

PAH VARIABILITY IN VERTICAL PROFILES FROM OTALŻYNO, HUCZWA AND STOCZEK PEATLAND SITES

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Abstract: The purpose of this study was to examine differentiation and variation of the spectrum of unsubstituted polycyclic aromatic compounds found in vertical profiles extracted from three peatland sites. Three peatland deposits were selected for the study: Otałżyno near Kartuzy (northern Poland), Stoczek near Radzyń Podlaski (central Poland) and Huczwa near Tomaszów Lubelski (south-eastern Poland). Polycyclic aromatic hydrocarbon content was determined by chromatography. Determination of hydrocarbon content in the extracts identified 17 compounds (15 of which are included on the so-called “list” of priority PAHs that EPA recommends should be targeted for measurement in the natural environment). Peats from the Otałżyno deposit (a raised bog) are characterised by lower PAH content – 436 $\mu\text{g}\cdot\text{kg}^{-1}$ compared with peats from the Huczwa and Stoczek (fens), where PAH content is 754 and 567 $\mu\text{g}\cdot\text{kg}^{-1}$, respectively. The PAH spectrum of the peats studied is dominated by tricyclic compounds, primarily by phenanthrene. This is in contrast with the PAH spectra of modern deposits in which tetra- and pentacyclic compounds account for the majority of polycyclic aromatic hydrocarbons. Bog peats have higher acenaphthene content, but at the same time have relatively low levels of fluoranthene relative to phenanthrene, if compared with peats from the Huczwa and Stoczek deposits.

Key words: peat, raised bog, fen, polyaromatic hydrocarbons.

Słowa kluczowe: torf, torfowisko wysokie, torfowisko niskie, wielopierścieniowe węglowodory aromatyczne.

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1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a group of organic compounds widely found in the environment. These compounds can be synthesised by plants, they can be the metabolic products of microorganisms that putrefy plant and animal remains; they are generated during diagenesis and catagenesis of organic matter, particularly during petroleum formation and transformation of organic material into coal as well as in volcanic eruptions. Polyarenes are also formed naturally as a result of pyrolysis of organic material du-

ring fires of natural plant communities (Koziański, Saade 1998, Alves et al. 2010). It is believed that fires of prairies, steppes, peat lands and forests are the largest natural sources of PAHs released to the atmosphere. The amounts and types of PAHs produced during the combustion of plant material depend upon the quantity of material, the character and intensity of burning, and the plant species involved (Oros, Simoneit 2001a; Oros, Simoneit 2001b). At present, enormous amounts of unsubstituted polyarenes are discharged into the environment mainly as a result of the burning of fossil fuels in power plants, of hard coal conversion in coking

plants, the burning of coals in homes, burning of liquid fuels in vehicles, processing of petroleum in refineries as well as the extraction, transportation and storage of liquid fuels. The afore-mentioned polyarenes are also released during steel-making processes and by burning municipal waste.

Unsubstituted PAHs have been the subject of numerous medical studies and environmental surveys since it was established that some of them have toxic and carcinogenic properties. The largest threat to the quality of the natural environment is posed by high molecular weight unsubstituted PAHs, particularly those with five or more rings (e.g. benzo[a]pyrene, benzo[b]fluoranthene, benzo(ghi)perylene), because they have a harmful effect on animals and, due to their slow rate of degradation in the environment, accumulate in soils and sediments. However, lower molecular weight PAHs (naphthalenes, fluorenes, phenanthrenes) also pose a threat to the environment as they show acute toxicity towards aquatic organisms.

PAHs which are present in the atmosphere in the gaseous state or in the solid phase are to some extent taken up via sorption and accumulated by plants, mainly through leaves, as a result of both dry gaseous deposition and dust deposition as well as wet deposition (Blasco et al. 2006). It has also been found that plants can take up PAHs from the soil via the root system (Liu, Korenaga 2006). The magnitude of bioaccumulation of pollutants through leaves is dependent on leaf-air exchange processes, pollutant properties as well as the species of plant involved and exposure duration. PAHs are lipophilic organic compounds that accumulate in plant tissues. It has been found that as the lipid content of plants decreases, so does PAH concentration (Simonich, Hites 1994). An important part in sorption of PAHs by plants is played by the cuticle, which is composed of various lipid substances (Salloun et al. 2002, Chen et al. 2008.). Surveys of pine needle samples have revealed that, of unsubstituted PAHs, phenanthrene is the dominant one, accounting for 50–80% of the compounds studied (Tremolada et al., 1996). In addition, studies of PAHs present in the atmosphere have shown that phenanthrene is the dominant component of PAHs occurring in the gaseous phase (Ramirez et al. 2011).

