	80
Height of the Maximums	in %	100	86	73

The results give an explicit answer to the question if new technologies create a higher health risk for the population. This is not the case, because new technologies not only reduces the mass of particulate matter but also reduces the number of fine and ultrafine particles as well. The suspicions that the opposite is the case have been taken from a Swiss investigation on stationary slow rotating diesel engines in the so called VERT project. There the old technology engine was optimized to low NOx-emission , which usually is obtained with a high exhaust gas recirculation rate. This lead to a strong agglomeration of the particulate matter emitted. The new technology engine in this Swiss investigation was optimized for fuel economy and had therefore a very low exhaust gas recirculation. Therefore the resulting particulate matter emitted was smaller in diameter, due to less agglomeration. It is clear that this comparison is totally misleading. To underline this size distribution results additionally electron microscopy investigations were done.

4.2 Electron microscopy

The samples for the transmission microscopy were deposited on coated copper nets. Figure 10 show structure of coating, statistical randomly holes and the adsorbed particle depositions.

525 tds, 50 km/h
swirl chamber

530 d, 120 km/h
common-rail
high pressure
direct injection

320 d, 50 km/h
high pressure
direct injection

320 d, 120 km/h
high pressure
direct injection

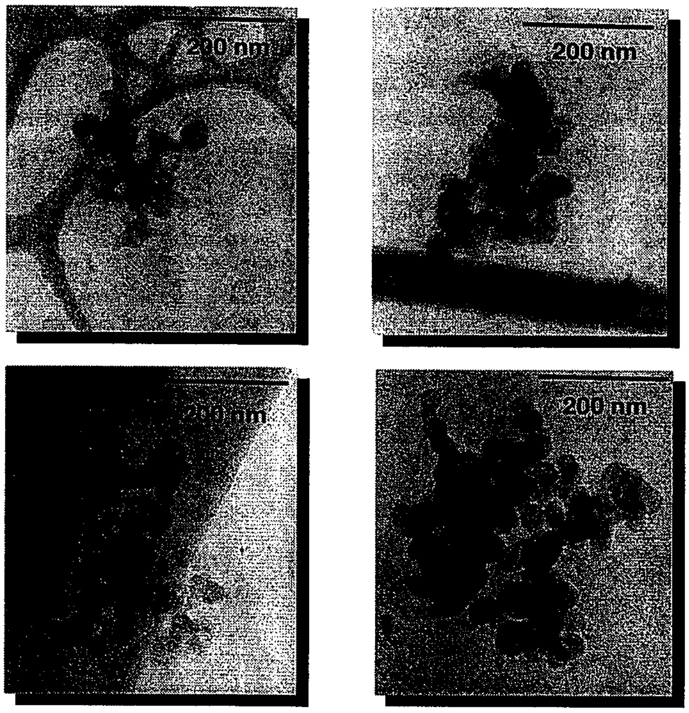


Figure 10: Morphology for the 3 cars tested

Figure 10 shows examples of results, including the size of the primary particles and the morphology of particulate matter from the 3 cars operated in different driving conditions. This pictures do not allow a judgment of the size distribution, because they represent only a tiny part of the sample grid, for which a low loading was chosen to see the primary particle size. Otherwise an artifact aggregation due to the sampling time could occur. The aggregate particle example should be seen as a single substitute out of the whole distribution curve. From the left to the right it can be seen: Particles emitted from the 525tds (old technology) at 50 km/h constant driving. The primary particles have a size between 10 to 30 nm, mostly 20 nm. To the right an example of the 530 d car (high pressure injection and common rail), 120 km/h constant driving. Here the primary particles have a size between 17 and 40 nm, mostly 30 nm. The next picture shows the 320d - (high pressure direct injection with distribution pump) - at 50 km/h driving speed. The size of the primary particles vary from 20 to 42, mostly 25 nm. The last picture on the right side shows the particles from the same engine at 120 km/h with a size range of the primary particles from 25 to 50 nm , mainly 25 nm. These examples are summarized in table 3.

