

Non-Esterified Plant Oils as Fuel-Engine Characteristics, Emissions and Mutagenic Effects of Particulate Matter

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3.10 Non-Esterified Plant Oils as Fuel-Engine Characteristics, Emissions and Mutagenic Effects of Particulate Matter

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ZUSAMMENFASSUNG

Pflanzenöle können einen gewissen Beitrag für eine erneuerbare, nahezu CO₂-neutrale Kraftstoffversorgung leisten. Die nicht-veresterten Pflanzenöle haben im Gegensatz zu veresterten Ölen eine günstige Energie- und CO₂-Bilanz. Deshalb werden hier die naturbelassenen, aber raffinierten Pflanzenöle auf ihre Eignung als Kraftstoff in Dieselmotoren und deren Emissionen detailliert untersucht. Versuche wurden mit drei verschiedenen Dieselmotoren durchgeführt.

ABSTRACT

Plant oils may be used as a sustainable, nearly CO₂ neutral fuel for diesel engines. This work investigates experimentally the particulate and gaseous emissions of diesel engines fuelled with non-esterified, pure plant oils with the quality standard of DIN V 51605 (Weihenstephan RK-Qualitätsstandard 05/2000). The data are collected from three engines:

- Common rail passenger car engine from Opel AG
- Truck engine from VOLVO
- Truck engine from MAN AG

The emissions of the MAN engine have been used to perform AMES tests to analyze possible health impacts of plant oil operation.

The experimental data show a reduction of particulate matter compared to traditional gasoil which may yield up to 50 % for engines which are correctly adjusted to plant oil operation. The particulate matter shows same primary particle sizes but the agglomerates as collected on TEM grids are different – the plant oil soot particles tend to form larger aggregates [4]. The gaseous emissions of CO and hydrocarbons HC are generally lower compared to the operation with gasoil. However, the NO_x emissions are slightly higher. This may be contributed to the measured higher combustion chamber pressures and temperatures when fuelled by plant oils.

Emission samples have been extracted from ESC cycles of 13 step tests to perform the AMES test which give indication on carcinogen substances. The AMES test results gave no indication of mutagenic effects exceeding the detection limits. No significant differences could be found comparing the emissions of plant oil and gasoil operation. Thus, it can be stated that the emission from plant oil operation does not have a

health impact different to traditional gas oil. This is in contrast to some other publications – a deeper insight shows that these investigations did not properly modify the engine for plant oils. It is mandatory to make the engine modification to pre-warm the plant oils to 90°C prior to injection. The engine's warm-up phase needs special care to avoid any coking at the injection system and combustion chamber surfaces. The publications, where a higher health risk was claimed to be found in the exhaust of plant oil fuels, did not pre-warm the plant oils – cold plant oils have been injected in the combustion chamber instead. This results in incomplete atomization and incomplete combustion with a lot of hazardous emission species. Such an operation will damage the engine after relatively short times and is, therefore, not realistic.

The investigated fuels had some influence on the engine characteristics. Higher temperatures and pressures in the cylinder have been detected for some plant oils compared to gasoil. This increase is explained by the higher oxygen content within the plant oils.

INTRODUCTION

The known fossil fuel reserves decrease, whereas the consumption increases. Part of the fuel consumption can be covered by plant oils. There are over 200 oil plants known which may also grow in arid areas. Thus, even poor, third world countries may grow sustainably there

own energy demand. In Europe, there are numerous co-generation plants with plant oils (mainly palm oil) in operation and some logistic enterprises operate their whole fleets of trucks with pure plant oils. The use of food plants for fuel production and the possible ecological impacts triggered an ethic discussion, which is not subject of this investigation.

PLANT OILS AS DIESEL ENGINE FUEL

The air temperature after compression is approx. 450 to 550°C for most diesel engines. This temperature range is sufficient for plant oils which have an ignition temperature of approx. 200 to 300°C. However, the kinematic viscosity of plant oils with approx. $\eta = 80 \text{ mm}^2/\text{s}$ at room temperature is much higher compared to gas oil with approx. $5 \text{ mm}^2/\text{s}$. The high viscosities of plant oils cause high mechanical stress to the injection system and the atomization is poor, providing large fuel droplets, incomplete combustion, coking in the combustion chamber and thus damages to the injection components, cylinder and piston rings.

Basically two techniques are known to adjust plant oils to advanced diesel engines:

- a) Chemical modification of the plant oils
- b) Engine adjustments to pure plant oils

To a.: Esterified plant oils have a viscosity which is similar to gas oils. Methyl esters of the oils can be used without further engine modifications. Two disadvantages are known. One is the energy consumption of the chemical process and the other is the chemical instability of some sealing materials against esters, which act as solvents. Special sealings are required in the engine for esterified oils. This paper deals exclusively with pure plant oils. Esterified oils are not considered any more.