Peats, which are considered a valuable archive, have been used in recent years to reconstruct climatic and environmental changes (Shotyk et al. 2003, Bindler 2006, De Vleeschouwer et al. 2007, Ferrat et al. 2012). Peats are organogenic rocks formed as a result of biochemical changes in remains of dead plants under the conditions of high humidity and lack of oxygen. Various types of peat differ from each other in terms

of organic and inorganic component content and these differences are a result of peats being formed under various oxygen conditions as well as various geological, hydrological, geomorphological and physical and chemical conditions, and from various plant associations (Christianis et al. 1998, Orru, Orru 2006). The occurrence of PAHs in peats formed in pre-industrial times can only be explained on the ground that they came from natural sources – i.e. inclusion, in peat, of compounds found in putrefying plant material, atmospheric deposition of compounds produced in fires of plant communities or by reference to the fact that these compounds were formed as a result of changes occurring during the biochemical phase in early sediment diagenesis. The aim of this study was to examine differentiation and variation of the spectrum of unsubstituted polycyclic aromatic compounds found in vertical profiles extracted from three peatland sites.

2. Experimental procedures

In Poland there are approx. 50 thousand peatland sites having a total surface area of ca. 12 thousand km² (Ilnicki, Żurek 1996), most of which are located in north-eastern Poland in areas away from urban and industrial centres. Fens, which are most numerous, account for 92% of Poland's peatlands (Tobolski 2000). Peatlands vary greatly in their surface area, although there are only 290 peatland sites with a surface area exceeding 500 hectares (Ilnicki, Żurek 1996). Most of the peatlands have been converted into meadows, pastures and forests.

Three locations were selected for the study: Otałżyń near Kartuzy (northern Poland), Stoczek near Radzyń Podlaski (central Poland) and Huczwa near Tomaszów Lubelski (south-eastern Poland), with one borehole drilled in each peatland site.

The Biale Błoto peatland site at Otałżyń (raised bog) (Przodkowo commune), having a surface area of ca. 54 hectares, lies in the Kaszubskie Lakeland, approx. 5 km north of Kartuzy (Fig. 1) (Ostrzyżek, Dembek 1996). It is a raised sphagnum bog which developed on land shaped by the ice sheet of the upper stadial of the Vistulian glaciation (classified as belonging to the northern Poland glaciation group). This bog is located within a moraine upland mainly composed of glacial till (Prussak, Główniak 2002). Along the southern and western boundary of the bog and partly along the northern boundary sands and silty sands overlying glacial tills are present, while along the northern and eastern boundary glacial tills are found. The bog comprises four types of peat: the upper layer of the de-



Fig. 1. Localization of investigated peatlands
Ryc. 1. Położenie badanych torfowisk

posit is made of fuscum peat (formed from peat moss *Sphagnum fuscum*), underlain by a layer of cotton grass peat. In deeper layers this cotton grass peat grades into cotton grass sphagnum peat which in turn is underlain by layers of woody peat, while the base of the deposit is composed of woody transitional peat (Fig. 2). The bog as a whole rests on silts, which were found in the borehole at a depth of 9 metres (Granacki 1958).

The Huczwa peatland site (fen) (Rachanie commune), having a surface area of ca. 87 ha, is located in south-eastern Poland approx. 14 km north-east of Tomaszów Lubelski, on the Sokal Plateau-Ridge, which has a number of areas covered by strongly calciferous loess that was formed during the upper stadial of the Vistula glaciation. This deposit lies in the valley of a small river called Sikława, a tributary of Huczwa. Stretched along the valley are beech stand forest complexes as well as farming fields and copses. The Huczwa deposit is a rush fen with an average depth of 4.92 m, and a maximum depth of 7.5 m (Zaremba 1958). The upper layers of the fen are composed of sedge peat under which there is a layer of sedge and rush peat. The next layer consists of sedge and moss peat that merges into moss peat. In the base of the fen there are deposits of algae and lime gytija (Zaremba 1958).

