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Mass, Size, Number and Surface of diesel soot particles of DI engines with common rail

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Mass, Size, Number and Surface of Soot Particles Of DI Engines with common-rail in Diesel passenger car

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Abstract

Modern diesel engines in passenger cars emit less than 90% of the particulate matter emission compared with the technology ten years ago. Regarding the size of soot particles the maximum of the frequency distribution is about 100nm not significantly affected by different technologies, e.g. prechamber and swirl chamber concepts. The number of particles has decreased proportional to the mass of particles. The size of the primary particles, their number and the dimension of the spherical calotte from soot aggregate particles determine the surface of soot particles. Together with the mass and number also the surface of diesel particles is decreasing. The insoluble fraction of particulate matter from modern diesel technologies and modern diesel fuels - in particular low sulfur fuels – has decreased as well.

Introduction

For many years diesel vehicles are used as energy efficient, low consuming, low CO-, HC- and benzene-emitting and emission stable vehicles with relative low CO_2 values. On the other hand higher particle emission and since the introduction of the three way catalytic converter technology for gasoline engines also higher NOx emission compared with gasoline vehicles have caused an ongoing discussion in particular about the role of diesel soot in the human body. In animal experiments the effects of high particle doses have been investigated, see figure 1.

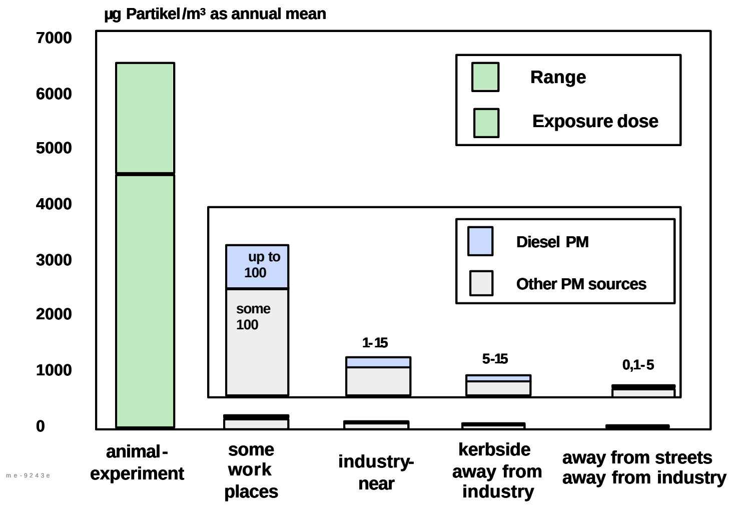


Figure 1: Particle concentration in animal experiments and in the environment [1]

With the highest dose - used in animal experiments - rats developed tumors due to overloading of the rat lung [2]. In respect to Paracelsus that the dose is responsible for the health effect [3] the mass reduction was the target up to now.

In latter years discussions about the size, the number and the surface in connection with the chemical composition have increased [4,5,6,7,8,9]. Therefore in this paper some considerations about these criteria are discussed.

Particle Mass

Due to health concerns about the role of diesel soot in the human lung already mentioned before particle mass emission of diesel vehicles during the last twenty years decreased steadily, especially from passenger cars [9], see figure 2, but also from heavy duty vehicles [7,11].