To b: The plant oil's viscosity decreases with increasing temperatures. At 80 to 90°C, the viscosity drops below $10 \text{ mm}^2/\text{s}$ [2] which is suitable even for modern common rail injection systems. Consequently, the pure plant oils must be heated prior to the pump and the injection. Above mentioned damages are avoided and the engines can be operated permanently. Several engine manu-

facturers allow the usage of pure plant oils, provided that a reliable pre-warming system is installed.

Several techniques are known to master the cold start, the challenging phase when plant oil is used. Detailed technical information to modification techniques for plant oils are found in [3]. The two-tank system has been proven in the last years as most simple, most economic and most reliable solution. The cold engine is always started by normal gasoil. If the engine is warm enough, the plant oil is heated by a heat exchanger installed in the loop of the cooling water. But the engine must be operated in the last minute by gasoil to flush the injection system and fill it with the gasoil for the next cold start. A disadvantage is the additional tank for the gasoil.

INVESTIGATED ENGINES

The specifications of the three diesel engines under investigation are listed in tables 1, 2 and 3.

The OPEL engine in the lab of the University of Applied Sciences Offenburg served for the basic investigations on the impacts of plant oil on engine behaviour and emissions. The VOLVO truck served for test cycles in real traffic operation and to get information on the behaviour of the after-treatment system for plant oil fuels. The MAN engine emissions was dedicated for the AMES tests to gain information on the health impact. All engines have been converted correctly to plant oil operation.

Manufacturer	GM Opel
Type	Diesel engine 1.7 CDTI ECOTEC®
Number of cylinders	4 in line
Volume	1,686 cm ³
Valve number	16
Diverse	Common rail injection, EGR
Bore / stroke	79.0 / 86.0 mm
Compression relation	18.4 : 1
Max. output	74 kW at 4400 rpm
Max. torque	240 Nm at 2,300 rpm

Table 1: Opel 1.7 CDTI ECOTEC®, laboratory engine of the University of Applied Sciences Offenburg

Manufacturer	VOLVO Euro 5
Type	Diesel engine VOLVO FH 480
Number of cylinders	6 in line
Volume	12,800 cm ³
Diverse	Pump nozzle injection Intercooled turbo compression
Compression relation	
Max. output	353 kW
Max. torque	2300 Nm

Table 2: VOLVO FH 480, installed in VOLVO FH13 truck

Manufacturer	MAN Euro 4
Type	Diesel engine D2066 LF36
Number of cylinders	6 in line
Volume	10,520 cm ³
Diverse	Common rail, EGR and DPF
Max. output	324 kW kW

Table 3: MAN D2066

TEST SET-UPS

Dynamometer for Opel 1.7 CDTI

The engine was equipped with the aggregates and electronics as in the passenger car. An asynchronous generator is used as brake. This e-machine acted also as starter. The fuel system involved a tank for gasoil, a tank with auxiliary pump for the plant oil, filters, volume flow meter, fuel pump, heat exchanger for plant oil, high pressure pump and injectors. The extraction tubes were heated up to approx. 200°C avoiding condensing effects. Figure 3.10-1 shows the arrangement of the extraction positions and temperature sensors in the exhaust system, figure 3.10-2 is a photo of the dynamometer. The dynamometer is described already in [4, 11].

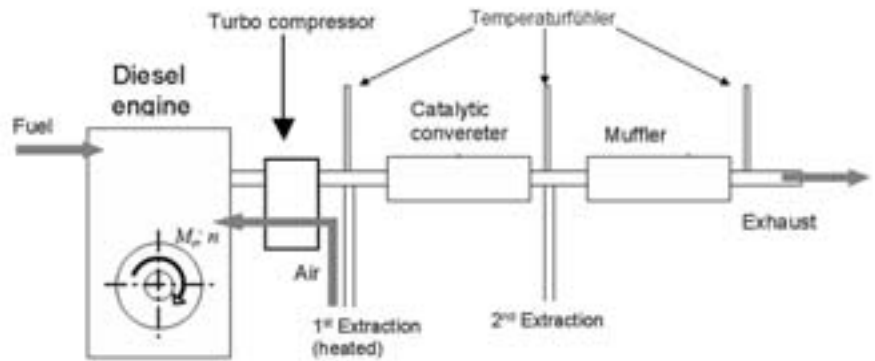


Fig. 3.10-1: Exhaust system with extractions and sensors

VOLVO FH 480, installed in VOLVO FH13 Truck

The goal of the measurements in the operating truck was the emission data collection, i.e. gaseous and particulate emissions in real traffic cycles.