The Stoczek peatland site (fen) (Czemierniki commune) is located in eastern Poland, ca. 15 km south of Radzyń Podlaski, in an area along the boundary between the Lubartów Upland (the Południowopodlaska Lowland macroregion) and the Parczewska Plain (the Western Polesie macroregion). The peat bog occupies

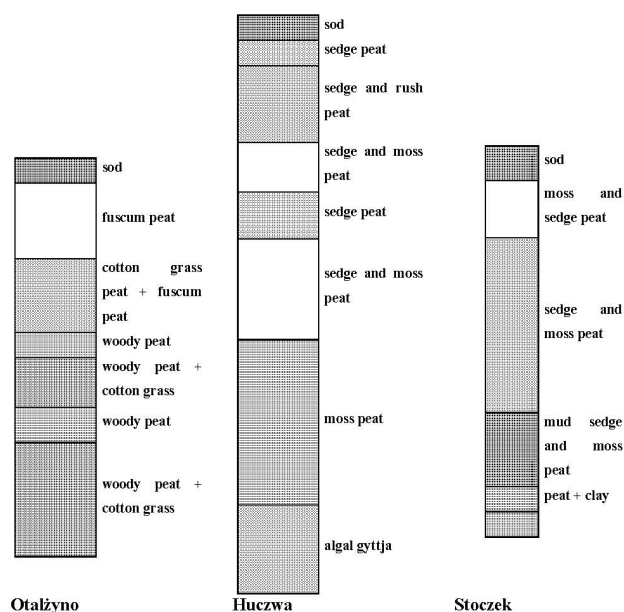


Fig. 2. Lithostratigraphic profiles
Ryc. 2. Profile litostratygaficzne

a section of the wide valley of the Tyśmienica river (a tributary of Wieprz) in its middle course, within the limits of the village of Stoczek. What mainly determined the surface features of this area was the ice sheet of the Odra glaciation (Central Poland glaciations). The moraine upland on which the Stoczek site is located is composed of glacial till layers many of which are covered by an overburden of fluvioglacial and glacial sands (Łozińska-Stępień 1986). The Stoczek deposit is composed of layers of sedge and sphagnum peat merging upwards into sphagnum and sedge peat.

The test samples were prepared by cutting cores (Otałzyno – 4.9 m and Huczwa – 8.8 m) and Stoczek (3.5 m) into 10-centimetre sections. It was decided to use the samples extracted from the greatest depth for palynology analysis with a view to determining the time the peat deposition started, whereas the other samples were analysed using a range of chemical tests.

Polycyclic aromatic hydrocarbon content was determined in dichloromethane extracts obtained from peat using SOXTEC apparatus with the liquid-solid extraction method. After the extracts had been desulfurized on copper, they were concentrated in a TurboVap workstation (using spiral nitrogen flush). Chromatographic analyses were carried out using a 6890N gas chromatograph featuring an Agilent GC-MSD 5973 mass selective detector. The procedure of separating the compounds being investigated was performed using an HP-5MS non-polar capillary column (length of 25 m, diameter of 0.20 mm, film thickness of 0.33 μm

composed of 5%-diphenyl-(95%)-dimethyl polysiloxane, using increments of 10°C/min within the temperature range 70°C – 200°C and an increment of 2.5°C/min within the 200°C – 300°C range, with the mass selective detector operating at 280°C, in the SIM mode. The analyses were carried out employing the external standard method with the use of certified standards: PM-612 mixture produced by ULTRA Scientific and pure substances of benzo[e]pyrene and perylene from Promochem. Determination of hydrocarbon content in the extracts identified 17 compounds (15 of which are included on the so-called “list” of priority PAHs that EPA recommends should be targeted for measurement in the natural environment): acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benz[a]anthracene, chrysene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene, indeno[1,2,3-cd]pyrene, dibenz[a,h]anthracene, benzo[ghi]perylene, benzo(e)pyrene and perylene. As a reference material a Marshland sample was used, in accordance with ISE 2011.2 Wageningen report.

