



Polycyclic aromatic hydrocarbons in crude oils from Poland

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The content of 17 polycyclic aromatic hydrocarbons (PAHs) in oil samples derived from all three oil-bearing regions of Poland, i.e. from the Carpathians, the Carpathian Foredeep and the Polish Lowlands (the Fore-Sudetic-Wielkopolska, the Western Pomeranian and the Peribaltic areas), were determined. The concentrations of PAHs determined in these crude oils varied from 22.85 to 332.45 ppm. The average content of PAHs in crude oils from the Carpathians and the Carpathian Foredeep (191.18 ppm) was several times higher than that in crude oils from the Polish Lowlands (44.70 ppm); there was throughout, though, a relatively small variability in the content of perylene, varying from 0.50 to 0.80 ppm. Of PAH compounds determined, the largest share was occupied by 3-cyclic hydrocarbons (an average of 72%) and 4-cyclic (about 24%), whereas the content of 5- and 6-cyclic hydrocarbons did not exceed 3%. Phenanthrene and fluorene were prevailing PAH compounds. The diverse PAH composition in oil samples from the Polish Lowlands compared to those from the Carpathians and the Carpathian Foredeep seems to have resulted more from the composition of original organic matter than from the course of geochemical and biochemical processes (which occur during biomass decay) and the subsequent migration of these compounds in rock formations.

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INTRODUCTION

The polycyclic aromatic hydrocarbons (PAHs), many of which possess toxic, mutagenic and carcinogenic properties, are pollutants commonly present in small amounts in all environmental systems (McGroddy and Farrington, 1995; Ollivon *et al.*, 1995; Maliszewska-Kordybach and Terelak, 1996; Bojakowska and Sokołowska, 1998; Harvey, 1998). Some of these polyarenes present in the environment are of natural origin; they occur in plants or are the products of metabolic processes in microorganisms that decompose organic remains; in addition, they originate during geochemical and biochemical processes that lead to the formation of fossil fuels (including crude oil generation) or are synthesised during volcanic eruptions, and forest, bush and peat fires (Evans *et al.*, 1990; Capaccioni *et al.*, 1995; Jiang *et al.*, 1998; Koziński and Saade, 1998; Harvey, 1998; Howsam and Jones, 1998). However, most PAHs in recent soils and sediments originate from

anthropogenic sources, particularly as the result of burning fuels for domestic, industrial and transport needs, coal coking and oil treatment, and steel and aluminum manufacturing. PAHs also reach the environment via liquid fuel spills during transport, storing or car handling (Behymer and Hites, 1988; Harvey, 1998; Howsam and Jones, 1998).

Polycyclic aromatic hydrocarbons occur in the environment as a mixture of compounds containing two or more aromatic (benzene) rings. Their particular composition varies according to rock and sediment type. The greatest hazard is caused by high molecular compounds, especially by those containing five or more rings. These harm and degrade slowly in the environment, and so accumulate in soils and recent sediments. If the composition of a PAH mixture in fossil fuels is affected by the composition of original material, the depositional conditions (pH, Eh, temperature), the course of biochemical processes during organic decay and the course of carbonification and hydrocarbon generation after burial, the