Dynamometer for MAN engine

The state of the art dynamometer for the MAN engine is completely equipped including all necessary sensors and extraction installations for the exhaust samples to perform the AMES tests.

MEASUREMENT TECHNIQUES

General engine data

All conventional engine data like speed of revolution, torque, temperatures (exhaust, cooling water, inlet air) and fuel volume flow were recorded.

PRESSURE INDICATION

One cylinder of the OPEL engine was equipped with a piezo-electric pressure indication sensor which was installed instead of the glow plug. The AVL Indimaster 6704 analyzed the induced pressures. Thus, the indicated mean pressure p_i is obtained versus the crank angle, serving as basis for p_i -V-diagrams and others.

EMISSION GAS ANALYSIS

The DiGas 440 of AVL measured the gaseous emissions. The gas was conditioned: The gas was cooled to separate the condensed liquid from the gases. A filter separated practically all particles. A gas blower increased the gas pressure and the gas was then guided into the gas analyzer after two additional particle filters. The gas data were continuously re-

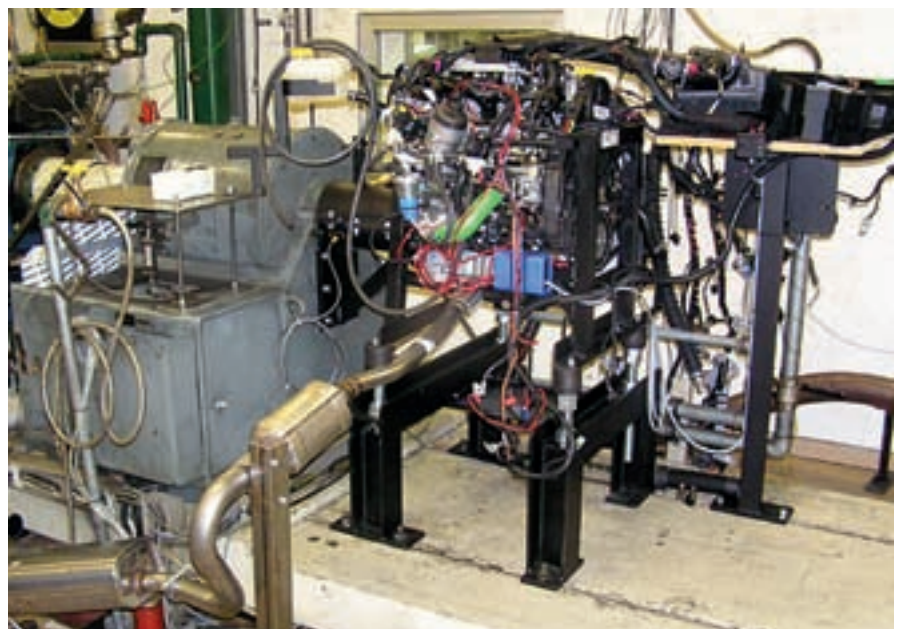


Fig. 3.10-2: EURO 4 Diesel engine connected to the engine dynamometer of the University of Applied Sciences Offenburg

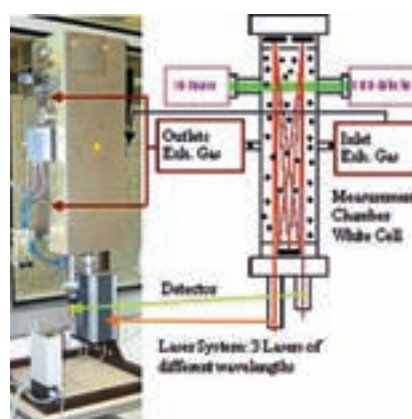


Fig. 3.10-3: LPME System WIZARD DQL [6]

