

# Characterization of the polycyclic aromatic hydrocarbons emitted from a compression ignition engine powered with biofuels of the 1<sup>st</sup> and 2<sup>nd</sup> generation

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## 1. Introduction

Motor vehicle emissions are an important source of atmospheric air pollution, contributing to the development of respiratory system diseases, tumours, and genetic changes. International regulations discern some substances that are harmful to the environment, such as carbon monoxide (CO), unburnt hydrocarbons (HC), nitrogen oxides (NO<sub>x</sub>), or particulate matter (PM). The emissions of the chemical compounds mentioned above from most vehicles are subject to limitations imposed by European standards. The limits depend on vehicle type and have been laid down in standard requirements referred to as Euro 5, Euro 6, etc. [1]. The acceptable emissions of other environmentally harmful substances are reduced by successive introduction of increasingly stringent quality requirements, which limit the sulphur, benzene, or polycyclic aromatic hydrocarbons (PAHs) contents of motor fuels.

Among the fuels used in transport, diesel oil is the one that significantly contributes to the emission of particulate matter, with which PAHs are introduced among other pollutants into the environment. The PAHs make a group of over 100 chemical compounds and about 16 of them have been indicated by the United States Environmental Protection Agency (US EPA) and International Agency for Research on Cancer (IARC) as substances proven to produce or potentially capable of producing carcinogenic and mutagenic effects.

The aromatic hydrocarbons come from unburnt or partly burnt fuel and lubricating oil and their sorts and quantities depend on engine type, vehicle load and speed, fuel composition, and effectiveness of the exhaust treatment system. Research carried out [2] has revealed that the unburnt diesel oil is a source of PAHs having low molecular mass (acenaphthalene, fluorene), while PAHs with medium molecular mass (e.g. fluoranthene, pyrene) and high molecular mass (e.g. benzo(b) fluoranthene, benzo(a)pyrene) are products of fuel combustion [3, 4]. Experiments carried out by Rhead et al. [5] have shown that during the combustion process, the products of partial fuel combustion undergo pyrosynthesis and form new aromatic compounds, which originally were not present in the fuel.

According to research works done by other teams [6, 7], aromatic hydrocarbon derivatives, such as nitro-PAHs and oxy-PAHs, may be directly emitted from the engine, formed in reactions with the OH and NO<sub>3</sub> groups derived from the atmospheric air, or formed in result of heterogenic reactions between PAHs and ozone.

The fatty acid methyl esters (FAME) introduced into widespread use were expected to reduce the emissions of regulated harmful

substances; the emissions of aromatic hydrocarbons were also to be reduced in comparison with those observed for fossil fuels being in use because the methyl esters practically do not contain aromatic hydrocarbons. The information available from the literature and concerning the impact of biodiesel on PAH emissions is insufficient and often contradictory. Admittedly, many researchers observed some reduction in the PAH emissions, but this was closely related to engine operation conditions [8, 9]. George Karavalakis et al. [10], who carried out comparative investigation of the emissions of PAHs produced by the combustion of blends of diesel oil and three sorts of methyl esters (obtained from soybean oil, palm oil and rapeseed oil), have shown that the origin and quality of a biofuel has an impact on the sort and quantity of the PAHs emitted. The results of their works have revealed that in comparison with the diesel oil, the esters contributed to an increase in the emissions of PAHs with low molecular mass and to a decrease in the emissions of PAHs with higher molecular mass. The total PAH emissions from mixtures of methyl esters obtained from waste oils were higher than those from mixtures of diesel oil and other biofuels obtained from fresh feedstock [11]. In researchers' opinion, this is related to higher susceptibility of esters obtained from waste oils to oxidation.

Another sort of the biocomponents that do not contain aromatic hydrocarbons is the hydrogenated vegetable oil (HVO), obtained from vegetable oils unsuitable for food production and/or from waste animal fats. Soo-Young No has indicated in his review article [12] on the use of HVO for the fuelling of compression ignition (CI) engines that most experiments revealed a reduction in the NO<sub>x</sub>, PM, HC, and CO emissions in comparison with such emissions from the conventional diesel oil and biodiesel used for the fuelling of heavy vehicles. Anja Singer et al. [13], who analysed the toxicity of polycyclic aromatic hydrocarbons emitted from a heavy vehicle powered with pure HVO, have shown that the PAHs thus emitted were characterized by a lower toxicity coefficient in comparison with the PAHs emitted in result of the combustion of esters and diesel oil. Similar relations were revealed by them at tests carried out for light vehicles meeting the Euro 3 requirements. By contrast, the toxicity coefficient of the PAHs emitted from the combustion of HVO in vehicles meeting the Euro 6 requirements was higher than that of the PAHs emitted from the combustion of petroleum-derivative diesel oil.