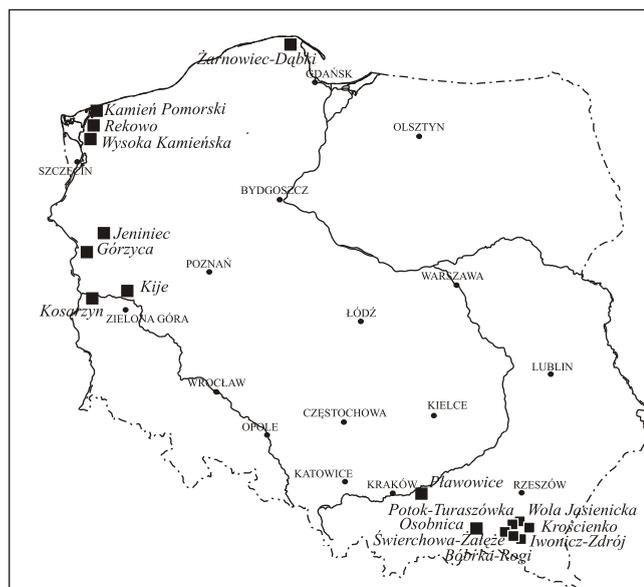


Fig. 1. Location of crude oil deposits sampled

PAH composition in recent soils and sediments is largely linked to the products of burning released to the atmosphere.

Crude oils generally contain several percentage of PAHs, with substituted naphthalene and phenanthrene being prevalent. The proportion of unsubstituted PAH is relatively small; in general, crude oils are characterised by a very low content of 5- and 6-cyclic aromatic hydrocarbons of this type (Bence *et al.*, 1996). The highest concentrations of high molecular PAHs (about several hundreds ppm) were recorded in hydrothermal oils in the Gulf of California; these were generated by alteration of organic matter at temperatures of 260–350°C (Simoneit and Fetzer, 1996). The prevalence of 4-, 5- and 6-cyclic aromatic hydrocarbons over 2- and 3-cyclic ones, and compounds unsubstituted over alkylated compounds is also characteristic for products of oil combustion as opposed to non-combusted oils (Bence *et al.*, 1996). Of the PAH mixture contained in exhaust gases, about 50% belong to compounds which “survived” combustion, even though the “survival rate” of individual PAH compounds during burning varies from 0.04 to 0.87% (Beak *et al.*, 1991; Tancell *et al.*, 1995; Howsam and Jones, 1998).

There is little information on the occurrence of environmentally important PAHs in fossil fuels. The abundant literature on the organic chemistry of ancient environments deals mostly with alkylated PAHs such as methylphenathrenes, good markers that enable determinations of oil maturity and its source of origin (Pu *et al.*, 1990; Ramanampisoa and Radke, 1995; Smith *et al.*, 1995; Requejo *et al.*, 1996). Yet knowledge of the ranges in concentration of high molecular PAHs in crude oils is essential because oil exploitation, transport, storing and processing, as well as combustion of crude oils and their derivatives, are a significant source for releasing these compounds to

the environment. There has been a lack of information on the concentrations of unsubstituted PAHs in crude oils from Polish deposits.

SCOPE AND METHODS OF INVESTIGATION

The content of PAHs was determined in 30 samples of crude oils derived from all three oil-bearing regions of Poland, i.e. from the Carpathians, the Carpathian Foredeep and the Polish Lowlands (the Fore-Sudetic-Wielkopolska, the Western Pomeranian and the Peribaltic areas).

In the Carpathians 18 samples from deposits, located in the Silesian and the Subsilesian units, were examined. These samples were derived from the following deposits: Krościenko, Potok-Turaszówka and Bóbrka-Rogi in which oil-bearing horizons occur within the Ci kowice Sandstones, Iwonicz-Zdrój and Osobnica the oils of which occur within the Ci kowice and the Istebna Sandstones, and Wola Jąsienicka in which oil-bearing horizons occur within Early Cretaceous rocks, i.e. the W głowice Marls and Sandstones (Depowski, 1987). These crude oils were generated primarily from organic matter contained in Late Cretaceous and Tertiary host rocks (ten Haven *et al.*, 1993). In the Carpathian Foredeep 3 oil samples from the Grobla-Pławowice deposit located north of Bochnia were examined. In this deposit crude oil has accumulated in Cenomanian sandstones and Oxfordian limestones. In the Western Pomeranian area 3 oil samples (from the Rekowo, Kamie Pomorski and Wysoka Kamie ska deposits) and in the Fore-Sudetic-Wielkopolska area 4 samples (from the Jeniniec, Górzycza, Kije and Koszarzyn deposits) were investigated. Oil deposits in the Fore-Sudetic-Wielkopolska and the Western Pomeranian areas relate to the Zechstein Main Dolomite; hydrocarbons generated from this lithostratigraphic unit are recognised as autochthonous (Karnkowski, 1993). In the Łeba Elevation of the middle Peribaltic area, where oil-bearing horizons occur in the upper part of Middle Cambrian quartz sandstones, 2 oil samples from the Żarnowiec-Dąbki deposit were studied (Karnkowski, 1993). The location of these sampled oil deposits is shown in Figure 1.