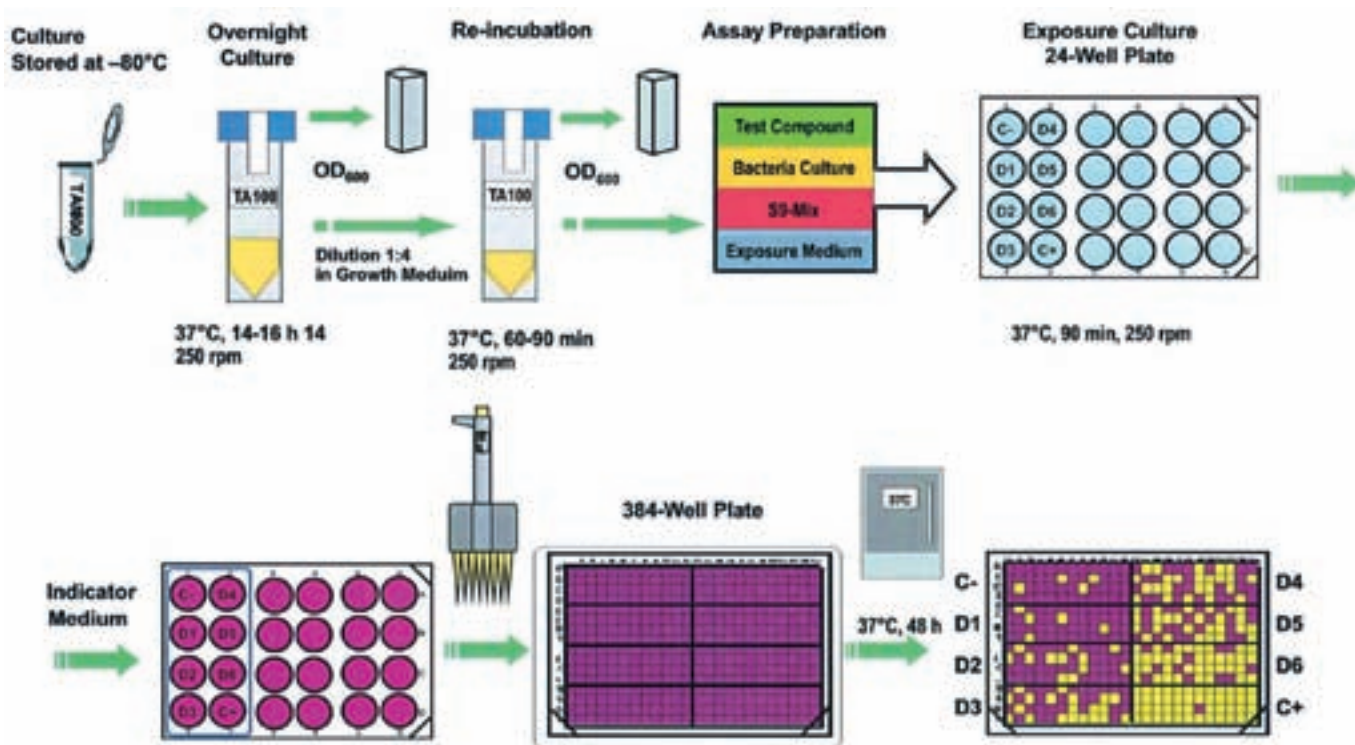


Fig. 3.10-4: Visualisation of the AMES fluctuation test [7]

corded at 1 Hz. The starting procedure with warm up phase and auto calibration of DiGas 440 was respected. The exhaust gases CO, CO₂, hydrocarbons HC, O₂ and NO_x were recorded.

PARTICULATE EMISSION Long Path Multi-Wavelength Extinction LPME

The LPME technique to measure on-line the particle mean diameter and the particle concentration in the hot, undiluted exhaust is described in detail in [4, 5]. Figure 3.10-3 explains the set-up.

Transmission Electron Microscope TEM

The particles have been captured in the exhaust flow on grids which have been analyzed in the Institute of Electron Microscopy of the University Karlsruhe.

Gravimetry

Gravimetric measurements were taken by all engines according to the normalized procedure.

AMES TEST

The AMES test -developed in the seventies by B. Ames- is aimed to identify mutagenic substances. Test media are special bacteria mutants of "Salmonella typhi-mu-rium" with histidin deficiency. The applied auxotrope mutants are not

able to synthesize histidin so that they can not multiply on histidin free nutrition. Mutagenic substances may cause mutations which enable the bacteria again to synthesize histidin. These "His-revertants" can multiply again in histidin free environment. The number of revertants after an incubation of 48 hours is a measure for the mutagenic effect of the substance under investigation.

The applied AMES fluctuation assay is from the company Xenometrix GmbH. The test sequence is visualized in figure 3.10-4. The following test phylums have been used for the different mutagenics:

- TA 98 to detect screen thrust mutagenics
- TA 100 to measure base exchange mutagenics
- TA Mix (generic development of Xenometrix)

The change of the nutrition's pH value is an indication for the change of the histidin concentration. The pH indicator measures thus the concentration of the revertants on the micro plate.