Duckhan Kim et al. [14] carried out investigations on pollutant emissions, including PM emissions, from the combustion of various fuel blends in a CI engine on a chassis dynamometer. The measurements revealed that when the engine was fed with diesel oil with 10–30 % HVO content then the particulate matter emission was reduced by about 12–15 % by mass in comparison with that measured for pure petroleum-derivative diesel oil. Comparative

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dynamometer tests of fuel blends with HVO and FAME contents and pure diesel oil as a reference fuel were also carried out by Federico Millo et al. [15]. These researchers showed that the PM emission dropped for a fuel blend consisting in 30 % of HVO and in 70 % of diesel oil at all engine test loads; for a similar fuel sample with FAME content, a drop in the PM emission in comparison with that from the diesel oil was only observed at higher engine loads. The authors explained this fact by the qualitative characteristics of FAME, especially low volatility and high final boiling point.

This paper presents qualitative and quantitative characteristics of polycyclic aromatic hydrocarbons emitted from the exhaust system of a motor vehicle with a compression ignition engine. The experiments described here were carried out within a project aimed at evaluation of the impact of fuel biocomponents, i.e. fatty acid methyl esters (FAME) and hydrogenated vegetable oil (HVO), on the air quality in urban areas and on the potential health effects.

## 2. Experiments

To determine the characteristics of the polycyclic aromatic hydrocarbons formed during the fuel combustion process, a test rig was prepared, which consisted of a Fiat Panda car with a four-cylinder compression ignition engine model I.3 Multijet. The vehicle met the Euro 5 standard requirements. The exhaust system of the car was appropriately modified to enable particulate matter collection both when the vehicle was provided with a diesel particle filter (DPF) and a catalyst and when it was operated without the filter and catalyst. The measurements were carried out in static engine operation conditions, at vehicle speed of 43.75 km/h, engine speed of 1340 rpm, accelerator pedal position of 20 %, engine load equal to 45.7 %, and engine coolant temperature of 94°C.

The vehicle was fed with three fuel types in succession. As the reference fuel, the commercial diesel oil was adopted. The other two fuel blends were composed for project purposes and contained biocomponent admixtures, i.e. fatty acid methyl esters (FAME) and hydrogenated vegetable oil (HVO). The composition of the test fuels was as follows:

- commercial diesel oil with up to 7 %, by volume, FAME content (B7),
- biofuel with 20 %, by volume, FAME content (B20),
- biofuel with 7 % and 13 %, by volume, FAME and HVO contents, respectively (SHB20).

Before commencing the engine tests, the fuel blends prepared were tested in order to determine their quality characteristics as specified in Polish Standard PN-EN 590 [16]. Selected results of these tests have been given in Table 1.

The particulate matter emitted during engine operation was collected on filters made of glass fibre coated with PTFE (EMFAB TX40HI20WW 70MM), installed in the vehicle exhaust system, which was specially modified. After a particulate matter sample was taken, the filter was stored in a closed vessel at a temperature of about –18°C until the measurements were carried out. The procedure of identification and determination of individual aromatic hydrocarbons consisted of several stages. At the first stage, the PAHs were extracted in an ultrasonic bath with the use of a hexane/acetone mixture (4 : 1) and the extract was dried. At the second stage, the PAHs were separated from the said extract by means of extraction to the solid phase with the use of hexane and dichloromethane. At the last stage, the PAH solution in dichloromethane was concentrated to a volume of 1 ml and then the PAH contents were determined with the use of a gas chromatograph 7890A GC coupled with a mass spectrometer MS 5975C. The contents of the following aromatic hydrocarbons in the extract were determined: naphthalene, acenaphthylene,

acenaphthalene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benzo(a)anthracene, chrysene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)fluoranthene, benzo(a)pyrene, indeno(1,2,3-c,d)pyrene, dibenzo(a,h)anthracene, benzo(g,h,i)perylene. The results of measurements of PAH contents given in the table below have been calculated per one hour of motor vehicle operation.