In oil samples derived from these deposits 17 PAHs, i.e. acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benzo(a)anthracene, chrysene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(e)pyrene, benzo(a)pyrene, perylene, indeno(1, 2, 3-cd)pyrene, dibenzo(ah)anthracene, benzo(ghi)perylene, are listed in environmental studies by the US EPA (US Environmental Protection Agency). The fraction of aromatic hydrocarbons was obtained using a columnar chromatography method (Al_2O_3 , SiO_2). The analyses were performed using a Hewlett Packard gas chromatograph type 5890 II with a mass spectrometer GS-MSD 5981 detector. A HP-5 non-polar capillary column (length — 25 m, diameter — 0.2 mm, film — 0.33 μm) (5%)-diphenyl-(95%)-dimethylpolysiloxane was applied. The temperature was set from 70 to 200°C with a rate of 10°C/min and from 200 to 300°C with a rate of 2.5°C/min. The

Table 1

PAHs in crude oils of different regions from Poland (ppm)

Hydrocarbons	Carpathians and Carpathian Foredeep regions (<i>n</i> = 21)	Polish Lowlands (Sudetic-Wielkopolska and Western Pomeranian areas) (<i>n</i> = 7)	Łeba Elevation (<i>n</i> = 2)
Acenaphthylene (Ace)	$\frac{< 0.15 - 3.97}{1.29}$	$\frac{< 0.15 - 1.56}{0.74}$	< 0.15
Acenaphthene (Acf)	$\frac{< 0.15 - 7.04}{2.47}$	$\frac{< 0.15 - 0.80}{0.39}$	< 0.15
Fluorene (Fl)	$\frac{7.25 - 83.94}{31.37}$	$\frac{2.25 - 14.38}{8.60}$	$\frac{1.86 - 6.48}{4.17}$
Phenanthrene (Fen)	$\frac{48.59 - 175.18}{102.42}$	$\frac{13.72 - 26.71}{21.26}$	$\frac{16.16 - 38.86}{27.51}$
Anthracene (Ant)	$\frac{0.98 - 3.37}{1.75}$	$\frac{0.58 - 1.92}{1.23}$	$\frac{0.33 - 0.92}{0.63}$
Fluoranthene (Flu)	$\frac{2.33 - 8.52}{4.70}$	$\frac{0.08 - 0.79}{0.41}$	< 0.15
Pyrene (Pir)	$\frac{4.14 - 18.92}{11.04}$	$\frac{0.69 - 3.56}{2.04}$	$\frac{1.13 - 3.72}{2.43}$
Benzo(a)anthracene (BaA)	$\frac{1.01 - 7.19}{3.47}$	$\frac{< 0.23 - 1.09}{0.27}$	< 0.23
Chrysene (Ch)	$\frac{7.48 - 51.28}{27.18}$	$\frac{2.02 - 26.30}{8.29}$	$\frac{6.51 - 14.43}{10.47}$
Benzo(b)fluoranthene (BbF)	$\frac{0.30 - 2.79}{0.94}$	$\frac{< 0.05 - 0.22}{0.09}$	$\frac{0.19 - 0.31}{0.25}$
Benzo(k)fluoranthene (BkF)	< 0.05	< 0.05	< 0.05
Benzo(e)pyrene (BeP)	$\frac{0.36 - 10.66}{3.12}$	$\frac{< 0.05 - 1.25}{0.40}$	$\frac{0.49 - 3.52}{2.01}$
Benzo(a)pyrene (BaP)	$\frac{< 0.05 - 1.22}{0.43}$	$\frac{< 0.05 - 0.12}{< 0.05}$	$\frac{< 0.05 - 0.29}{0.16}$
Perylene (Per)	$\frac{0.31 - 1.17}{0.65}$	$\frac{0.52 - 0.77}{0.62}$	$\frac{0.52 - 0.56}{0.54}$
Indeno(1, 2, 3-cd)pyrene (IndP)	$\frac{< 0.10 - 0.41}{0.10}$	< 0.10	< 0.10
Dibenzo(ah)anthracene (DahA)	$\frac{< 0.10 - 0.74}{0.14}$	$\frac{< 0.10 - 0.12}{< 0.10}$	< 0.10
Benzo(ghi)perylene (Bper)	$\frac{< 0.10 - 0.93}{0.14}$	$\frac{< 0.10 - 0.41}{0.12}$	$\frac{< 0.10 - 0.31}{0.18}$
Total PAH _A	$\frac{71.05 - 245.25}{139.29}$	$\frac{17.75 - 43.77}{32.22}$	$\frac{18.50 - 46.41}{32.46}$
Total PAH _B	$\frac{18.42 - 77.41}{46.38}$	$\frac{3.81 - 28.28}{11.01}$	$\frac{7.83 - 18.34}{13.08}$
Total PAH _C	$\frac{1.29 - 16.39}{5.50}$	$\frac{0.84 - 2.55}{1.40}$	$\frac{1.39 - 5.11}{3.25}$
Total PAH ₁₇ = PAH _A + PAH _B + PAH _C	$\frac{91.76 - 332.45}{191.20}$	$\frac{22.85 - 68.55}{44.70}$	$\frac{27.77 - 69.89}{48.83}$