There are two AMES techniques acknowledged by the OECD:

- Conventional technique with incubators and agar plates
- AMES fluctuation assay with micro plates and agar plates

The advantages of the applied fluctuation test are:

- Controlled quality of the "Salmonella typhimurium" phylums
- Available ready to use reagents
- Only 30 mg of the test substance is sufficient
- Fast test procedure
- Feasible automation by use of micro titration plates

MEASUREMENTS RESULTS

The following data of the individual engines were measured for different conventional and plant oil fuels:

- Exhaust gas composition
- Particulate matter emission
- Thermodynamic data in the combustion chamber (indicated pressures and derived data)
- Search for mutagenic substances in the particulate matter by the AMES test

OPEL 1.7 CDTI ECOTEC®

Some measurement results of the Opel engine are published in [4].

Exhaust gas analysis

Table 4 is representative for gaseous emissions. The differences between the different fuels were low and sometimes in the measurement uncertainties.

2300 rpm 47 kW	CO	O ₂	CO ₂	HC	NO _x
	ppm vol	ppm vol			
Gas oil	0	8,3	9.2	0	652
Rape seed oil	0	8,7	9.3	0	670
Sun flower oil	0	8,7	9.0	0	716
Soya oil	0	8,5	9.5	0	720
Peanut oil	0	8,6	9.4	0	779

Table 4: Opel engine; results of the gas analysis at 2300 rpm and 47 kW at lambda \approx 1.6; the HC and CO concentrations were below the measurement limits

The exception was the NO_x emission where the gas oil delivers lowest values for all load conditions.

Pressure indication

The absolute cylinder pressure for plant oil was consistently higher for the plant oils in all load conditions of the engine. The differences were as high as 10 %, i.e. 125 bar in peak pressure for gas oil versus 138 bar for sun flower, rape seed and peanut oils (see [4], figure 3.10-6). This effect is contributed to the oxygen content in the plant oil molecules which lead to higher combustion temperatures – the same effect which is responsible for higher NO_x values in the exhaust. Figure 3.10-5 traces the measured heat release over the crank angle at 2,300 rpm and 50 kW for rape seed oil and gas oil. The combustion with rape seed oil was faster than with gas oil. As a result, the pressure of rape seed and other plant oils exceeded the one of gas oil in the combustion chamber [4].

Figure 3.10-6 summarizes the main engine data temperature and pressure in the combustion chamber and NO_x emission versus the speed of revolution and the power for the two fuels conventional gas oil and rape seed oil.

The efficiencies with gas oil were higher compared to rape seed oil, figure 3.10-7. This can be explained to a certain extent by the engine optimization to gas oil; the injection sequence was not modified in the frame of these investigations. The in-