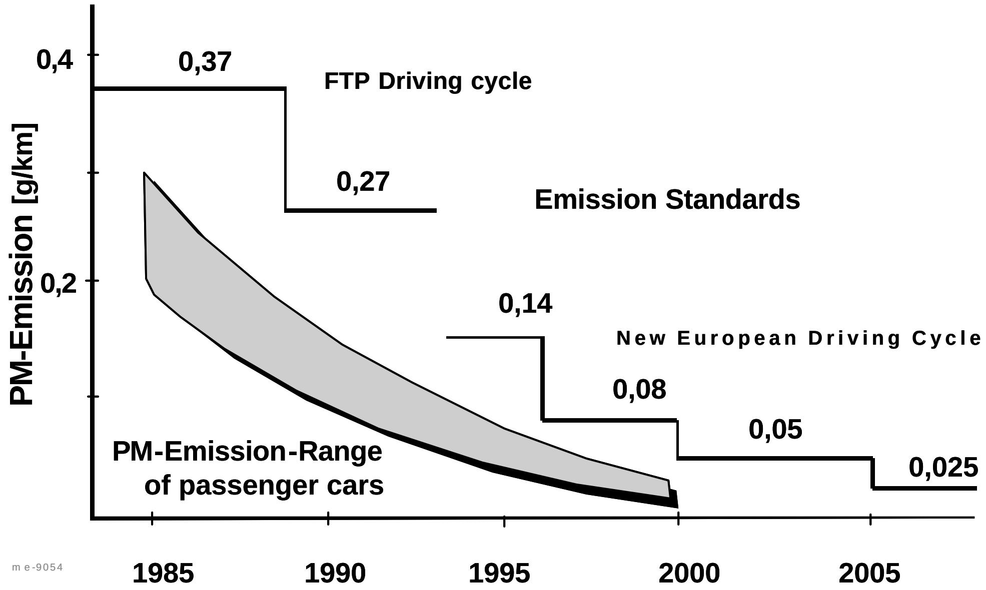


Figure 2: Development of particle emission and particle emission standards in Europe

New technologies have already reached remarkable low particle mass emissions, see figure 3.

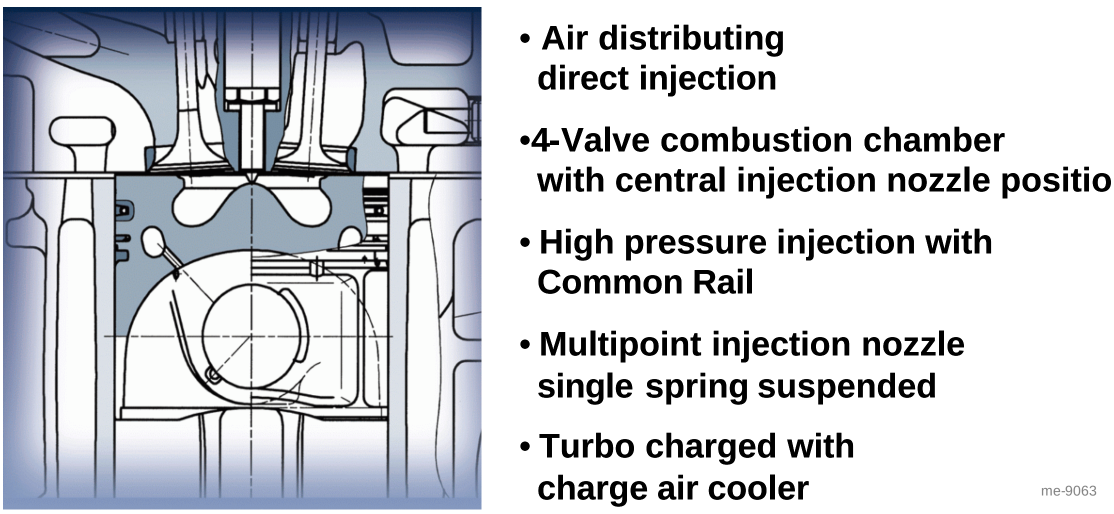


Figure 3: Combustion chamber of a direct injection common rail diesel passenger car engine

The latest and most promising technology is direct injection with a common-rail system [12], as shown in figure 4. Due to the tremendous progress in electronics and in high-pressure pumps exact timing and duration of fuel injection is the key element for low emissions.