$\frac{\text{min. content} - \text{max. content}}{\text{mean value}}$; total PAH_A — total of 3-cyclic aromatic hydrocarbons [acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene]; total PAH_B — total of 4-cyclic aromatic hydrocarbons [fluoranthene, pyrene, benzo(a)anthracene, chrysene]; total PAH_C — total of 5- and 6-cyclic aromatic hydrocarbons [benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(e)pyrene, benzo(a)pyrene, perylene, indeno(1, 2, 3-cd)pyrene, dibenzo(ah)anthracene, benzo(ghi)perylene]

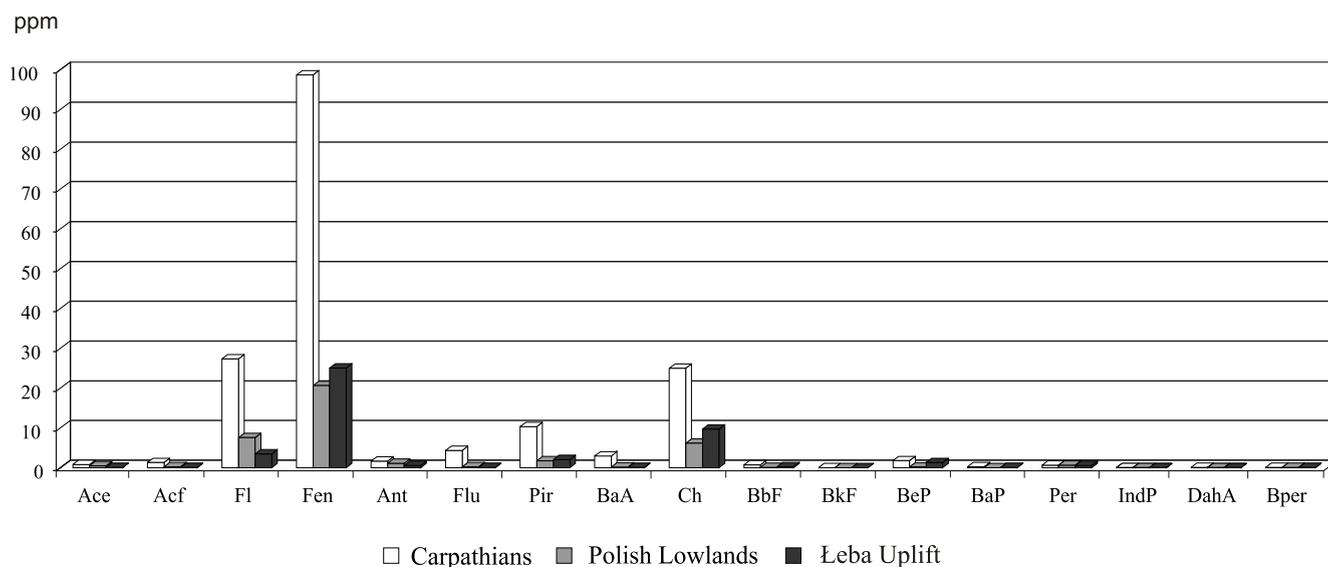


Fig. 2. PAHs in crude oils from different oil-bearing areas of Poland

For explanations see [Table 1](#)

analyses were performed using an outer standard method; the following standards were applied: PM-612 from ULTRA Scientific or certified perylene and benzo(a)pyrene. The samples were analysed at the Central Chemical Laboratory of the Polish Geological Institute.

RESULTS AND DISCUSSION

In the crude oils examined derived from these Polish deposits of different ages, the total content of 17 PAHs varied from 22.85 to 332.45 ppm ([Tab. 1](#)). The crude oils from deposits of the Fore-Sudetic-Wielkopolska, the Western Pomeranian and the middle Peribaltic areas, as well as from deposits of the Carpathians and the Carpathian Foredeep, showed distinct differences ([Fig. 2](#)). The crude oils from the Carpathians and the Carpathian Foredeep