3. Results and discussion

Peats from the **Otalzyno bog** were found to contain 209–2050 $\mu\text{g}\cdot\text{kg}^{-1}$ of PAHs, and their geometric mean content was 436 $\mu\text{g}\cdot\text{kg}^{-1}$ (Table 1). It is noteworthy that the spectrum of the PAHs is dominated by tricyclic hydrocarbons of which phenanthrene accounts for the largest share (average content – 104 $\mu\text{g}\cdot\text{kg}^{-1}$), and by fluorine present at a concentration half that of phenanthrene (average content – 53 $\mu\text{g}\cdot\text{kg}^{-1}$). Tetra- and pentacyclic hydrocarbons were found in significantly lower concentrations, except for perylene. However, high concentrations of perylene were only found in woody peat extracted from deeper layers, at a depth of 3.4–4.9 m (Fig. 3). As can be seen in the figures presenting variability of hydrocarbon content in the vertical profile, acenaphthylene, acenaphthene, fluorene are present in high concentrations in lower sections of the profile, whereas the other tricyclic or tetracyclic compounds occur at high levels in the upper section of the profile, particularly in those composed of sphagnum peat (Fig. 3). It was found that woody peats were characterized by relatively higher fluorine content and lower fluoranthene content relative to sphagnum peat. Pyrene/phenanthrene as well as anthracene/phenanthrene ratios are indicative of a very low share of compounds of pyrogenic origin (Fig. 4). It is only in peat layers at a depth of ca. 0.7 m that increased chrysene, pyrene and benzo[b]fluoranthene content is noticeable as well as relative increase in their concentration relative to phenanthrene,

which was probably caused by bog or forest fires. Peats from peatlands that have been subjected to burning are characterised by increased tetracyclic hydrocarbons content (Bojakowska, Sokołowska, 2003).

Peats from the **Huczwa** deposit were found to contain 402–1464 $\mu\text{g}\cdot\text{kg}^{-1}$ of PAHs; their average concentration was 785 $\mu\text{g}\cdot\text{kg}^{-1}$ and their geometric mean content – 754 $\mu\text{g}\cdot\text{kg}^{-1}$ (Table 1). The average levels of phenanthrene, which is the dominant among the PAHs concerned, were 276 $\mu\text{g}\cdot\text{kg}^{-1}$, while fluoranthene ranks second in terms of hydrocarbon content – 157 $\mu\text{g}\cdot\text{kg}^{-1}$. Just as in the case of the Otalzyno bog, the share of tetra- and pentacyclic compounds is significantly lower. The pyrene/phenanthrene ratio and anthracene/phenanthrene ratio imply a very low share of compounds of pyrogenic origin. However, peats deposited at a depth of ca. 3.8 m contain noticeably higher levels of most of the PAHs being investigated (Fig. 5), including those of pyrogenic origin, which indicates that fires burned there in the relevant time period in the past. Evidence for this is also provided by the relative increase, in these peat layers, of pyrene and chrysene content compared with phenanthrene (Fig. 6).

The **Stoczek** deposit was found to contain 281–1448 $\mu\text{g}\cdot\text{kg}^{-1}$ of PAHs; their average concentration was 628 $\mu\text{g}\cdot\text{kg}^{-1}$ and the geometric mean content was 567 $\mu\text{g}\cdot\text{kg}^{-1}$. Average content values for phenanthrene and fluoranthene were 153 and 115 $\mu\text{g}\cdot\text{kg}^{-1}$, respectively. What merits special mention is the increased perylene content (geometric mean – 37 $\mu\text{g}\cdot\text{kg}^{-1}$), which is characteristic of the peats deposited in the lower section of the core (Fig. 7). The peat layer lying at a depth of ca. 2.2 m is characterised by increased levels of the most of the hydrocarbons being investigated; however, no increase was found in pyrene and chrysene content relative to phenanthrene, which implies increased accumulation of PAHs. The graphs show an increase in chrysene, pyrene and benzo(b)fluoranthene content relative to phenanthrene, which suggests that they are of pyrogenic origin (Fig. 8).

Of the three peat deposits investigated, Otalzyno peats have the lowest PAH content (Otalzyno being a raised peat bog). The PAH geometric mean content in these peats – 436 $\mu\text{g}\cdot\text{kg}^{-1}$ – is significantly lower compared with Huczwa and Stoczek peats (fens), which contain 754 and 567 $\mu\text{g}\cdot\text{kg}^{-1}$ of PAHs, respectively. The PAH levels found in the afore-mentioned deposits are comparable to those found in peats from other deposits, including peat lands located in Poland and Switzerland (Berset et al. 2001, Malawska et al. 2002, Malawska et al. 2006a, Malawska et al. 2006b, Zacccone et al. 2009). Unlike PAH spectra in modern deposits, dominated