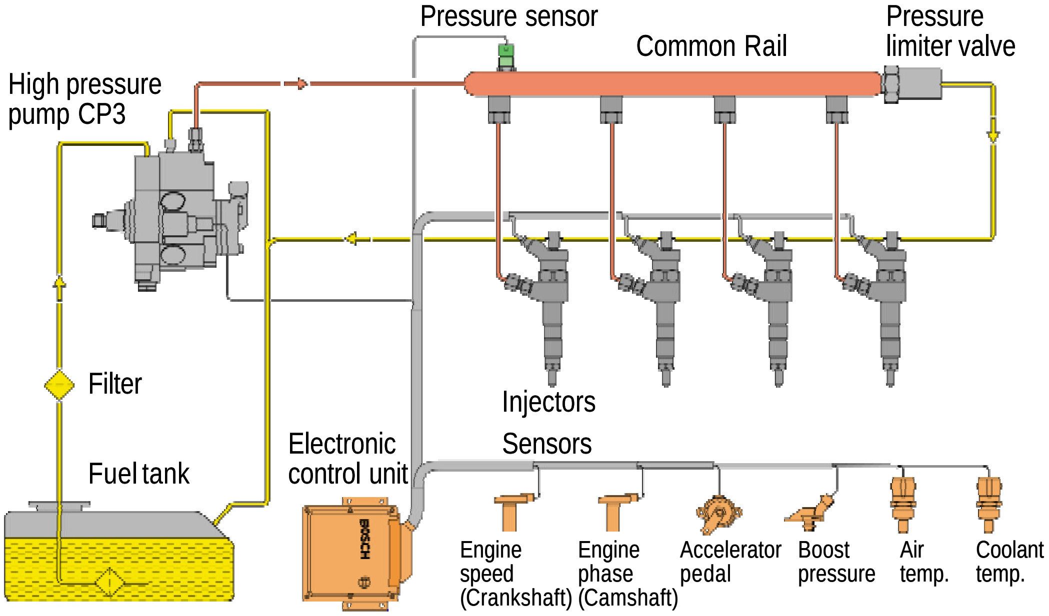


Figure 4: Principle of a direct injection common rail diesel system me-0262cr

Due to the progress in latter years improvements in respect of emissions and performance were reached so that both the environment and the customer were satisfied.

Size distribution

The penetration of particles into the human lung and the deposition within the bronchia, the lower respiratory tract until the alveolars is dependent on the size of the particles [13]. Therefore size distribution measurements are very important. These measurements have been made at the BMW diesel development department, at the university of Vienna and at the AVL Institute in Graz [10] and the results are shown in figure 5 for a constant speed of 50 km/h.

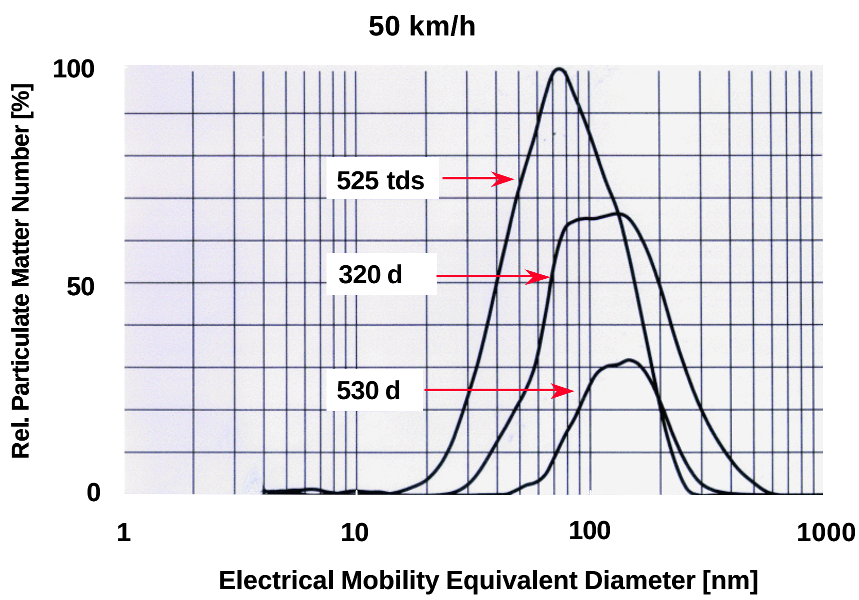


Figure 5: Size distribution of a direct injection common rail diesel system compared with other Technologies me-9070gr

The figure shows clearly that the maximum of the size distribution for the common rail system is around 100 nm with only few particles below 60 nm. Also other passenger cars show similar results, see figure 6. The measurements were undertaken by the European car manufacturer association ACEA with conventional and advanced diesel passenger cars at two speeds [14]. The diameter range here is mainly from 60 to 80 nm for advanced diesel technology. The mass range related to km was mainly between 5 mg and 40 mg.