deposits, linked to Cainozoic and Mesozoic flysch rocks, were characterised by an average content of PAHs (191.20 ppm), which was four times higher compared to those derived from Palaeozoic rocks of the Polish Lowlands (44.70 ppm).

Of the 3-cyclic aromatic hydrocarbons examined (acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene), phenanthrene and fluorene showed the highest concentrations, whereas acenaphthylene the lowest. The crude oils from the Carpathians showed much higher concentrations of these hydrocarbons (phenanthrene — 102.42 ppm and fluorene — 31.37 ppm) compared to those from the Polish Lowlands containing 21.26 and 8.6 ppm, respectively ([Tab. 1](#)). By comparison, the crude oils in the Alaska North Slope pipeline coming from the oil fields of Prudhoe Bay, Kuparuk, Endicott and Lisburne, related to Triassic-Cretaceous shales, were featured by higher levels of phenanthrene (262.5 ppm) and fluorene (92.9 ppm) (Bence *et al.*, 1996). In the Polish crude oils, 3-cyclic aromatic hydrocarbons were dominant, reaching about 72% of the total PAHs. The lowest share of

these hydrocarbons was recorded in crude oils from the Wysoka Kamie ska borehole (Polish Lowlands), reaching 56.3%, and in those from the Pławowice deposit (Carpathian Foredeep) — 61.5–65.5%. Among the PAHs determined, phenanthrene was nearly constant constituting 53±6% of the total 17 compounds examined.

