Emissions of particulate matter from diesel engines
- Determination of the particle number concentration in diesel exhaust gas and
- Emissions of heavy-duty diesel engines with focus on particulate matter

Yvonne Ruschel[1], Olaf Schröder[1], Jürgen Krahl[1,2], and Axel Munack[1]

Abstract

Particulate matter (PM) has become perceived as one of the major harmful emissions from diesel engines. PM emissions are subject to diesel engine emission legislations worldwide. Epidemiological and toxicological studies have indicated that adverse health effects increase with decreasing particle size and increasing particle number. Therefore, the determination of the particle number distribution is of same importance as the assessment of the total mass of PM. In the first part of this paper, measurement techniques of particulate matter and two instruments for determination of particle number concentration and particle size distribution in diesel exhaust gas are introduced. The second part gives an overview of particulate emissions of a heavy-duty diesel engine running on four different fuels.

Keywords: Biodiesel, diesel fuel, particulate matter, ultrafine particles, health effects, ELPI, SMPS

Introduction

Particulate Matter (PM) has become perceived as one of the major harmful emissions from diesel engines. PM emissions are subject to diesel engine emission legislations worldwide. According to the U.S. Environmental Protection Agency (EPA), particles are defined as all solid or liquid substances in diluted exhaust gas that can be collected on a filter at a temperature of under 51.7 °C (which is 125 °F) (Code of Federal Regulations). The temperature reduction of the exhaust gas is achieved by diluting the exhaust gas with air.

PM consists of a variety of organic and inorganic substances. The main constituents of the organic substances are unburned or partially burned fuel and lubrication oil compounds. The inorganic substances include soot (carbon), sulphates, water and metallic compounds. Metal and rust particles from the engine or the exhaust gas system as well as derivates of organo-metallic fuel and lubricant additives are included in the metallic compounds. The percentage of these substances in the total particle mass depends on a multitude of parameters. In addition to constructive parameters such as design of the combustion chamber and the injection system, the mode of operation, or rather the overall load configuration, the fuel and lubricant quality as well as the wear of the engine are also included here (Wachter F. and Cartellieri W. P. 1987).

Recently emissions of fine and ultrafine particles from diesel engines induced a broad discussion in Germany. Since January 2005 the limit for the emission of fine particles has been tightened in the context of a council directive of the European Union (99/30/EG).

According to this directive the limit of 50 µg/m³ particles in ambient air should not be exceeded. The compliance of the limit cannot be guaranteed yet all over in Germany.

In 2006, an exceedance of the limit on more than 35 days per year occurred at 100 of the 450 measuring stations.

In detail, particles from diesel engines show a bimodal size distribution consisting of a nuclei mode and an accumulation mode as can be seen in figure 1.

[1] Federal Agricultural Research Centre, Institute for Technology and Biosystems Engineering, Braunschweig, Germany

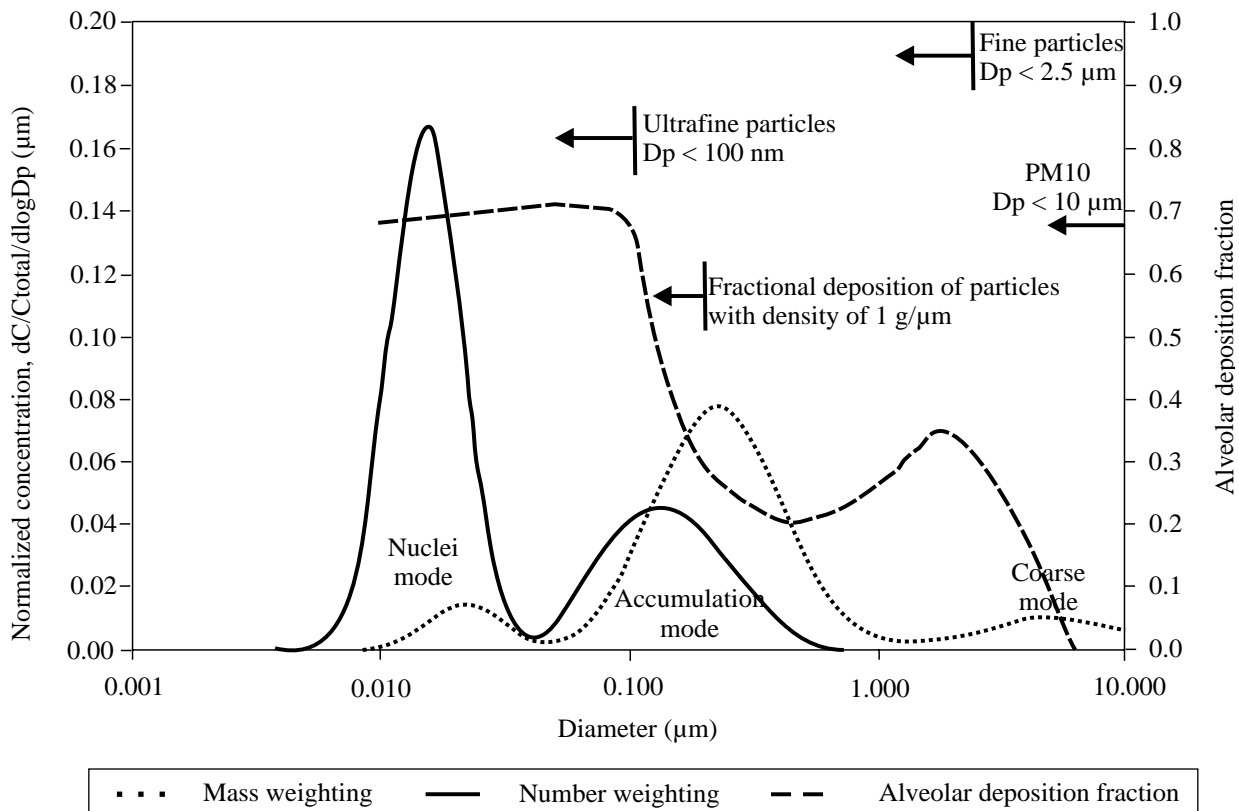[2] University of Applied Sciences Coburg, Coburg, Germany

Figure 1:

Typical diesel engine exhaust particle distribution by mass, number and alveolar deposition (Kittelson D. B. 2000)

Nuclei mode particles, which can depose deep in the lungs, dominate the majority of particles but have only little effect on the total mass. Epidemiological and toxicological studies indicated that adverse health effects from exposure to PM may increase with decreasing particle size even if the particles consist of toxicologically inert materials (Seaton A. et al. 1995, Donaldson K. et al. 2000). Ultrafine particles can penetrate the alveolus region, resting there for months. Here the particles can cause allergic or inflammatory reactions leading to bronchitis and asthma (Mayer A. 2001). Therefore the determination of the particle number distribution is just of the same importance as the PM mass that is subject to regulation. Consequentially, the particle number is currently under discussion as future regulated value for Euro 6 legislation.