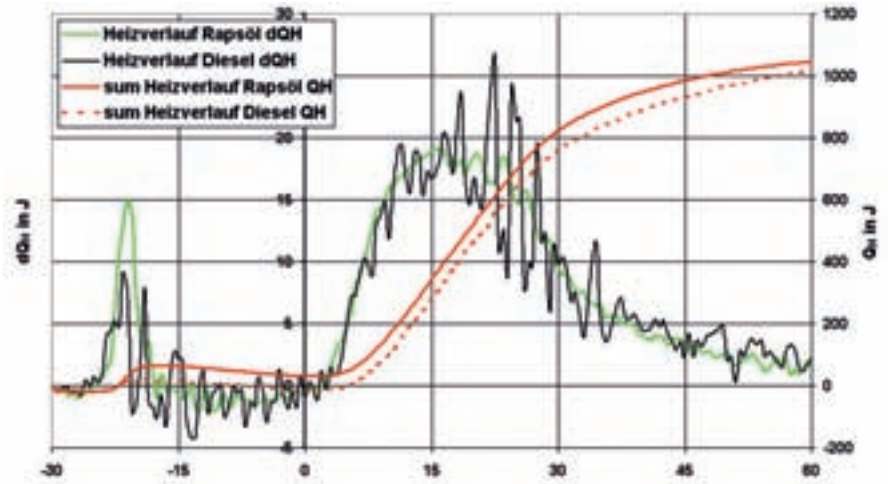


Fig. 3.10-5: Heat release with rape seed oil and gas oil

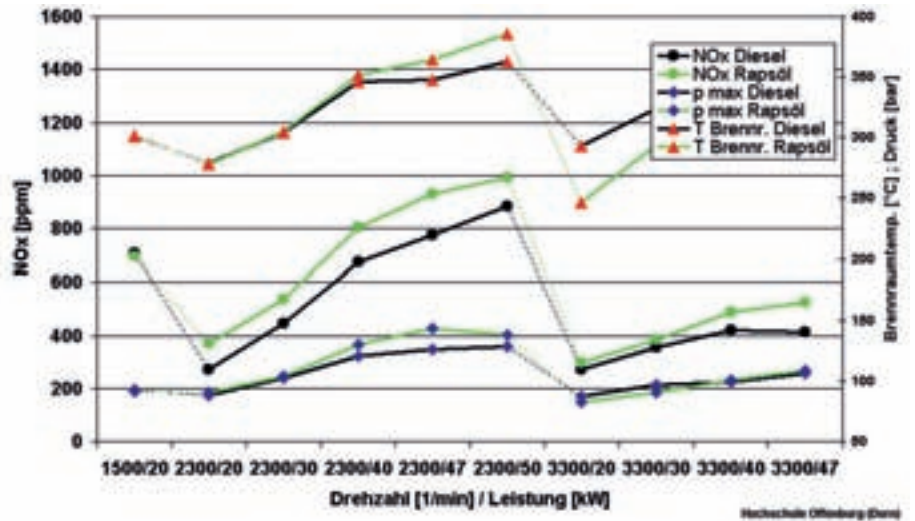


Fig. 3.10-6: Comparison of fuels: Gas oil and rape seed oil. Temperature and pressure in combustion chamber and NO_x emission

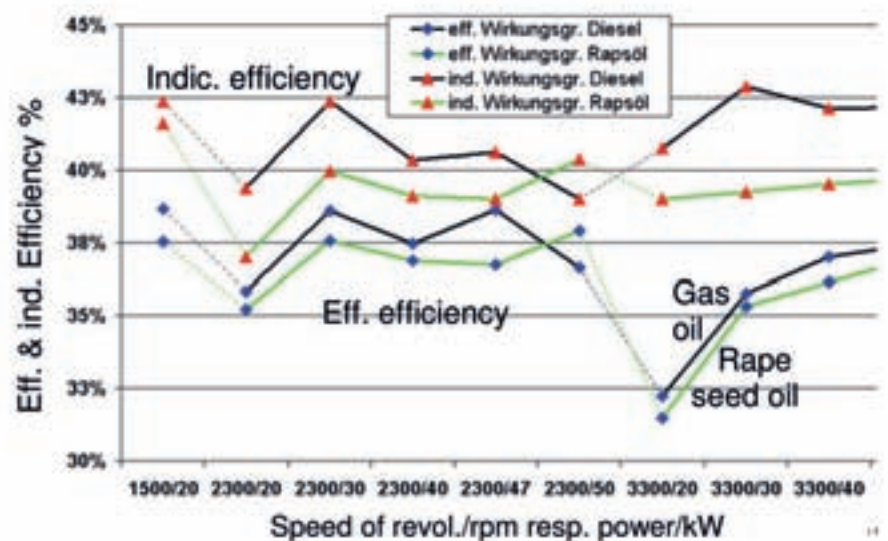


Fig. 3.10-7: Comparison of fuels: Gas oil and rape seed oil. Efficiencies

jection should be shifted to an earlier time according to experiences with plant oils. The mass specific calorific value of plant oil is lower than that of gas oil, but the density is higher so that there should be no major effect due to this difference.

Particulate matter emission

The sizes of the primary soot particles are the same as outlined in [4, 5]. Recent measurements reveal some differences in the particle behaviour which can be seen in the TEM pictures of figures 3.10-8 and 3.10-9. These particles have been collected at the same engine condition at 2300 rpm and 47 kW, but with different fuels. Conventional gas oil emitted the particles shown in figure 3.10-8 and rape seed generated particles of figure 3.10-9.

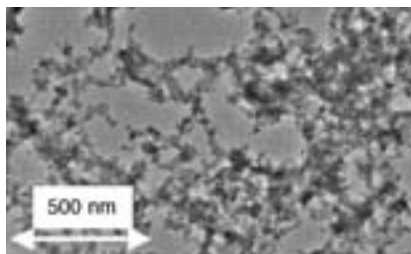


Fig. 3.10-8: TEM picture of particles emitted from gas oil fuel. OPEL engine at 2300 rpm and 47 kW

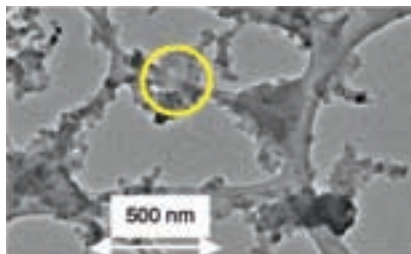


Fig. 3.10-9: TEM picture of particles emitted from rape seed oil fuel. Opel engine at 2300 rpm/47 kW

The primary particles emitted by gas oil fuel formed more chain like aggregates whereas the rape seed oil particles formed more compact, baked together aggregates, i.e. less chain like. The similar effect was found for other plant oils. However, this effect must be investigated further as there might be secondary influences like humidity which may cause these differences. It is too early to speculate that the different particle agglomerates cause a different biological impact.