Of 4-cyclic aromatic hydrocarbons [fluoranthene, pyrene, benzo(a)anthracene, chrysene] determined, chrysene showed the highest concentrations, and benzo(a)anthracene the lowest. Considerably higher average levels of these compounds (46.38 ppm) were recorded in crude oils of the Carpathians and the Carpathian Foredeep compared to those from the Polish Lowlands (11.0 ppm). The Carpathian Foredeep oils also had a lower content of fluoranthene than did the Carpathians oils ([Tab. 1](#)). The 4-cyclic aromatic hydrocarbons formed on average of 24% of the total PAHs. The highest proportion of these compounds was recorded in the Wysoka Kamie ska borehole (Polish Lowlands) (41.3%) and from the Pławowice deposit (Carpathian Foredeep) (29.0–32.6%), and the lowest in the crude oils of the Górzycza (16.7%) and Kosarzyn (15.3%) boreholes in the Polish Lowlands.

Of 5- and 6-cyclic aromatic hydrocarbons, benzo(a)pyrene and benzo(b)fluoranthene showed the highest concentrations. The content of benzo(k)fluoranthene in the crude oils examined was below detection limits, whereas indeno(1, 2, 3-cd)pyrene and dibenzo(ah)anthracene were recorded only in crude oils from the following: Bóbrka-Rogi (Carpathians), Pławowice (Carpathian Foredeep), Górzycza (Polish Lowlands). The highest levels of 5- and 6-cyclic compounds were noted in crude oils of the Carpathians deposits at Grobla and Bóbrka. Most of the examined Carpathians crude oils contained benzo(b)fluoranthene (average 0.94 ppm) and benzo(a)pyrene (0.43 ppm), whereas in Zechstein crude oils of the Polish Lowlands, these compounds were commonly below detection limits. In all the oil samples examined perylene showed relatively small variation; its content in most samples

varied from 0.50 to 0.80 ppm. Perylene, a compound formed as a result of biotic and abiotic transformation of organic matter, is regarded as an indicator of anoxic sedimentary environments (Silliman *et al.*, 1998). The 5- and 6-cyclic aromatic hydrocarbons described make up only about 2.6% of the total PAH content in the crude oils examined.

The observed variation in PAH content was primarily caused by original organic composition. Hydrocarbons accumulated in rocks of the Main Dolomite (including the Wysoka Kamie ska and Kamie Pomorski deposits) were produced from sapropelic organic matter (kerogen) of type II (Kotarba *et al.*, 1998), whereas those in Carpathian flysch rocks include humic organic matter (kerogen of type III) (Czepiec and Kotarba, 1998). Primary differences may have been connected not only with the different chemical compositions of terrestrial and marine plants, but also with plant taxonomic differences in successive geologic eras.

Although the crude oils examined are characterised by much higher concentrations of PAHs compared to those from solid fossil fuels (their concentrations in crude oils run to tens or more ppm, whereas in peats there are hundreds ppb, and, in brown and hard coals, several ppm), they contain much less perylene than brown coals, and less benzo(a)pyrene than hard coals (Bojakowska and Sokołowska, 1999).

The proportion of individual compounds in the overall PAH composition is interesting. A nearly constant level of

perylene in the PAH mixture examined was observed; this prevailing PAH compound constitutes generally $53 \pm 6\%$ of the total PAHs. In addition, a greater share of high molecular compounds, i.e., benzo(b)fluoranthene and benzo(a)pyrene, was noted.

Both the diverse concentrations of PAHs in crude oils of the Polish Lowlands, the Carpathians and the Carpathian Foredeep deposits and the relatively small diversity in the content of perylene may indicate a relatively small influence of geochemical and biochemical processes taking place during biomass decay and subsequent hydrocarbon migration in rock formations, but rather primarily an influence of original organic matter composition.

A characteristic high share of 3-cyclic aromatic hydrocarbons, especially phenanthrene and fluorene in crude oils, may be useful for identifying environmental pollution sources by oil-derived products. In recent soils and aqueous sediments 4-, 5- and 6-cyclic compounds, especially fluoranthene, are prevalent (Bojakowska *et al.*, 1999). Thus an increase in the proportion of 3-cyclic aromatic hydrocarbons in environmental samples and an increase in the content of phenanthrene and fluorene versus fluoranthene may indicate pollution of the environment by oil-derived products.

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