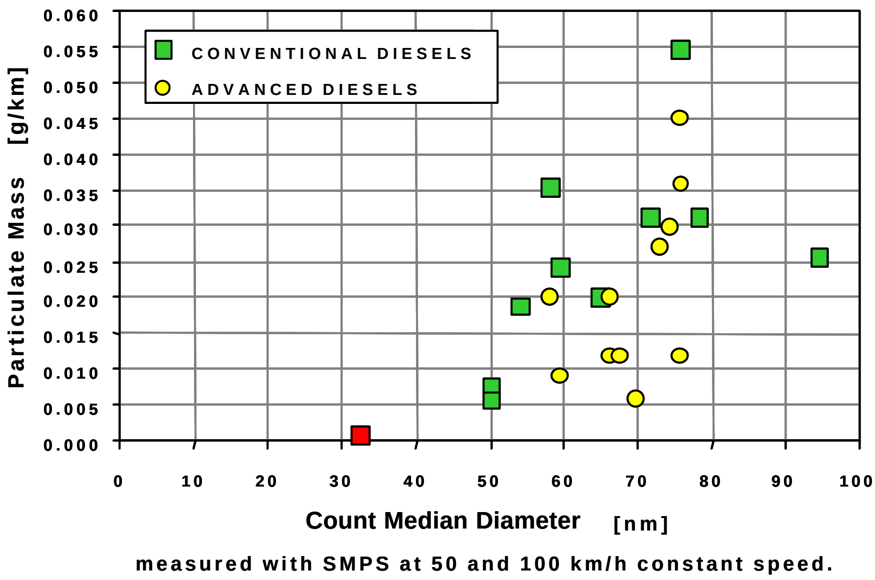


Figure 6: Mass to Size Relation of diesel soot particles me-0264

Figures 5 and 6 show clearly that vehicles with advanced diesel technology do not change the diameter significantly with decreasing particle mass.

Morphology

The electron microscopy was taken at the test bench from a BMW 530d passenger car in the dilution tunnel at a speed of 120 km/h using a Berner cascade impactor for a few minutes, to avoid aggregation during the sampling process [10].

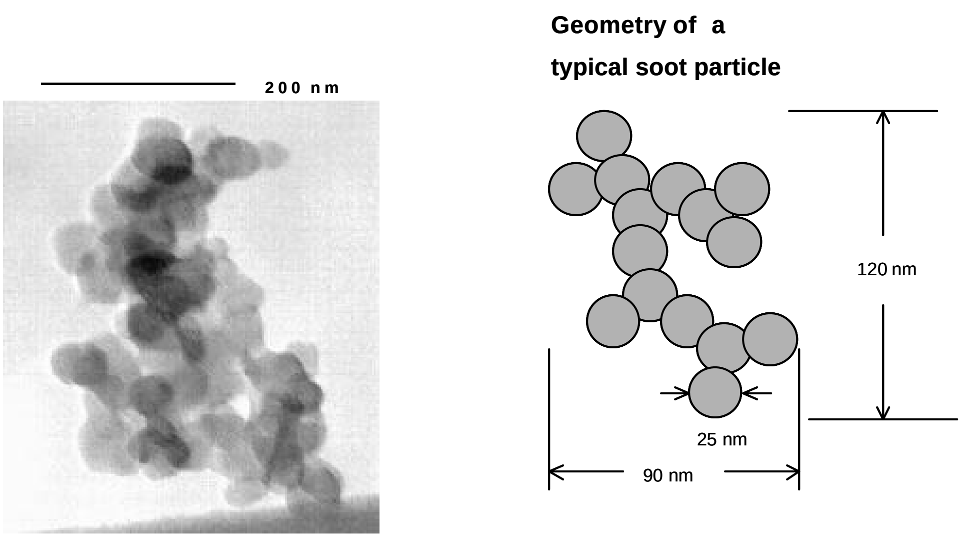
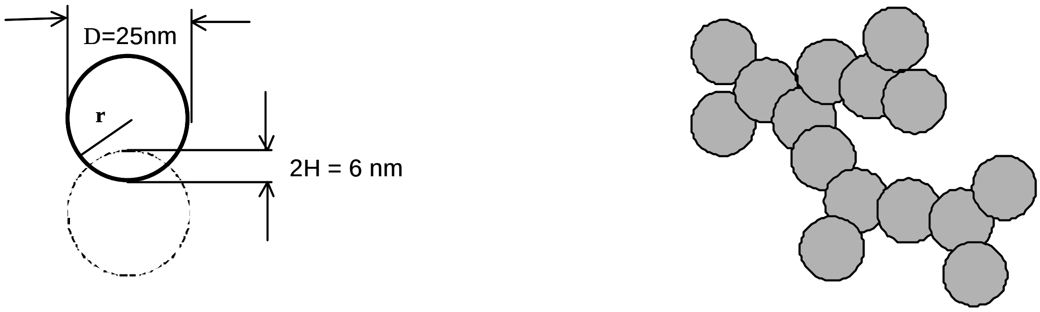