Table 1

Selected standardized characteristics of the biofuels and diesel oil under test

Parameter	B20	SHB20	B7	Test method
Cetane number	53.6	53.1	52.5	EN ISO 5165
Density at 15 °C [kg/m <sup>3</sup> ]	843.3	829.9	839.5	EN ISO 12185
PAHs content [% (m/m)]	1.1	1.3	1.5	EN 12916
Sulphur content [mg/kg]	4.6	4.8	5.7	EN ISO 20846
Flash point [°C]	66.5	62.0	61.0	EN ISO 2719
Water content [mg/kg]	150	70	80	EN ISO 12937
FAME content [% (v/v)]	20.8	5.7	6.7	EN 14078
Oxidation stability [g/m <sup>3</sup> ]	1	5	5	EN ISO 12205
Viscosity at 40 °C [mm <sup>2</sup> /s]	3.088	2.983	3.002	EN ISO 3104
Cold filter plugging point (CFPP) [°C]	-7	-30	-30	EN 116
Lubricity at 60 °C [μm]	199	276	218	EN ISO 12156-1

## 3. Measurement results

### 3.1. Exhaust system without a particulate matter filter

In the tests carried out during the operation of a motor vehicle without a particulate matter filter and a catalyst installed in its exhaust system, the largest amount of PAHs (5 450 ng/ml) was detected in the exhaust gases emitted when the diesel oil B7 was used as a fuel. A lower PAH content (2 703 ng/ml) of the exhaust gases was recorded for the B20 fuel and the lowest PAH emissions (2 148 ng/ml) occurred during the combustion of the SHB20 fuel. It has been found that pyrene predominated among all the PAHs detected in the exhaust gases produced in result of the combustion of all the three fuel blends under test, but for the B7 fuel, the pyrene content was more than twice as high as that recorded for the B20 and SHB20 fuels. In the extracts of the particulate matter obtained from the combustion of the B7 fuel, considerable amounts of fluoranthene, phenanthrene, and chrysene were detected. Exact values of the emissions of all the PAHs identified have been specified in Table 2.

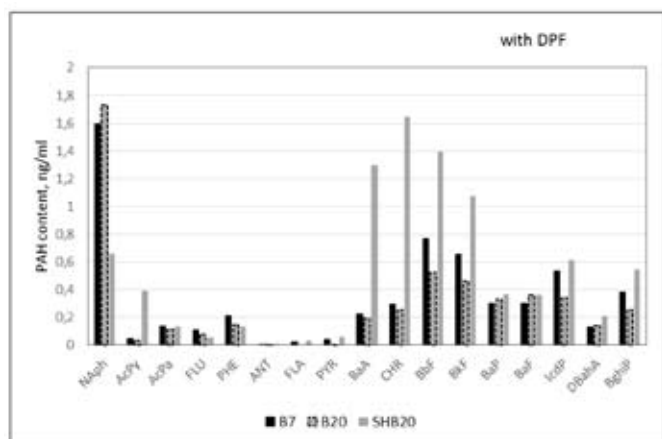
The exhaust gases generated by the combustion of the B20 fuel in comparison with those obtained from the B7 fuel contained less fluoranthene, pyrene, benzo(a)anthracene, chrysene, benzo(b)fluoranthene, benzo(k)fluoranthene, and benzo(g,h,i)perylene, with the contents of the other PAHs being higher. Although the total PAH emissions from the combustion of the B20 fuel were lower than those recorded for the commercial diesel oil B7, the fuel with 20 % FAME content generated more benzo(a)pyrene than the other fuels under test did.

**Table 2**  
PAH emissions for the B7, B20 and SHB20 fuel. Exhaust system without a DPF

	PAH compound	Abbreviation	Unit	Without a DPF		
				B7	B20	SHB20
1	Naphthalene	NAph	ng/ml	127	223	96
2	Acenaphthylene	AcPy		8.5	26.5	4.2
3	Acenaphthalene	AcPa		3.6	5.5	2.2
4	Fluorene	FLU		18.3	40.6	7.2
5	Phenanthrene	PHE		405.5	600	157.5
6	Anthracene	ANT		37.3	49.8	17.3
7	Fluoranthene	FLA		1 185	174.5	195.5
8	Pyrene	PYR		2 970	1 245	1 245
9	Benzo(a)anthracene	BaA		68	32.9	54
10	Chrysene	CHR		337.5	114.5	212.5
11	Benzo(b)fluoranthene	BbF		141.5	47.6	86
12	Benzo(k)fluoranthene	BkF		85	45.4	42.4
13	Benzo(a)pyrene	BaP		12.5	23.8	6.9
14	Benzo(a)fluoranthene	BaF		21.4	45.2	8.3
15	Indeno(1,2,3-c,d)pyrene	IcdP		12	12.8	5.7
16	Dibenzo(a,h)anthracene	DBahA		2.2	6.8	1.4
17	Benzo(g,h,i)perylene	BghiP		15.6	9.6	6.1
Total PAHs				5 450.9	2 703.5	2 148.2