MAN ENGINE D2066 LF36

The MAN engine was operated in the European Stationary Cycle ESC which

was also selected in [9] so that a direct comparison is feasible.

Emissions

The emissions of the two fuels rape seed oil and gas oil have been investigated. The results of the limited exhaust gas components in the ESC cycle are listed in table 5: CO, NO_x, hydro carbons HC and particulate matter PM. A similar effect as in the OPEL engine was observed. The NO_x emission was higher by approx. 10 %, whereas the other exhaust components were appreciably lower for rape seed oil as fuel.

ESC cycle	CO	NO _x	HC	PM
	g/kWh			
Gas oil	0.11	3.0	0.029	0.033
Rape seed oil	0.03	3.4	0.017	0.013

Table 5: Comparison of exhaust gas components during ESC cycle of gas oil and rape seed oil as fuel

AMES tests

This engine served primarily for the sample collection of the particulate matter for the AMES tests. Particulate emissions have been collected from rape seed fuel emissions and gas oil emissions at 3,300 rpm and 50 kW. The exact AMES procedure is described, e.g. in [8]:

- Samples were extracted in dichlor methane (DMC)
- The extraction took place in a soxhlet apparatus at 75°C and for a period of 9.5 Stunden.
- 70 ml DMC are applied per sample
- The extracts are dissolved after vacuum evaporation in 3.68 ml dimethyl sulfoxide (DSMO)

The following concentrations could be calculated by the difference of the filter weights:

Rape seed oil: 0.3 g/l
Gas oil: 0.175 g/l

These concentrations were used as initial solutions for the mutagenic Ames tests. The bacteria phylums TA 98, TA 100 und TA MIX were applied for the investigations. They were tested with and without S9-Mix.

The extracts were diluted in several steps, always by a factor of 3.5 for the Ames tests, table 6. The concentrations have

been selected according the comprehensive AMES investigation by the "Technologie und Förderzentrum Bayern" [10]. Different substances as positive check were used specific to the phylum, table 7. The solvent DSMO served as negative check.

Dilution-step	Dilution	Concentration rape seed oil extract	Concentration gas oil extract
		g/l	g/l
1	1	0.30	0.175
2	0.286	0.086	0.05
3	0.0816	0.024	0.014
4	0.023	0.007	0.004
5	0.0067	0.002	0.001
6	0.0022	0.0006	0.0003

Table 6: Dilution steps of PM for AMES tests

Phylum	Positive check
TA 98 -S9	2-NF, 2000 ng/ml (2-Nitrofluoren)
TA 100 - S9	4-NQO, 100 ng/ml (4-Nitroquinolin-N-Oxid)
TA Mix -S9	4-NQO, 500 ng/ml
+S9, all phylums	2-AA 5000 ng/ml (2-Amino-anthrazen)

Table 7: Substances for positive check

Figures 3.10-10 to 3.10-13 visualize for one test series the number of revertants for the PM emitted by plant oil and gas oil fuels with bacteria phylums TA100 - S9 and TA100 + S9 with the different dilution steps. The tests with the other phylums listed in table 7 showed quite similar results.



Fig. 3.10-10: Number of revertants with bacteria phylums TA 100 - S9 for PM from rape seed oil fuel

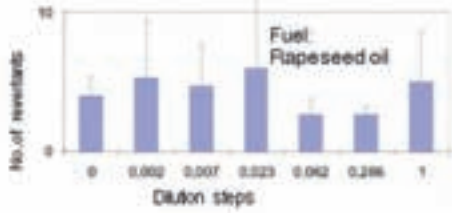


Fig. 3.10-11: Number of revertants with bacteria phylums TA 100 + S9 for PM from rape seed oil fuel

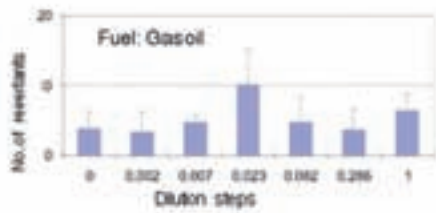


Fig. 3.10-12: Number of revertants with bacteria phylums TA 100 - S9 for PM from gas oil fuel

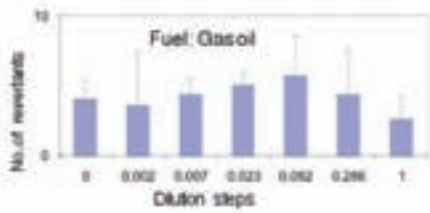


Fig. 3.10-13: Number of revertants with bacteria phylums TA 100 + S9 for PM from gas oil fuel