Figure 7: Morphology of diesel soot particles with a geometrical diameter of 250 nm me-0260m1

The size of this particle is about 350 nm in length and about 200 nm in width resulting in a geometrical diameter of about 250 nm. This aggregate particle consists of about 100 primary particles with a typical size of 25 to 30 nm. Primary particles hardly exist alone. The maximum of the number distribution is around 100nm. Typical aggregate particles of this size consist of about 15 primary particles, see figure 7.

For a detailed analysis morphological electron microscopy was combined with mathematical exploration of several characteristics, see figure 8.



Primary particle

Diameter of the primary particle
Diameter = $D_{\text{primary particle}}$ = 25 nm
Surface = $S_{\text{primary particle, not corrected}} = p \times D^2 = 4p \times r^2$
Volume $V_{\text{primary particle, not corrected}} = 4/3 \times p \times r^3$

$S_{\text{primary particle}} = 4p \times r^2 - 2p \times r \times H$
$V_{\text{primary particle}} = 4/3 \times p \times r^3 - p/3 \times 2 \times H^2(3r-H)$

Density (r) of soot = 1,9 g/cm^3 (10^{-15}ng/nm^3)

Weight of the primary particle = $15{,}54 \times 10^{-9}$ ng

Aggregate particle

Geometrical Diameter	= 120 nm
Aerodynamic Diameter	= 90 nm
Mobility Diameter	= ? nm

To determine the weight of the aggregate particle the spheric calottes must be extractet.
Correction after the extraction of the spheric calottes:

$S_{\text{aggregate particle}} = 15 \times p \times D^2 - 14 \times 2 p \times r \times H$
$V_{\text{aggregate particle}} = 15 \times 4/3 \times p \times r^3 - 14 \times p/3 \times H^2(3r-H)$

Weight of the aggregate particle = 218×10^{-9} ng

Me-0259pape

Figure 8: Typical diesel soot aggregate particle from the exhaust of a passenger car

Assuming that about $0.5 \cdot 10^{14}$ particles per km are emitted the resulting weight with 22 mg/km is in line with the gravimetric measured particulate emissions on the test bench.

Number

Due to the decreasing mass of particulate matter it is more and more difficult to measure the low amounts of particle emission from passenger diesel cars. To overcome these difficulties there are discussions to use the number of particles rather than the mass, to judge further improvements. Figure 9 shows the number of exhaust gas particles per km driven for different European diesel cars. It is evident that modern diesel engines emit less particle mass as well as less particle numbers.
The number of particles is different for different engines and different technologies and is also dependent on the fuel. At constant speeds the values are lowest, if speeds are changing often the number is increasing [7].

