

## Aus dem Institut für Technologie und Biosystemtechnik

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### Measurement of particle emissions from diesel engines operating on different fuels

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#### Abstract

Diesel engines are the most common power sources in agriculture. Generally nearly as much diesel fuel as gasoline is needed in Germany for public transportation. Because diesel engines are a major source for fine particles (< 2.5  $\mu$ m) and the main source for ultrafine particles (< 0.1  $\mu$ m) it is necessary to determine the emissions and to characterise the particulate matter (PM). The goal of our research was to investigate and to compare particulate matter emissions from biodiesel and conventional diesel fuel. In this context biodiesel is synonymous with rape seed oil methyl ester (RME). The contribution of agriculture to PM emissions from diesel engines was about 6 % during 1996 and 2000 according to the diesel fuel consumption in Germany (Jahresbericht Mineralölwirtschaftsverband, 2000).

The Institute of Technology and Biosystems Engineering is equipped with an engine test stand to measure regulated and non-regulated emissions from engines. In detail three measurement techniques are available to detect particulate matter. The most important technique is the exhaust dilution tunnel in accordance with ECE standards with a filter unit at its end for gravimetric analysis. The second technique is a BERNER-Low-Pressure-Impactor to study the particulate mass distribution in the range from 0.015 to 16.0  $\mu$ m. The third available instrument is a Scanning-Mobility-Particle-Sizer (SMPS) that detects the particle number distribution for particles with an electronical mobility diameter from 10 to 300 nm.

Three different fuels were tested in four modes of the 12-mode test (ECE-R 49). Besides RME, two common diesel fuels were chosen. In general RME showed for the mentioned measurement techniques the lowest particle emissions in the four investigated engine test points. Only partial load and rated power showed with the SMPS an increased particle number concentration between 10 and 45 nm for biodiesel versus the mineral diesel fuels. This is a remarkable result because former experiments indicated an opposite trend. Probably the progress in engine development, for example the increased injection pressure up to 1600 bar and higher, causes this positive effect. But it is necessary to point out that both the emissions for biodiesel and conventional diesel are lowered with a modern engine.

Beyond that it could be found that a low sulfur content correlates with lowered particulate matter emissions. But the effect was obviously less than the general chemical differences between mineral and renewable diesel fuel.

Keywords: Biodiesel, Diesel, Dilution Tunnel, Impactor, Particle Number Distribution, Particle Mass Distribution, SMPS, Sulfur Content

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#### **1** Introduction

Diesel engines become more and more popular. In 2001 in Germany more than 33 % of all new sold passenger cars were diesel fueled (Jahresbericht VDA, 2001). Five years ago the percentage was only 12 % (Statistik Kraftfahrtbundesamt, 2002). The contribution of agriculture to the particle emissions from diesel engines was about 6 % during 1996 and 2000 according to the diesel fuel consumption in Germany. This underlines the importance of emission research with a special focus on particles the more because diesel engines are a major source for fine particles (< 2.5  $\mu$ m) and the main source for ultrafine particles (< 0.1 µm) (Wichmann, 2002). Both kinds of particles are considered to be harmful to human health and even to be responsible for the carcinogenicity of diesel exhaust emissions (DEE). In this context it is necessary to know that ultrafine particles are obviously more toxic than fine particles regarding the same mass application rate (Wichmann, 2002; Heinrich, 1998). Furthermore, the particle number concentration is a very important property to measure and to discuss because of the high deposition capability (50 to 60 %) of ultrafine particles in the alveolar respiratory tract (Heinrich, 1998). At the Institute of Technology and Biosystems Engineering

Table 1: Fuel properties three measurement techniques are in use to detect PM emissions: A dilution tunnel to collect and measure the particle mass, a BERNER-Low-Pressure-Impactor to determine the particle mass distribution and a scanning mobibility particle sizer to measure the particle number concentration.