### 3.2. Exhaust system with a diesel particle filter (DPF)

In the tests carried out during the operation of a motor vehicle with a particulate matter filter and a catalyst installed in its exhaust system, the largest amount of PAHs (9.008 ng/ml) was detected in the exhaust gases emitted when the SHB20 fuel was used. A lower PAH content (5.782 ng/ml) of the exhaust gases was recorded for the B7 fuel and the lowest PAH emissions (4.982 ng/ml) occurred during the combustion of the B20 fuel. In the exhaust gases obtained from the B20 fuel, a compound of considerable importance was naphthalene (1.733 ng/ml) and its amount was higher than that recorded for the commercial diesel oil B7 (1.600 ng/ml). The benzo(a)pyrene and benzo(a)fluoranthene contents in the B20 combustion gases were also higher. In contrast, the contents of the other PAHs identified were lower. In the SHB20 fuel combustion gases, the following PAHs predominated: chrysene, benzo(b)fluoranthene, benzo(a)anthracene, benzo(k)fluoranthene, naphthalene, indeno(1,2,3-c,d)pyrene, and benzo(g,h,i)perylene. A comparison between the contents of individual PAHs produced during the combustion of the



three biofuels under test has been presented in Figure 1.

**Fig. 1. Comparison between the contents of individual PAHs produced during the combustion of the B7, B20, and SHB20 fuels in the system with a DPF**

During the combustion of all the three fuels under test, the benzo(a)pyrene contents in the exhaust gases were found to be at a similar level. The BaP contents detected in the extracts of the particulate matter collected during the combustion of the B7 (commercial diesel oil), B20, and SHB20 fuels were 0.304 ng/ml, 0.329 ng/ml, and 0.370 ng/ml, respectively. The differences were not very big; however, the BaP amount detected was the greatest for the SHB20 fuel.

### 4. Conclusions

The experiments carried out for the exhaust system without a particulate matter filter and a catalyst have shown that the biofuel containing a significant amount of biocomponents, i.e. fatty acid methyl esters (FAME) or hydrogenated vegetable oil (HVO), produces smaller quantities of polycyclic aromatic hydrocarbons (PAHs) than the commercial diesel oil (with up to 7 % FAME content) does. Nevertheless, in terms of one of the air pollution criteria, i.e. benzo(a)pyrene content of the exhaust gases, the biofuel with 20 % FAME content generated more BaP as against the biofuel containing HVO and the commercial diesel oil.

The installation of a particulate matter filter (DPF) in the exhaust system of the vehicle used for the tests brought about a reduction in the quantity of the particulate matter, and in consequence the PAHs, emitted to the environment to a many times lower level. However, contrary to expectations, the biofuel containing hydrogenated vegetable oil did not help to reduce the total emissions of aromatic compounds; just the opposite, the total content of aromatic compounds in the exhaust gases was higher than that recorded for the diesel oil and for the biofuel with the same FAME content. The experiment results suggest that the HVO combustion products are more susceptible to the processes that lead to the PAH formation than the products of combustion of FAME and petroleum-derivative hydrocarbons.

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#### RYNEK

##### Wyniki BASF w drugim kwartale 2016 r.

W drugim kwartale 2016 BASF odnotował nieznaczny poprawę w otoczeniu makroekonomicznym. Wzrost cen ropy naftowej i wzrost popytu od końca marca br. wskazują na rozwój. *Odnotowaliśmy solidny popyt, szczególnie w przemyśle samochodowym i budownictwie. Jednak sytuacja makroekonomiczna pozostaje trudna do przewidzenia* – powiedział dr Kurt Bock, przewodniczący zarządu wykonawczego BASF SE.