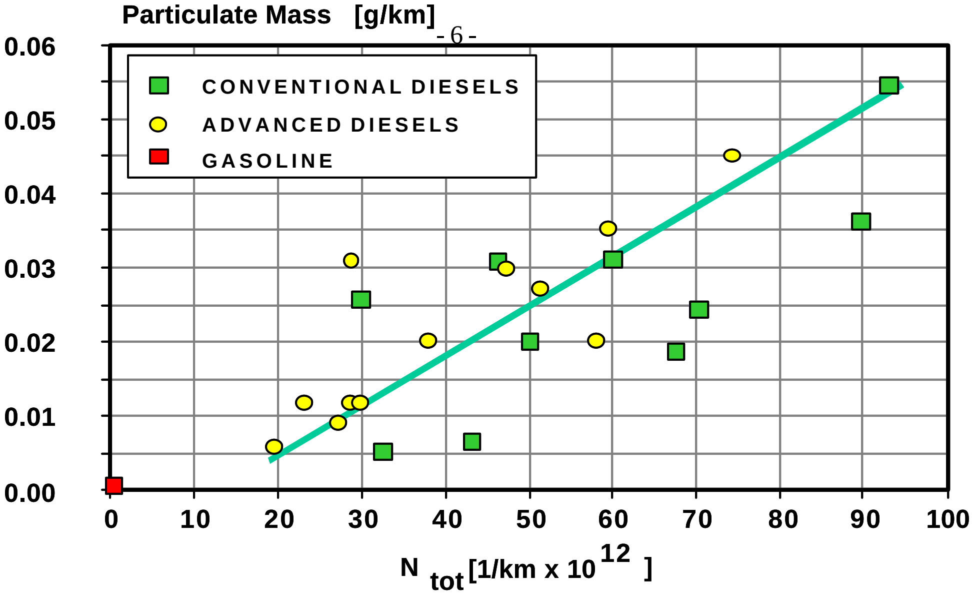
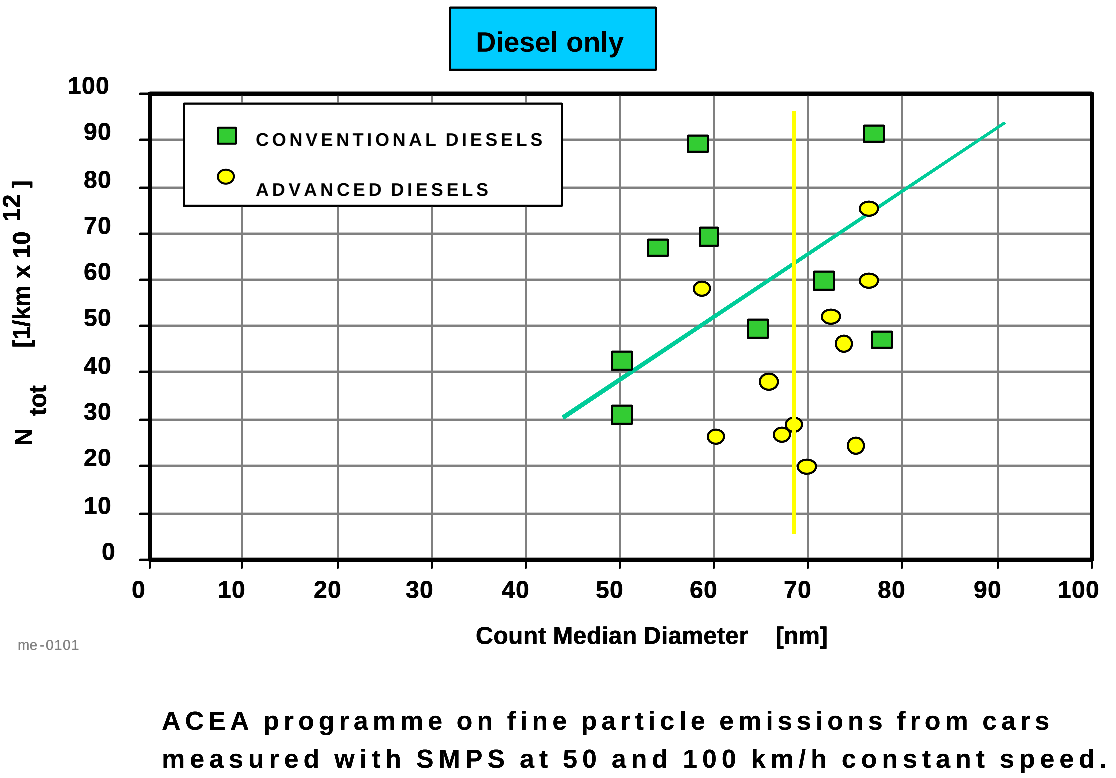