Beneath results on emissions this paper presents briefly two instruments for the determination of particle number concentration in diesel exhaust gas as well as the sampling procedure for PM. A scanning mobility particle sizer (SMPS) detects particles based on their electrical mobility. SMPS enables the determination of particles in the size range between 10 to 300 nm. A low pressure impactor (ELPI) detects particles between 30 nm and 10 μm. Fur-

thermore, the overall particulate mass (PM) can be determined gravimetrically.

The presented particulate emissions data of a heavy-duty diesel engine running on four different fuels were obtained be applying all three measurement methods.

Material and methods

Test Engine

Studies were carried out at the emission test stand of the Institute for Technology and Biosystems Engineering at the Federal Agricultural Research Centre (FAL) in Braunschweig, Germany. A Mercedes-Benz engine OM 906 LA (Euro 3) with turbocharger and intercooler was used. The engine was equipped with a pump-line injector system. It was operated without exhaust gas recirculation. Table 1 presents some of the engine characteristics.

Table 1:

Technical data of the test engine Mercedes-Benz OM 906 LA

Piston stroke	130 mm
Bore of cylinder	102 mm
Number of cylinders	6
Stroke volume	6370 cm³
Rated speed	2300 min⁻¹
Rated power	205 kW
Maximum torque	1100 Nm at 1300 min⁻¹
Compression ratio	17.4

Exact engine load during test cycles is guaranteed by crankshaft coupling to a Froude Consine eddy-current brake. Engine test runs were in accordance with the 13-mode European Stationary Cycle (ESC), for which the preset torque and revolution rates (related to maximum load at the actual speed or related to rated speed, respectively) as well as the time courses of engine torque and speed are displayed in subsequent figures 2 and 3.
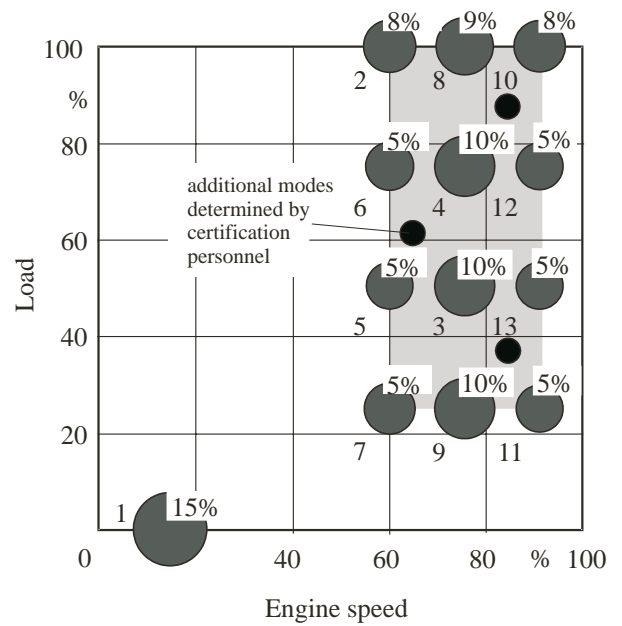

Figure 2:
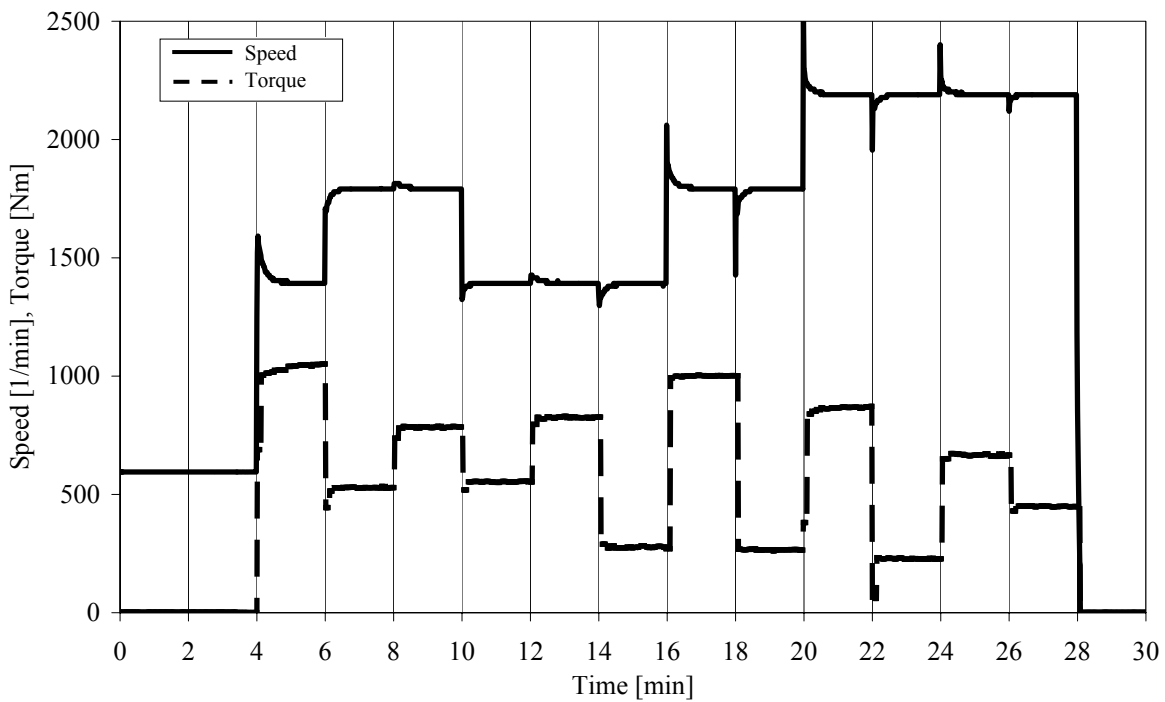
Modes of the ESC test cycle

Figure 3:

Exemplary courses of engine speed land torque for an ESC test cycle

In previous investigations higher emissions of particles in the nuclei mode were detected for RME compared to other fuels (Munack A. and Krahl J. 2005; Herbst L. et al. 2006; Ruschel Y. et al. 2006). Therefore, RME was compared to two common diesel fuels and one artificial blend. The following four fuels were used for engine operation:

DF: Reference diesel fuel
RME: Rapeseed oil methyl ester
B5Ult: 5 % RME + 95 % Aral Ultimate Diesel®
V-Power: Shell V-Power Diesel®

RME was provided by Carl Büttner Mineralöl-GmbH in Leer; DF from Haltermann Products BSL Olefinverbund GmbH in Hamburg. Aral Ultimate Diesel® and Shell V-Power Diesel® were obtained from public filling stations in Braunschweig.

Particulate measurement

The emissions of particles were determined by three different ways: The total mass of particulate emissions was measured gravimetrically according to the EU regulation (EU, 2005). Particle number and particle size distributions were measured with ELPI and SMPS.

Particulate Matter

PM measurements were accomplished after partial flow dilution of raw exhaust gas in a dilution tunnel (figure 4), which cools down the exhaust gas to under 51.7 °C. A dilution factor of about 10 is applied for determination. Dilution factors are calculated from separate recordings of CO_2 contents in exhaust gas, fresh air, and diluted exhaust gas.

The particles are collected on a two stage filter. Particle mass was determined gravimetrically after deposition to teflon-coated glass fibre filters (T60A20, Pallflex, diameter 70 mm).

The total volume (V_{SAM}) that is led through the filter results from the requirement that the sampling time for each testing phase must be at least four seconds per 0.01 weighting factor. This must also take place as late as possible and may not be concluded earlier than five seconds before the end of the phase. For sampling a mass flow controller is set in such a manner that it is in accordance with the individual weighting factors of each mode of the ESC test. Each sampling takes 60 seconds and ends three seconds before the end of the mode. In this time the dilution level is constantly monitored and the exhaust gas sample volume can be corrected through a lengthening or shortening of sample time if the dilution factor changes. At the end the weighting factor that results from the following equation must be kept at ± 7 % to the weighting factor of the ESC-test.

$$\frac{V_{SAM,i} \cdot \sum_i \left(V''_{EDF,i} \cdot WF_i \right)}{V_{SAM} \cdot q_i \cdot V''_{EDF,i}} \overset{!}{=} WF_i$$

with

V_{SAM}:   Total volume of samples
$V_{SAM,i}$:   Volume of sample in mode i

$V''_{EDF,i}$: Volume flow of exhaust gas in mode i
WF_i: Weighting factor of mode i
q_i: Exhaust gas dilution factor in mode i.

Weights of fresh and sampled filters were determined to an accuracy of ± 1 μg by means of a microbalance M5P from Sartorius, always preceded by at least 24 hours of
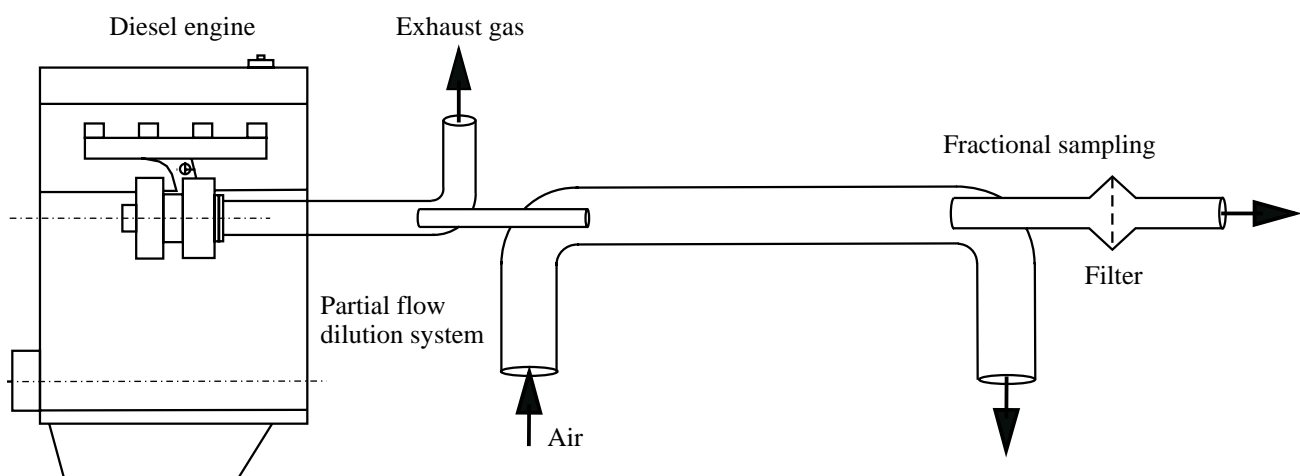

Figure 4:

Schematic diagram of the exhaust gas dilution tunnel

conditioning in a climate chamber held at 25 °C and 45 % relative humidity.

The particulate emission can then be calculated as

$$PT = \frac{M_{PF} \cdot \sum_{i}\left(V''_{EDF,i} \cdot WF_i\right)}{V_{SAM} \cdot \sum_{i}\left(P_i \cdot WF_i\right)}$$

with

PT: Specific particulate emission

M_{PF}:   Total mass on the particle filter

V_{SAM}:   Total volume of samples

$V''_{EDF,i}$:   Volume flow of exhaust gas in mode i

WF_i: Weighting factor of mode i

P_i: Power in mode i.

Particle size and number distribution

Determination of the particle size distribution is carried out by sampling with a multi-hole probe at the end of the exhaust gas dilution tunnel using a scanning mobility particle sizer (SMPS) system from TSI company. Secondary dilution is required to avoid an overloading of the measurement instrument.