Fig. 3.10-14: Test vehicle VOLVO truck FH 480

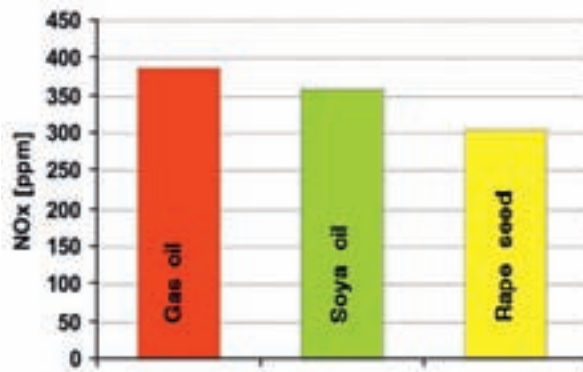


Fig. 3.10-15: Averaged NO_x emission of the VOLVO truck during one city traffic cycle

The number of revertants suggests that the number of revertants may be slightly higher for PM from gas oil emissions. However, all samples are in the range of the negative control sample. Furthermore, a dose effect could not be detected as the different dilution samples did not reveal any significant change in the detected revertant number. Therefore, no quantitative statements to different health impacts of the different fuels can be made. But it can be stated that there is no significant difference between the biological hazard of the emitted particulate matter. This may be true for all complete combustion processes as the nature of the soot is the same. However, strong differences may reveal in the emitted PM from incomplete combustion. The different particle characteristics are visualized in [4, 11]. The publication [9] reports AMES tests from a diesel engine operated by plant oil, but without pre-warming the plant oil. The caused

incomplete atomisation of the fuel and thus incomplete combustion must have generated PM emissions with a high content of unburned HC molecules of different nature as reported in [10]. Consequently, the AMES tests in [9] revealed high numbers of revertants, i.e. high mutagenic effects. A non-modified diesel engine can not be operated for a longer period of time with plant oil due to coking. Therefore, the results of [9] are of no relevance. A deeper discussion can be found in [12].

VOLVO TRUCK

The test vehicle is shown in Figure 3.10-14. The truck was operated in real city traffic. The route was repeated during the day.

The NO_x and HC emissions have been measured during the tests. Figures 3.10-15 and 3.10-16 summarize the averaged results per driving cycle for gas oil, soya oil and rape seed oil as fuels.

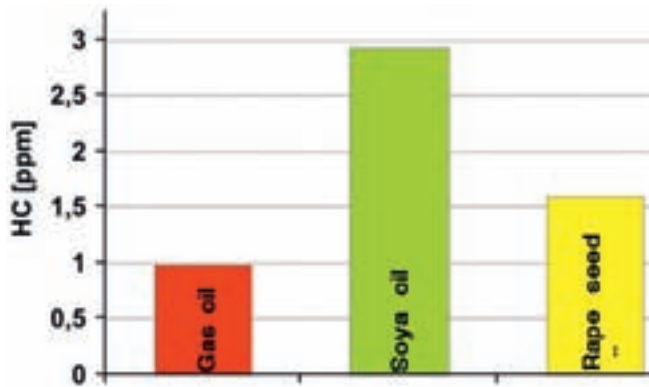


Fig. 3.10-16:
Averaged HC emission
of the VOLVO
truck during one city
traffic cycle

Summary

The emissions of three different diesel engines operated with gas oil and plant oils have been measured and compared. The emissions from plant oil operations achieved generally lower values compared to the operation with conventional fuel. The CO, HC and PM emissions were appreciably lower. NO_x is the exception – this emission was typically 10 % higher for plant oils compared to gas oil.

The AMES test revealed no significant difference in the mutagenic effect of the emitted particulate matter. Even so the concentration of the PM was selected like in a previous investigation with a tractor engine [10], the number of detected revertants (measure of mutagenic effect) was not above the negative control substance. This is in contrast to [10] where enough revertants have been found. It may be due to the better engines in this investigation (EURO 4 and EURO 5 engines). Anyway, the emissions from plant oil fuels did not exhibit real differences to the gas oil emissions in results of the AMES tests.

There are only few AMES investigations of PM emissions of diesel engines reported. The investigation [9] is not relevant as it deals with cold injected plant oil which will damage the engine in a short time. The mutagenic effect of the emitted particles is only high for incomplete combustion (see also [4]). Comprehensive investigations with correctly modified diesel engines for plant oils are described in [10] and [13]. They come to the conclusion that the mutagenic effects of rape seed oil fuel emission is by a factor of 2.5 to 1.1 lower than the effect found with conventional gas oil. Thus, the health impact should be lower with rape seed oil.

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