Besides the knowledge about particle mass and number distribution of the DEE from conventional diesel fuel it is essential to investigate which fuel parameters are probably responsible for low respectively high particle emissions. In course of a preliminary investigation three different fuels were tested in four modes of the 12-mode test (ECE-R 49). Table 1 shows the properties of the fuels. The common diesel fuels (DF 41 and DF 290) according to the European specification DIN EN 590 was delivered from Louis Drevfus & Cie, Hannover. DF 41 is a low sulfur diesel which is sold since October 2001 at all petrol stations in Germany and fulfills the standards for Euro-IV diesel fuel. DF 290 is a high sulfur fuel according to the old standard of Euro-III diesel fuel and no available at the German longer market. Connemann Company, Leer delivered the biodiesel (RME) according to the German E DIN 51606 specification.

Parameter	Biodiesel	Conventional Diesel	Conventional Diesel
	(RME)	Fuel	Fuel
		(DF 41)	(DF 290)
Density at 15°C [kg/m <sup>3</sup> ]	883.0	825.1	821.8
Kinematic Viscosity at 40°C [mm <sup>2</sup> /s]	4.5	2.373	2.222
Flashpoint [°C]	> 150	62.5	59.0
CFPP [°C]	- 20	- 27	- 14
Sulfur Content [mg/kg]	< 10	41	290
Carbon Residue [wt-%]	< 0.05	< 0.05	< 0.05
Cetane Number [-]	> 55	53.6	53.0
Ash Content [wt-%]	< 0.01	< 0.001	< 0.001
Water Content [mg/kg]	180	20	70
Acid Number [mg KOH/g]	0.145	0.05	
Iodine Number [-]	110		
Polycyclic Aromatic Content [vol-%]		4.9	4.2

#### 2 Engine and Engine Test Procedure

The investigations were carried out at a modern Mercedes Benz engine type OM 904 LA with turbo charger and intercooling. The 125 kW four cylinder aggregate with a unit-pump direct injection system is often used in light duty trucks. The engine was assembled to a test bench. figure 1 shows schematically the engine test stand to detect regulated and non-regulated emissions.



Figure 1: Scheme of the Emission Test Stand

In figure 2 the four chosen test modes from the ECE-R 49 test are shown.



Figure 2:

13-mode test (ECE R 49) and chosen test modes

#### 3 Measurement technique

#### 3.1 Dilution Tunnel

The measurement of the particle mass from engine exhaust is also regulated in the ECE-R 49. It is necessary to dilute the raw exhaust and to cool it down under 52°C to assure that all volatile exhaust components are condensed on the particulate matter. After that the particle emissions were collected on PTFE-coated fiberfilm filters (T60A20, Pallflex Products). Before and after the sampling procedure the filters were weighed with a microgram balance (Sartorius M5P,  $\pm$  5 µg accuracy) to determine the emitted particulate mass. Preceding each measurement, the filters were conditioned at  $22^{\circ}C \pm$ 3°C and a relative humidity (r.h.) of 45 %  $\pm$  8 % for at least 24 hours.

#### 3.2 BERNER-Low-Pressure-Impactor

The impactor is an instrument which classifies the particulate matter into ten different size classes. Table 2 shows the characteristics of the used instrument.

#### Table 2:

Stages and separation diameters of the BERNER-Lowpressure-Impacter

Impactor Stages	Separation Diameter [µm]	
10	8.000 - 16.00	
9	4.000 - 8.000	
8	2.000 - 4.000	
7	1.000 - 2.000	
6	0.500 - 1.000	
5	0.250 - 0.500	
4	0.125 - 0.250	
3	0.060 - 0.125	
2	0.030 - 0.060	
1	0.015 - 0.030	

The Berner impactor operates on the inertial behaviour of the particles. The exhaust stream is sucked through the successively arranged ten impactor stages. Starting with impactor stage number ten, the gas flows through the instrument. Every stage consists of a nozzle and an impaction plate on which a specific PM fraction is collected (figure 3). The impaction plate deflects the flow of the exhaust and causes a 90° shift in direction. Therefore a definite particle fraction with a specific aerodynamic diameter hits the impaction plate and is collected on its surface. Smaller particles will remain airborne and flow with the gas stream to the next stages.