The SMPS consists of a Kr-85 bipolar charger, an electrostatic classifier, both placed in the differential mobility analyzer (DMA), and a condensation particle counter (CPC). Before the sample aerosol flows through the DMA, particles larger 300 nm are removed by an impactor which is located in front of the DMA. The polydisperse aerosol is first led to a Boltzmann charge distribution by beta emission from the [85]Kr source (Liu B. Y. H. and Pui D. Y. H. 1974; Wiedensohler A. 1988). After passing the charger the aerosol is conducted into the electrostatic classifier. Figure 5 shows a general scheme of the DMA.

The electrostatic classifier consists of two concentric cylinders. The outer cylinder is grounded. Between the cylinders flows a sheath gas of filtered air. The charged sample aerosol is introduced along the inner wall of the outer electrode. The positively charged particles are accelerated toward the inner electrode by the electric field. As a result of the deflection the positively charged particles are dragged through the sheath air. Consequently, they quickly reach a terminal radial velocity that depends on the electrical mobility of the particles, that is independent of mass and density (Willeke K. and Baron P. A. 1993, Maricq M. M. et al. 2000, Hinds W. A. 1989). Depending on the voltage applied across the cylinders, only particles within a narrow range of electrical mobility, and therefore diameter, can pass through the exit aperture of the classifier (Knutson E. O. and Whitby K. T. 1975, Maricq M. et al. 2000). Afterwards the monodisperse aerosol is detected in the CPC (figure 6).
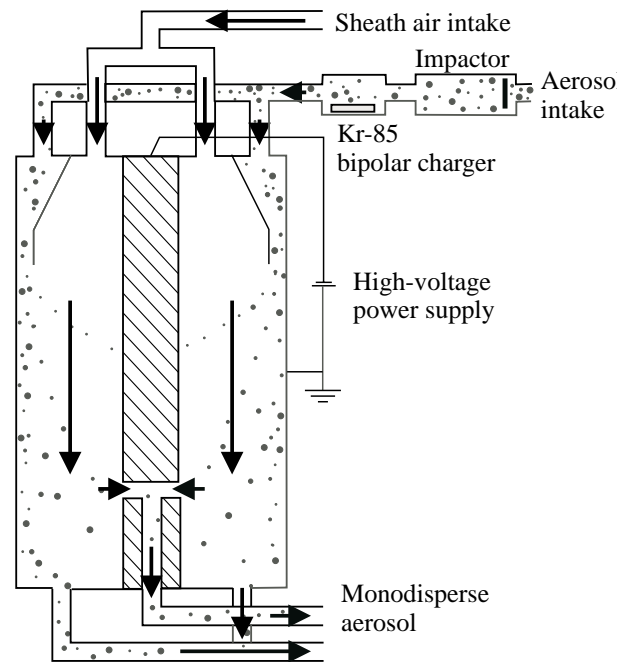

Figure 5:

Schematic of the differential mobility analyser (DMA)

Because particles of diesel exhaust gas are to small for direct optical detection methods they are first enlarged inside the CPC. This is achieved by surface condensation of supersaturated n-butyl alcohol, such that they increase quickly in size (up to about 10 µm). The enlarged particles are counted using laser diode light. The measurement signal used for counting is the scattered light of the single particle (Mayer A. 2001, ACEA 2002).

As alternative to the SMPS technique, the Finnish company Decati Ltd. offers an electrical low pressure impactor (ELPI). For the determination of the particle size distribution samples are taken with a multi-hole probe at the end of the exhaust gas dilution tunnel.

The principal components of the ELPI include a corona discharge, a Berner low pressure impactor, and a series of electrometers. The general scheme of the ELPI is shown in figure 7.
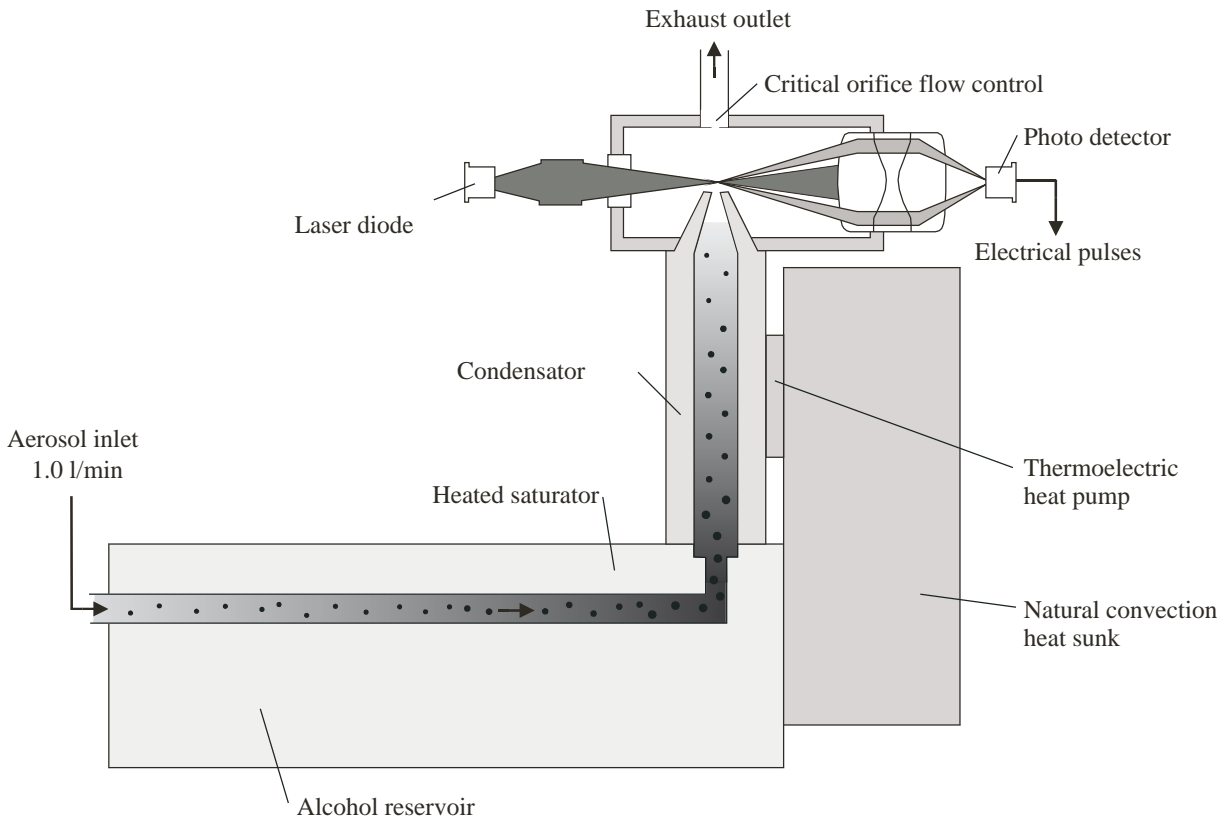
Exhaust outlet