Figure 9: Mass to Number Relation of exhaust gas particles from diesel passenger cars me-0102

But it has to be clarified whether the measured particles are solid or liquid because the health effects in the human body are totally different for a solid particle from a liquid one. While solid particles of the discussed size penetrate deep into the lung, having a 30% deposition rate and a residential time of 300 days a liquid particle remains in the upper airways and is removed within a few days [13]. We presume that particles



below 30 nm are liquid.

Figure 10: Number to Size Relation of diesel soot particles me-0101

From the ACEA investigation with different passenger cars it is evident that regarding new diesel technologies there is no correlation between the number and the count median diameter [14].

Surface

The surface can be measured with the so-called BET method – named after the inventors Brunauer, Emmett and Teller - and is for soot of diesel engines in the range from 40 to 110 m^2/g. Compared with soot of flames which has a surface of 20 m^2/g it is relatively large, compared with charcoal which has a surface of 1000 to 2000 m^2/g it is small [5].

Table 1: Particle surface of diesel soot particles from passenger cars

Surface = $S_{primaryparticle, not corrected}$ = $\pi \times D^2$ = 1960 nm^2

$S_{primaryparticle, corrected}$ = $4\pi \times r^2 - 2\pi \times r \times H$ = (1960- 235) = 1725 nm^2

$S_{aggregateparticle}$ = $15 \times \pi \times D^2 - 14 \times 2 \times 2\pi \times r \times H$ = $15 \times 1960\ nm^2 - 14 \times 2 \times 1725\ nm^2$ = 22 800 nm^2

With the data and assumptions – already given - the total surface of typical diesel soot aggregate particles is 106 m^2/g. This is in line with the measured data given in the literature [4,5].

When we compared the sizes of primary particles and aggregate particles from different engine technologies we found no significant differences. Therefore with the reduction of the particle mass the resulting total particle surface has also been reduced with the new technology. Table 2 gives the resulting specific surface for different European emission standards for passenger cars [16,17].

Table 2: Particle surface for different European emission standards

European Emission standards for			with a specific surface of 106 m^2/g
Euro 2	0,08 g/km	resulting in a	total surface of 8,48 m^2/km
Euro 3	0,05 g/km	resulting in a	total surface of 5,30 m^2/km
Euro 4	0,025 g/km	resulting in a	total surface of 2,65 m^2/km.

Volume and Weight

The volume of soot particles can be calculated as already shown before, see table 3:

Table 3: Particle volume for diesel soot particles of passenger cars

Volume $V_{primary\ particle,\ not\ corrected}$ = $4/3 * \pi * r^3$ = 8180 nm^3

$V_{primary\ particle,\ corrected}$ = $4/3 \times \pi \times r^3 - 1/3 \times \pi \times H^2 (3r-H)$ = (8180- 355) = 7825 nm^3

$V_{aggregate\ particle,\ corrected}$ = $15 \times 4/3 \times \pi \times r^3 - 14 \times 2 \times 1/3 \times \pi \times H^2 (3r-H)$ = 113 610 nm^3

The weight can be determined with the volume and density by calculation, see table 4:

Table 4: Particle weight of total diesel soot aggregat particles

Weight of a primary particle, not corrected in ng	$15,54 \times 10^{-9}$
Weight of a primary particle, corrected in ng	$14,48 \times 10^{-9}$
Weight of a aggregate particle, corrected in ng	216×10^{-9}
Number of particles /km averaged for 100nm	$0,5 \times 10^{14}$

Particle Weight averaged for 100 nm / km 11 mg / km

The weight of the particle fraction above 100nm – an agglomeration of more than 200 primary particles - contributes to the total weight about 15 mg/km. Therefore the calculation of the newest technology compared with the emission standards for passenger cars valid in the European Union seems reasonable, see table 5.