Sprzedaż grupy BASF zmalała o 24% w drugim kwartale, do 14,5 mld EUR, w porównaniu do tego samego okresu w 2015 r. Dwie trzecie tego spadku to rezultat efektów portfelowych (minus 16%). Główną przyczyną było zbycie aktywów obrotu gazem i działalności magazynowej w ramach wymiany aktywów z Gazpromem na koniec września 2015 r. Dodatkowo niższe ceny surowców, szczególnie w sektorze chemikaliów (Chemicals), doprowadziły do spadku cen sprzedaży (o 7%). Poza rozwiązaniami w sektorze rolnictwa (Agricultural Solutions), wszystkie segmenty wpłynęły na lekki wzrost poziomu sprzedaży (o 2%). W sektorze produktów chemicznych, który obejmuje segmenty Chemikalia (Chemicals), Wyroby Przetworzone (Performance Products) oraz Materiały i Rozwiązania Funkcjonalne (Functional Materials & Solutions), wolumeny wzrosły o 4%. Negatywne skutki walutowe dały się zauważyć we wszystkich działach (minus 3%).

W drugim kwartale br. zysk operacyjny (EBIT) przed kosztami nadzwyczajnymi zmniejszył się o 336 mln EUR do 1,7 mld EUR w porównaniu do znakomitego analogicznego kwartału ubiegłego roku. Znacząco wyższe zarobki w sektorze produktów chemicznych nie były w stanie zrekomensować znaczenie niższych dotacji z segmentu naftowego i gazowego. W porównaniu z poprzednim drugim kwartałem, EBIT zmniejszył się o 321 mln EUR do 1,7 mld EUR. Zysk netto zmniejszył się o 173 mln EUR do 1,1 mld EUR. Wskaźnik zysku na akcję wyniósł 1,19 EUR w drugim kwartale 2016, w porównaniu do 1,38 EUR w analogicznym okresie 2015 r.

Dostosowane do kosztów nadzwyczajnych i amortyzacji wartości niematerialnych, wskaźnik zysku na akcję wyniósł 1,30 EUR (drugi kwartał 2015: 1,49 EUR). (kk)

(<https://www.basf.com/>, 27.07.2016)

##### LANXESS podwyższa prognozy zysków za 2016 r.

Producent specjalistycznych środków chemicznych, koncern LANXESS, podwyższył prognozę zysków za rok 2016. Spółka przewiduje, że wskaźnik EBITDA przed uwzględnieniem pozycji nadzwyczajnych wyniesie pomiędzy 930 mln EUR a 970 mln EUR. Nowa prognoza jest przede wszystkim odpowiedzią na dobre perspektywy handlowe w nowych segmentach spółki na drugą połowę roku. Wcześniej zakładano zyski pomiędzy 900 mln EUR a 950 mln EUR.

W silnym drugim kwartale 2016 r. wskaźnik EBITDA przed uwzględnieniem pozycji nadzwyczajnych wzrósł o 8,5% do 293 mln EUR wobec 270 mln EUR rok wcześniej. Marża EBITDA przed uwzględnieniem pozycji nadzwyczajnych wzrosła rok do roku z 12,8% do 15,1%. Dobre wyniki ogólne spółka zawdzięcza przede wszystkim silnemu rozwojowi nowych segmentów LANXESS, tj. zaawansowane środki pomocnicze (*Advanced Intermediates*), specjalistyczne środki chemiczne (*Performance Chemicals*) i materiały o wysokiej wydajności (*High Performance Materials*).

– *Solidny wynik na działalności operacyjnej to efekt przede wszystkim wzrostu wolumenów, odchudzenia struktur kosztowych i poprawy oferty produktów poprzez dodanie „nowych” segmentów. To pokazuje, że dokonane przez nas zmiany umożliwiły nam stworzenie silnej i sprawnie działającej organizacji oraz że oferujemy właściwe produkty na właściwych rynkach* – uważa Matthias Zachert, Prezes Zarządu koncernu LANXESS. – *Z dużym optymizmem podchodzimy do drugiej połowy roku i spodziewamy się, że „nowy” LANXESS poprawi zyski w stosunku do roku poprzedniego. Pomimo trudnej sytuacji w branży kauczuku ponownie podwyższamy prognozę na cały rok.* (abc)

(więcej na [www.miesiecznikchemik.pl/Aktualności](http://www.miesiecznikchemik.pl/Aktualności))

(inf. Prasowa LANXESS, 11.08.2016)

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