Critical orifice flow control

Photo detector

Laser diode

Electrical pulses

Condensator

Thermoelectric
heat pump

Aerosol inlet
1.0 l/min

Heated saturator

Natural convection
heat sunk

Alcohol reservoir

Figure 6:

Schematic of the condensation particle counter (CPC)

Sample

Corona
charger

Flush pump and filter

External PC
or laptop

HV and power source

Impactor

Electro-
meters
and A/D

Internal
PC

(External
in/out)

Pressure
sensor

Controls
and
LCD display

Vacuum-
pump
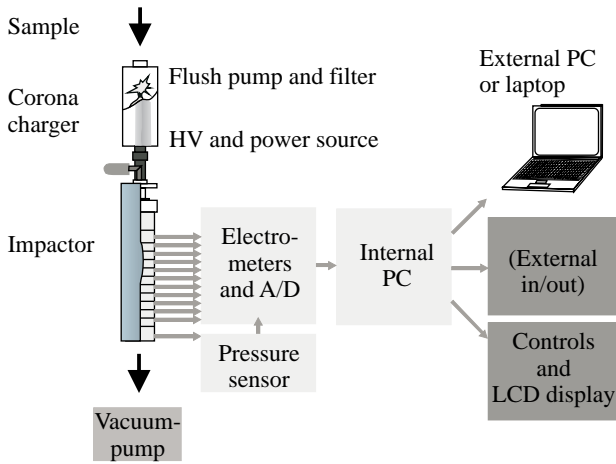
Figure 7:

Schematic of the electrical low pressure impactor (ELPI)

The diode type corona discharge provides unipolar charging of the particles in the sample aerosol. It is coupled with a trap, which has the function to remove ions and small (< 20 nm) particles that can otherwise produce extraneous currents at the electrometers. The amount of charge accepted by the particles increases with their mobility diameters.

After being charged the aerosol passes through a thirteen-stage cascade impactor, where the particles are separated in accordance to their aerodynamic diameter. The thirteenth stage removes particles larger than 10 µm. Each of the twelve lower stages is connected to an electrometer that measures the current deposited from particle impaction. The charge on each stage, resulting from particle impaction, is proportional to the number of impacted particles. With a dynamic response of 1 second, the ELPI is able to follow a transient driving cycle (Mayer A. 2001, ACEA, 2002).

Results

PM

PM emissions of the tested fuels are presented in figure 8. The EURO 3 limit of 0.1 g/kWh was met by all four fuels. The lowest emissions were obtained for RME. The highest emissions were determined for DF. The emissions of the two fuels B5Ult and V-Power lay between RME and DF. The value for B5Ult was lower than for V-Power. By use of RME it was possible to obtain reductions in the particle emissions versus DF.
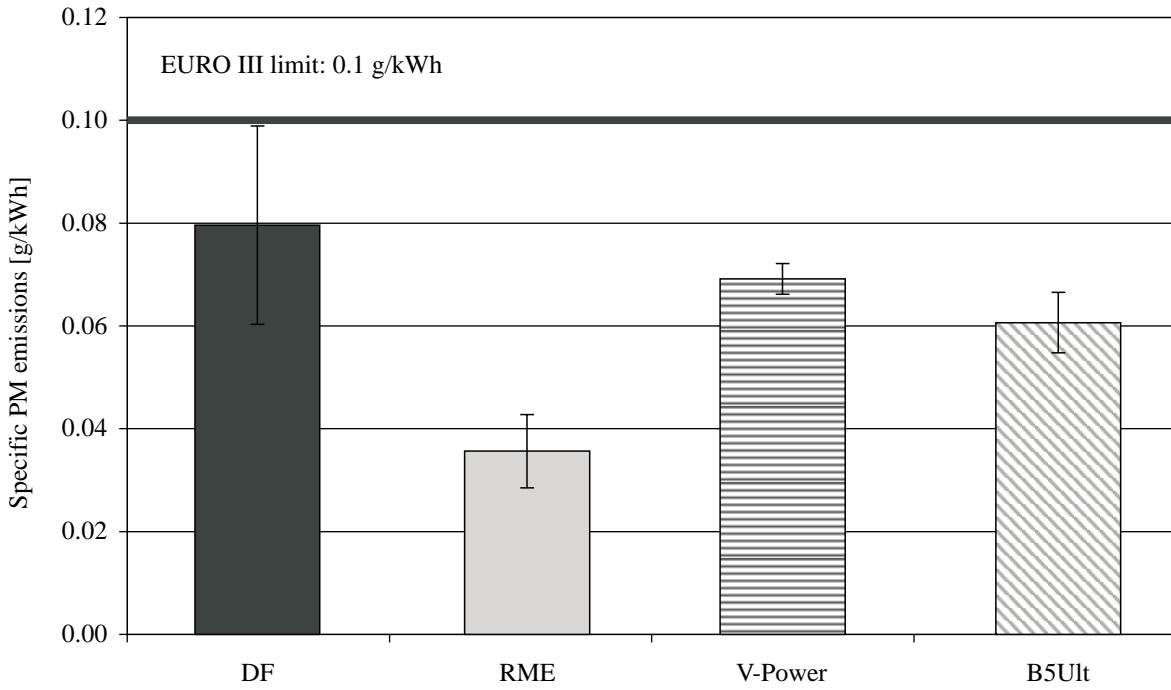
Figure 8:

Specific PM emissions (OM 906 LA, ESC test)

ELPI

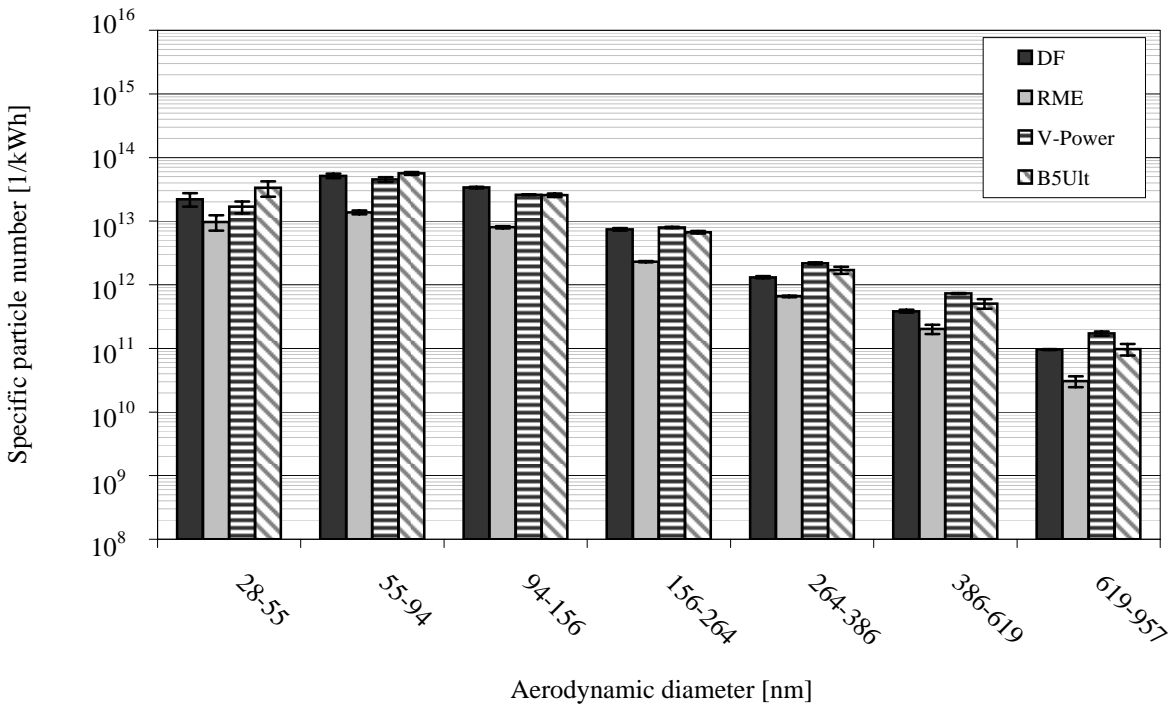The ELPI results are presented in figure 9.

Figure 9:

Specific particle number distribution in the exhaust gas (ELPI, OM 906 LA, ESC test)

RME showed the lowest emissions compared with the other fuels. The other three fuels exhibited a similar emission tendency among themselves over the entire size range. Up to impactor stage that classifies particles with diameters from 156 to 264 nm hardly no differences between V-Power, B5Ult and DF where observed; with exception of the size range from 28 to 55 nm, in which B5Ult showed increasing emissions. As for larger particles the emissions of V-Power and B5Ult were slightly higher versus DF. Starting from the size class range from 156 to 264 nm the highest emissions were detected for V-Power compared with the remaining fuels. In detail, B5Ult showed similar emissions as DF. Advantages concerning the emissions of particles where observed for RME.

SMPS

Analogically to ELPI and PM results, RME led as well to the lowest particulate emissions, when SMPS is used, figure 10.

RME demonstrated great potential to obtain a reduction in particle emission.

The determination of the particle size distribution with SMPS shows a bimodal size distribution (figure 1 and 10). In detail, the particle size distribution determined by the SMPS was slightly shifted to smaller values, such that the maximum of the nuclei mode was below 10 nm.

However, in individual modes different distributions were detected. At idle a high emission of ultrafine particles was measured (figure 11). At rated power the accumulation mode predominates. In the range of the accumulation mode the highest particle number emissions were found (figure 12). This leads to the result that – accumulated over the entire ESC test – a bimodal particle size distribution is observed.
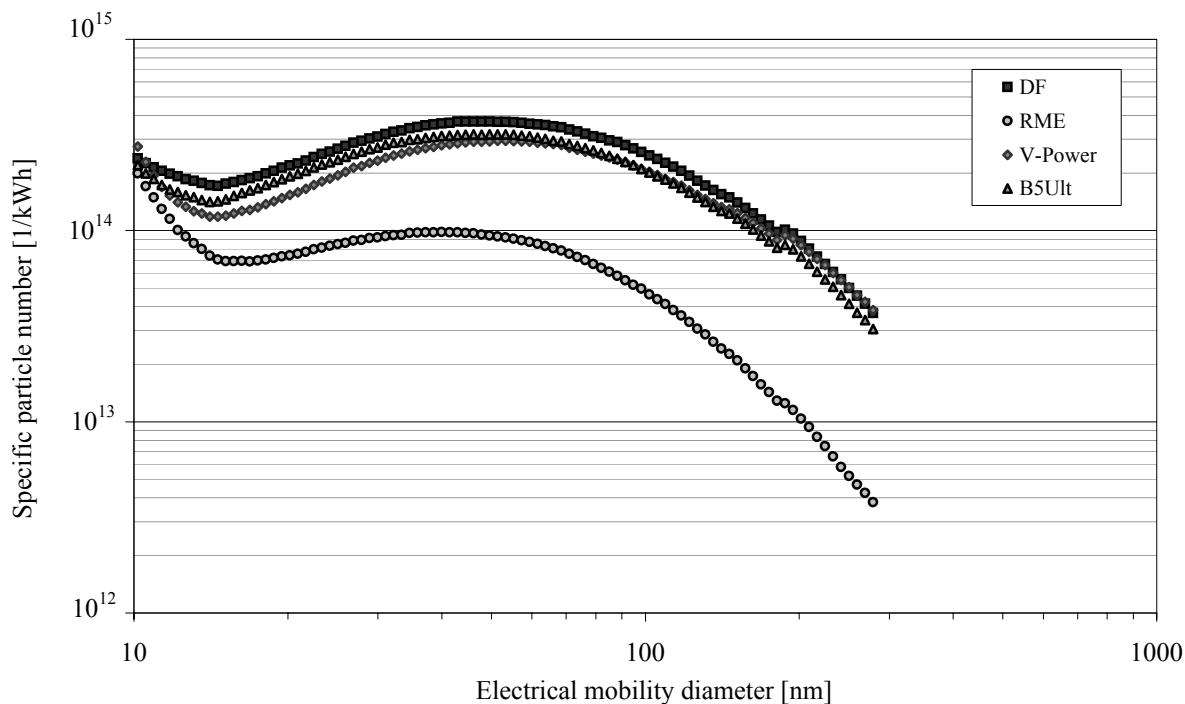