Fig. 3. Content variability of PAHs in vertical profile of Otałżyńskie torfowisko
 Ryc. 3. Zmienność zawartości WWA w profilu pionowym torfowiska Otałżyńskie

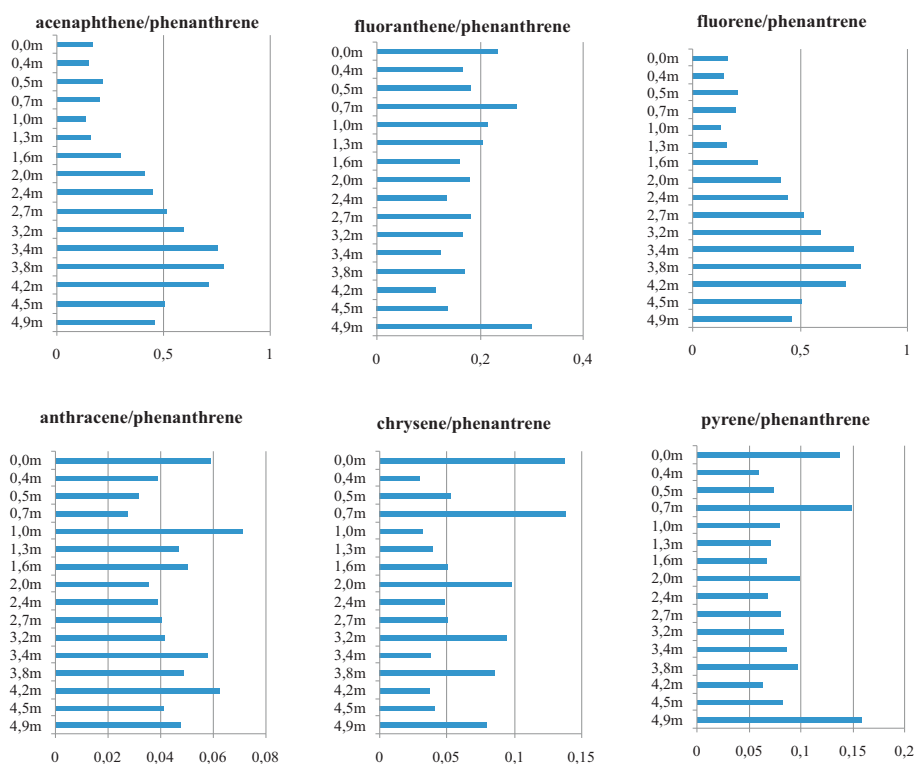


Fig. 4. Ratio variability of selected PAHs in Otałżyńskie torfowisko
 Ryc. 4. Zmienność stosunków ilościowych wybranych WWA w torfowisku Otałżyńskie