Figure 3: Scheme of an Impactor Stage

#### 3.3 Scanning-Mobility-Particle-Sizer (SMPS)

The principle of the SMPS is shown in figure 4.



Figure 4:

Scheme of the SMPS System (Instruction Manual, TSI Incorporated)

Before the aerosol enters the instrument it passes through an inertial impactor stage to separate particles above a known diameter from the actual interesting particle fraction (10 - 300 nm). Afterwards the aerosol enters the KR-85 Bipolar Neutralizer where the exhaust stream particles collide with bipolar ions. Consequential the aerosol reaches very fast a state of equilibrium with a bipolar charge distribution. The charge distribution follows a theoretically and practically verified model developed by Wiedensohler and Fissan (1988) for particle sizes in the submicrometer regime. Then the polydispers aerosol leaves the neutralizer into the classifier zone with two concentric metal cylinders. In accordance with figure 4 the polydispers aerosol and the sheath air enter the classifier from the top and stream along the outer and inner rod. Thereby the sheath air and the sample air don't mix with each other. The inner cylinder is charged with a well-defined negative electric charge. On the other hand the outer cylinder is electrically grounded so that an electric field is created between the two rods. Thus positively charged particles from

the polydispers aerosol are attracted to the inner cylinder and will be deposited there. The place where the particles depose depends on their electrical mobility and the charge of the cylinder. At the bottom of the inner rod there is a small slit where only particles with a defined electrical mobility can leave the classifier towards the condensation particle counter (CPC) as so called monodispers aerosol. During the measurement, the SMPS varies the voltage of the inner rod between 20 and 10 000 V depending on the particle sizes of interest. After the polydispers aerosol is classified into the monodispers aerosol the particle number is counted with a CPC. The aerosol flows into a heated chamber that is saturated with nbutanol. Particles and butanol vapor leave the chamber towards a condenser unit where the alcohol condenses onto the particles. The mechanism is a heterogeneous condensation. Finally the droplets' sizes are between two and three micrometer so that they can be counted optically.

#### 4 Results and Discussion

#### 4.1 Dilution Tunnel

Figure 5 shows exemplary the PM emissions from the tested fuels at maximum torque. It becomes obvious that RME emits less PM than the fossil fuels. The mass concentration is reduced about 40 % versus DF 290 and 35 % versus DF 41.



Figure 5:

Particle mass concentration for RME, DF 41 and DF 290 at maximum torque

#### 4.2 BERNER-Low-Pressure-Impactor

Figures 6 to 9 present the impactor results for all four tested modes of the ECE R49 test cycle. The samples for the impactor were taken out of the raw exhaust gas in accordance with figure 1.



Figure 6: Particle mass flow at idle



Figure 7: Particle mass flow at partial load (25 %)



Figure 8: Particle mass flow at maximum torque





For all test modes the particle mass flow is lower for RME than for the other diesel fuels. And again in comparison to figure 5, except for idle, DF 290 emits the highest particle mass per minute. This is especially very distinctive for maximum torque. Although at maximum torque the standard deviations for the DF 290 results are unproportional high, so that the picture might show too extreme differences to the other fuels, the tendency still exists.

For partial load and rated power, the maxima of the particle mass flow can be observed at separation diameters of 60 to 125 nm. Test mode idle shows an especial distribution for the three diesel fuels in comparison to the other test modes. While the maximum particle mass flow for DF 290 was found between 30 and 60 nm, DF 41 shows a maximum between 125 and 250 nm. The maximum for RME was found in between (60 - 125 nm). Although for the two diesel fuels at idle the total mass flow over all impactor stages is nearly the same, the shift to larger particle diameters for DF 41 versus DF 290 is remarkable. An opposite trend can be seen at mode maximum torque (figure 8) where the maximum for DF 41 lies between 60 and 125 nm and for DF 290 between 250 and 500 nm.

In general it is significant for the particle distributions that the preponderant part is smaller than a separation diameter of 1  $\mu$ m. This fact is very important with regard to the occupational health

effects of the particles, because of their presumably increased mutagenicity.