Figure 10:

Specific particle number distribution in exhaust gas (SMPS, OM 906 LA, ESC test)

Besides RME, all diesel fuels showed comparable emissions. However, the particulate emission of reference diesel fuel was slightly increased in comparison with V-Power and B5Ult.
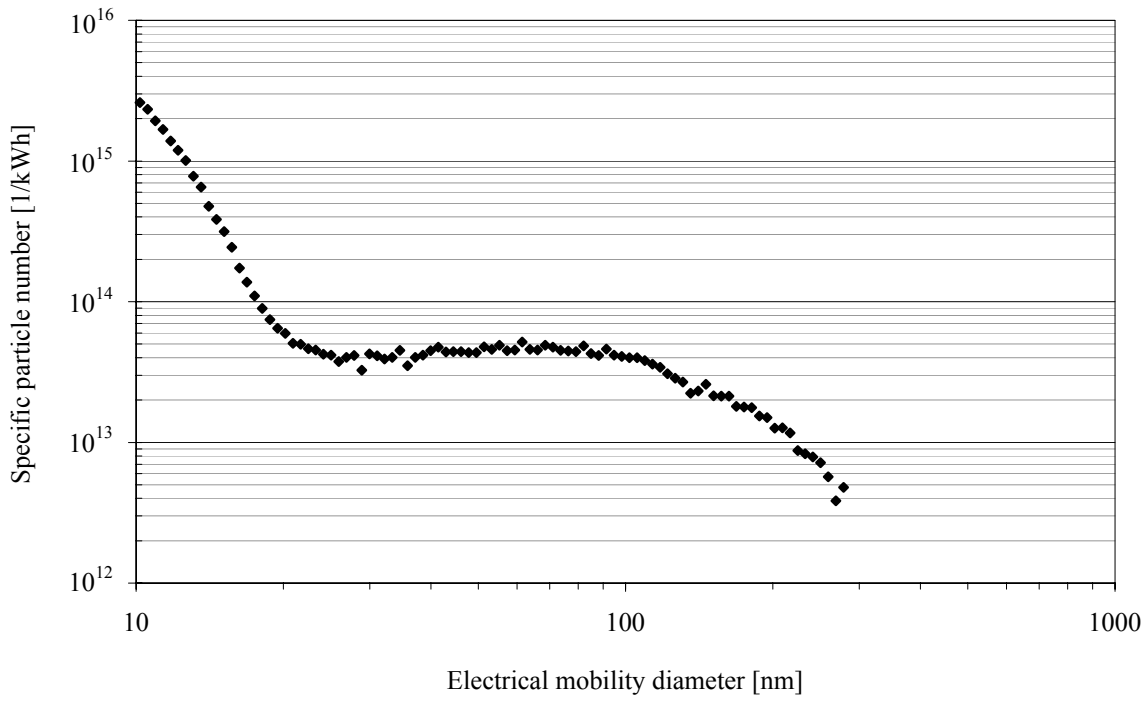
Figure 11:

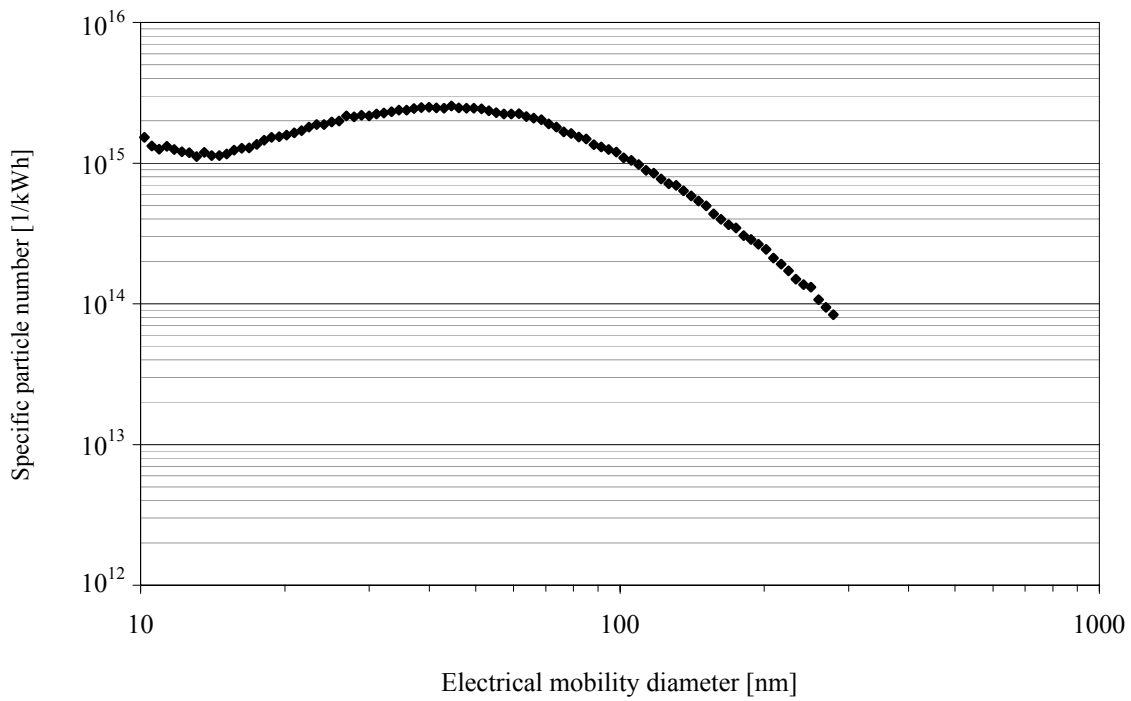Specific particle number distribution at idle (SMPS, **OM 906 LA, RME)**