Table 3: Size ranges of primary particles of the 3 engine concepts tested

passenger car type	Operation condition	Primary particle Size [nm]
525tds , swirl chamber	50 km/h	range 10 - 30, mean 20
320 d , high pressure di	50 km/h	range 20 - 42, mean 25
320 d , high pressure di	120 km/h	range 25 - 50, mean 25
530 d, hi. pres. di + c.rail	120 km/h	range 17 - 40, mean 30

4.3 Summary of the morphologic results:

The size of the primary particles of the engines tested vary between 10 and 50 nm, with a mean size range between 25 and 30 nm. No significant dependency of the size of the primary particles from the engine or the injection technology can be seen. But it can explicit be stated that new technology with high pressure direct injection do not shift the size of primary particles in the direction of smaller particles compared with the swirl chamber engine with indirect low pressure injection tested in this investigation. Morphological results assist the thesis that particles below 20 nm will not be solid soot particles, but must be liquid aerosol constituents.

4.4 Particulate composition

The composition of the particulate matter [18] was determined for 3 fractions, the soluble organic fraction of the fuel, of the lube oil and the insoluble fraction, mainly elemental carbon.

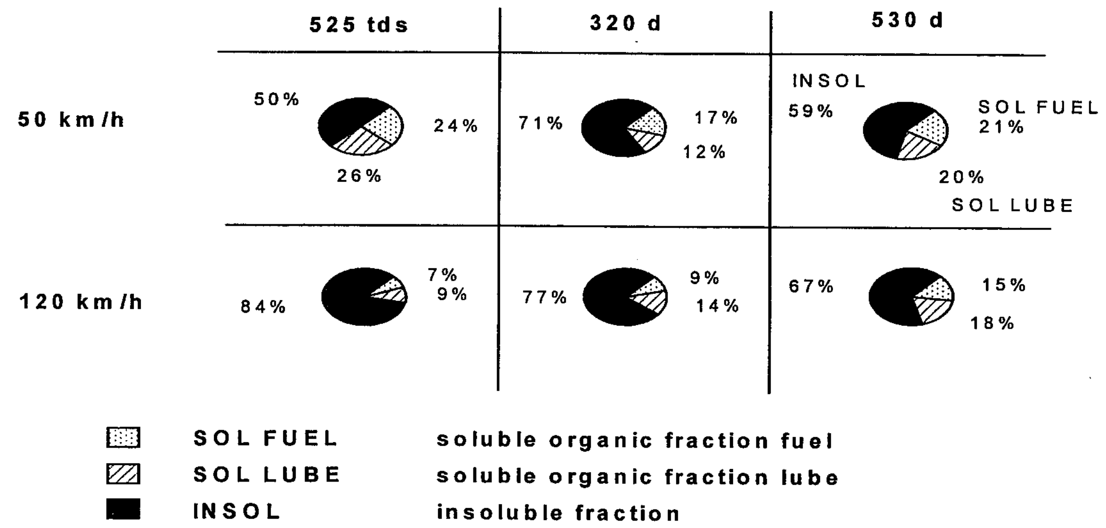


Figure 12: Chemical composition

Figure 12 summarizes the results obtained for the 3 engines and 2 operation conditions. At 50 km/h the swirl chamber engine has the lowest share of the insoluble fraction mainly due to the lower combustion temperatures and the greater surface to volume ratio at this combustion chamber concept. The differences for the 2 high pressure direct injection engines are probably caused through the operation condition in the engine map. The 320 d had a some what higher work load compared with the 530 d. This explains also the differences for the soluble organic fraction of the lube oil. Also the size of the displacement volume is contributing. Since all 3 engines were tested with the same fuel [19] no fuel effects are responsible. At 120 km/h - a operation condition with higher load and temperature - the amount of elemental carbon increases for all 3 engines but the ranking for the direct injection engines remain the same. The indirect injection swirl chamber engine has now the highest share of elemental carbon.

5. SUMMARY AND CONCLUSION

In a very cost and time intensive investigation with 3 different diesel passenger car engines the mass, size distribution, morphology and chemical composition of particulate matter were determined. Modern diesel direct injection engines including high pressure direct injection and the common rail system reduces not only the mass of particles significantly, but also reduces the number of particles in each size class. Especially elemental carbon - solid soot particles - are reduced in the same order of magnitude. No additional adverse health effects are connected with the introduction of new diesel technologies discussed in this investigation.

6. ACKNOWLEDGMENT

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