Contrary to the expectation that the different engine loads will lead to specific shifts for the maximums, no uniform trend can be seen for the tested modes.

#### 4.3 Scanning-Mobility-Particle-Sizer (SMPS)

Basically the particles between 10 and 300 nm can separated into two different ranges. The particles from 0 to approximately 40 nm belong to the so called nucleation mode and particles larger than 40 nm to the accumulation mode.

The following figures (figure 10 to 13) show the particle number distributions for the four test modes. They were sampled from the diluted exhaust gas (figure 1). Before the particle stream reached the SMPS it was diluted for a second time with a micro-dilution-tunnel which is not shown in figure 1.



Figure 10: Particle number distribution at idle



Figure 11: Particle number distribution at partial load



Figure 12: Particle number distribution at maximum torque





DF 41 and DF 290 show a close relationship apart from maximum torque (figure 12) where DF 290 emits noticeable more particles between 10 and 30 nm.

In addition to partial load (figure 11) also maximum torque (figure 12) and rated power (figure 13) exhibit more smaller particles for RME as for the conventional diesel fuels. These particles belong to the nucleation mode particles which are very sensitive temperature and dilution ratio changes. to Experiments conducted by FEV Motorentechnik Aachen showed that it is possible to reduce these particles to nearly zero by heating the diluted sample stream (Pungs et al., 2000). Particles larger than approximately 40 nm are dedicated to the accumulation mode particles and they are insensitive to varying sampling conditions (Hall et al., 2000; Abdul-Khalek et al., 1999). In this case the temperature and dilution ratio conditions were the same for all fuels. So the differing RME nucleation mode must have an other reason.

Diesel engines are especially developed for specified diesel fuel. Biodiesel with its slightly different physical and chemical properties shows a modified combustion behaviour. In former investigations a lot of unburned RME was found to be adsorbed on the PM filters (Prieger et al., 1996). From this it follows that the not optimised biodiesel combustion may cause additional fuel droplets which are counted by the SMPS instrument in the nucleation mode between 10 and 40 nm.

For the accumulation mode RME falls mostly below the particle number concentration of DF 41 and DF 290. The observed especial distribution at test mode idle for the impactor results cannot be repeated. Beyond that the results for the impactor and SMPS measurements are generally not comparable to each other. The measurement principles and the ways of sampling differ too much.

#### **5** Conclusion

For all three measurement techniques the lowest particle emissions were found for RME using the four engine test modes. Only for partial load and rated power the number of nucleation mode particles was increased for biodiesel. This is remarkable because former experiments indicated an inverted trend for total particulate mass, particle mass distribution and particle number distribution (Krahl et al., 2001; Prieger et al., 1996). Probably the engine design particularly with regard to the injection technique may be responsible for that effect. While older engines injected the fuel with e.g. 380 bar (MWM 302-2) the state of the art Mercedes Benz OM 904 LA engine reaches an injection pressure of 1600 bar. The pressure difference causes a significant particulate matter reduction for the conventional diesel fuels as well as for biodiesel in particular.

Another aspect is introduced by the fuel properties (table 1), mainly the different sulfur contents of the fuels. DF 290 has the highest amount of sulfur with 290 ppm and causes nearly always the highest PM emissions. Although the sulfur content of DF 41 with 41 ppm is definitely lower, the PM emissions are only slightly lower than for DF 290. Except for maximum torque the particle number distributions are even well comparable for the two mineral diesel fuels. For RME with a sulfur content lower than 10 ppm the reduction for particulate matter (PM) is significantly higher, but the effect of sulfur content reduction is obviously less than the general chemical differences between mineral and renewable diesel fuel. In addition to the sulfur

content, density, kinematic viscosity, and the flashpoint are parameters which distinguish the fossil fuels from the regenerative one.

The three different measurement techniques are good and necessary instruments for the investigation of particle emissions from diesel engines. They lead to different results with which it is possible to discuss different aspects of the emission behaviour of diesel engines operating on different fuels.

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