Figure 12:

Specific particle number distribution at rated power (SMPS, OM 906 LA, RME)

Influence of the dilution conditions on the particle size distribution – Temperature effects

The influence of the dilution temperature on the particle size distribution was investigated at the example of RME. Each measurement was fourfold repeated in mode 9 (1800 min[-1]; 265 Nm) of the ESC test. This test mode was selected, since in this mode both the nuclei mode and the accumulation mode are present and therefore the influence of the temperature on both ranges was expected to be observable quite well.

The influences of the temperature on the particle size distribution are shown in figure 13.

sition. For RME a high quota of soluble organic fraction (SOF) and only a small amount of insoluble fraction (ISF) was found; data not shown. It can be assumed that particles within the range of 10 to 40 nm, which were measured after the hot dilution at temperatures greater than 80 °C, are mainly soot, metallic ash or heavy hydrocarbons (Montajir R. M. et al. 2006).
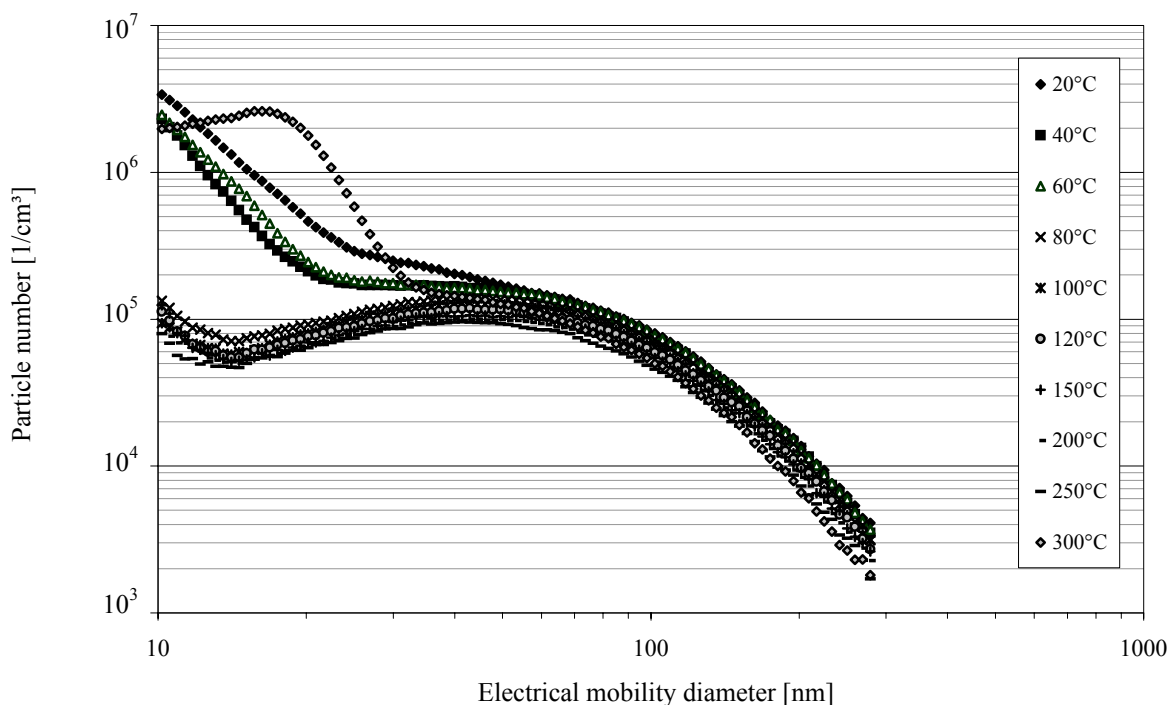

Figure 13:

Particle number distribution at mode 9 (SMPS, OM 906 LA, RME)

In the temperature range from 20 to 60 °C no differences could be observed. The distribution was dominated by particles within the range of 10 nm to 30 nm. With an increase of the temperature up to 80 °C a decrease in number of nuclei particles to one tenth was observed. In the temperature range from 80 to 250 °C the number of particles remained almost constant. At a temperature of 300 °C an increase of particles with a maximum at 17 nm was observed. The influence of the temperatures on the particle number within the size range from 10 to 20 nm has strong effects already at 80 °C. This suggests that particles in this size range may consist mainly of volatile substances. This result is supported by additional investigations of the particle compo-

Summary

Three methods for particle measurement were introduced, such as the legal method for PM emissions and two techniques to detect particle size (ELPI) and particle number distributions (SMPS).

Using these methods diesel engine emissions of a Euro 3 heavy-duty engine were analysed and compared for rapeseed oil methyl ester (RME), reference diesel fuel (DF) and the premium diesel fuels Shell V-Power Diesel® and Aral Ultimate Diesel®. The latter was blended with 5 % RME (B5Ult).

Previous investigations showed higher numbers of particles in the nuclei mode for RME versus DF. Therefore

more detailed information had to be obtained to what extent RME induces the formation of ultrafine particles, which are considered as harmful to human health.

In course of the actual investigations it could be demonstrated that these nuclei particles did not mainly consist of soot, but most probably of unburned fuel. It was shown that the sampling conditions – especially the temperature in the dilution tunnel – had a significant effect on the detected particle size and number distributions. Particularly ultrafine particles in the nuclei mode from 10 to 40 nm decreased when the temperature rose over 80 °C.

RME led to reductions of particle number emissions versus all other fuels, whereas the fossil based diesel fuels DF, B5Ult and V-Power did not vary significantly among each other.

Regarding particulate matter emissions (PM) a reduction by factor two was detected for RME versus DF.

In the result and at the example of the test engine it can be concluded that RME does not lead to higher ultrafine particle emissions than DF, B5Ult and V-Power.

Acknowledgments

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