Table 5: Particle mass for different European emission standards

Euro 2	0,08	g/km	or	80 mg / km
Euro 3	0,05	g/km	or	50 mg / km
Euro 4	0,025	g/km	or	25 mg / km

Chemical composition

The common rail technology (530d) has been compared at two speeds with two other BMW passenger car technologies – high pressure injection (320d) and swirl chamber (525 tds) – to get some information about the chemical composition as well [10].

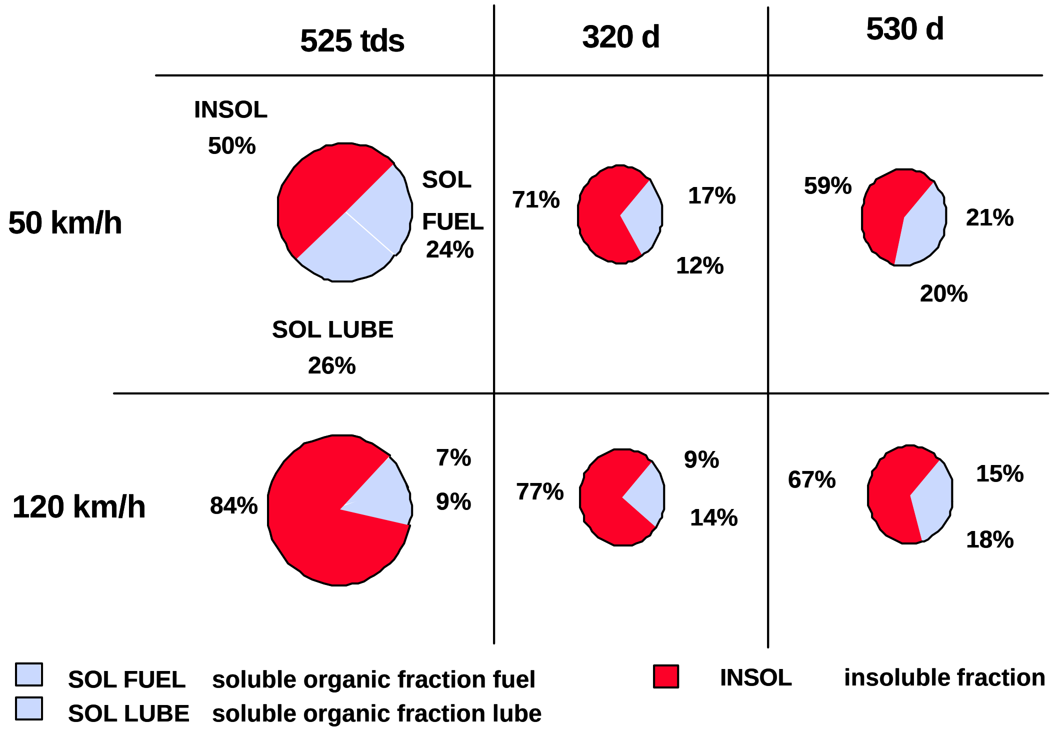


Figure 11: Chemical composition of particulate matter from different diesel technologies

Whilst higher speed increases the insoluble fraction, the soluble fraction particular of the fuel decrease. Compared with older technologies the share of the insoluble fraction at 120 km/h is lowest. Due to the good results on the solid particle side the soluble organics fraction of the fuel and the lube is higher for the common-rail-system.

Summary and conclusion

The particulate mass emission of modern diesel engines in passenger cars especially in recent years has decreased substantially. The size of soot particles is small enough to penetrate into the alveolar region of the lung. With a size of about 100nm however – where the main portion of particles is emitted – they are big enough to reach almost the minimum deposition in the human lung. The size of particles from modern diesel injection concepts is as large as the older prechamber and swirl chamber concepts. The number of particles is proportional to the mass of particles and therefore with the reduced mass also the number of particles has decreased. The surface of a soot particle is determined through the size of the primary particle, the dimension of the spherical calotte and the number of primary particles forming a typical soot aggregate particle. Investigations at BMW and within the ACEA programme show without any doubt that together with the mass also the surface of diesel particles is decreasing. The chemical composition of modern diesel technologies and modern diesel fuels - in particular low sulfur fuels – will decrease the insoluble fraction as well.

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