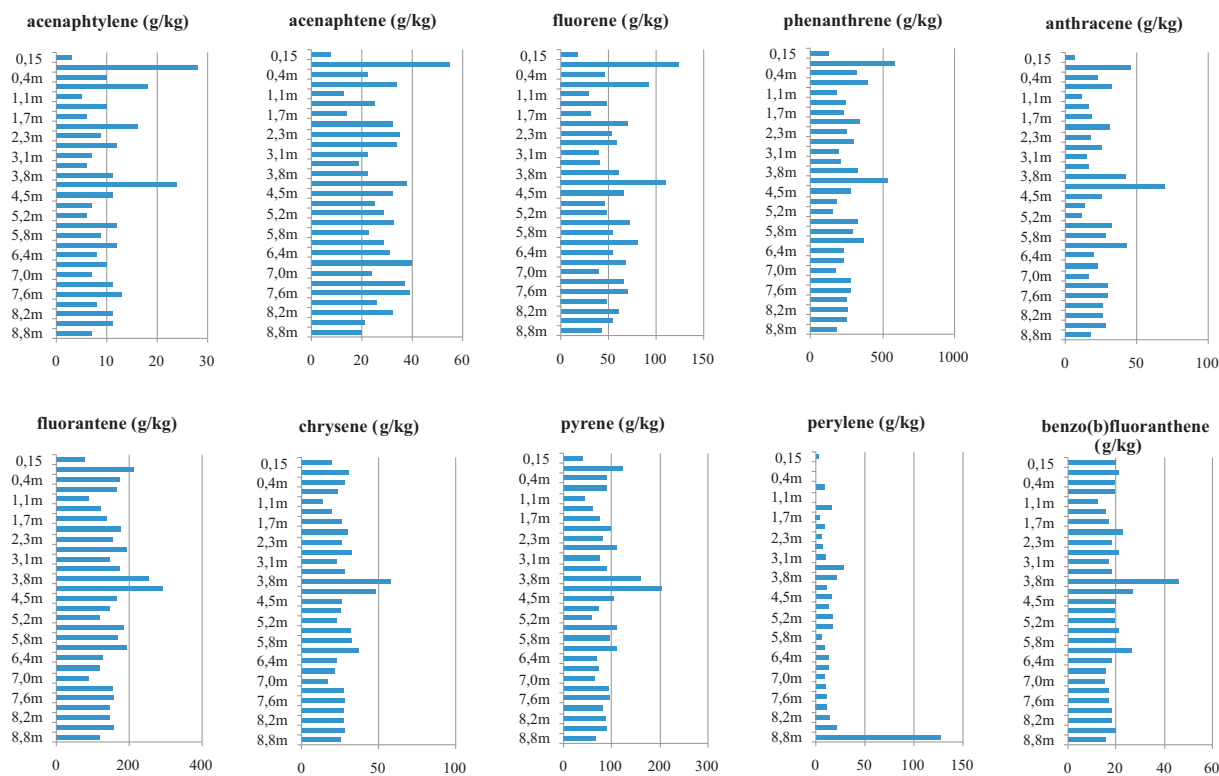


Fig 5. Content variability of PAHs in vertical profile of Huczwa peatland
 Ryc. 5. Zmienność zawartości WWA w profilu pionowym torfowiska Huczwa

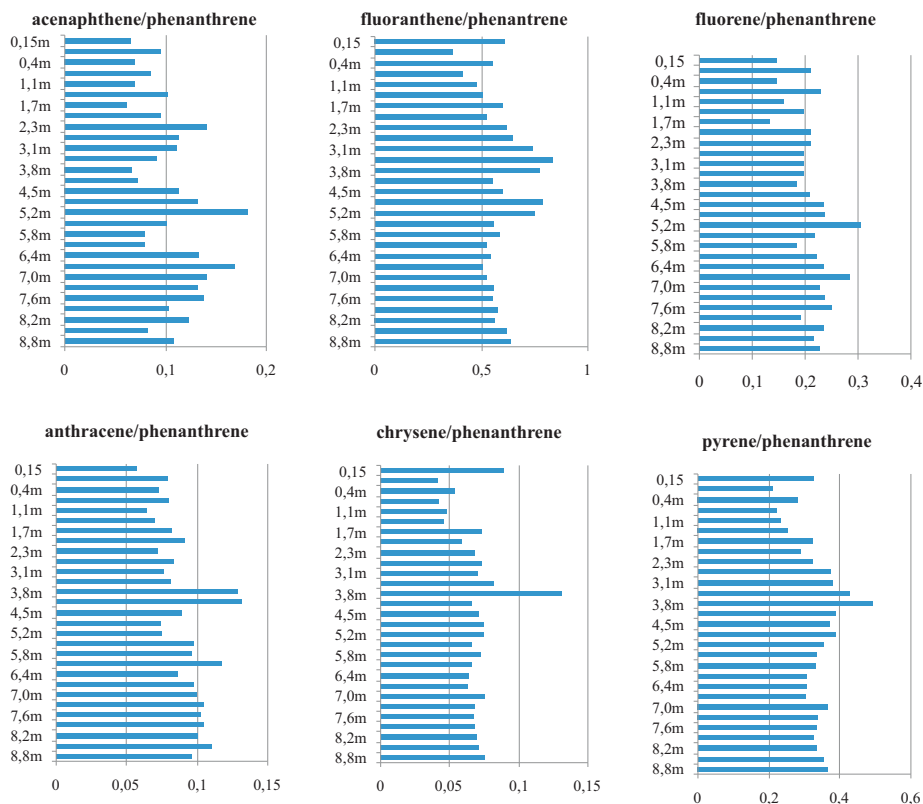


Fig. 6. Ratio variability of selected PAHs in Huczwa peatland
 Ryc. 6. Zmienność stosunków ilościowych wybranych WWA w torfowisku Huczwa

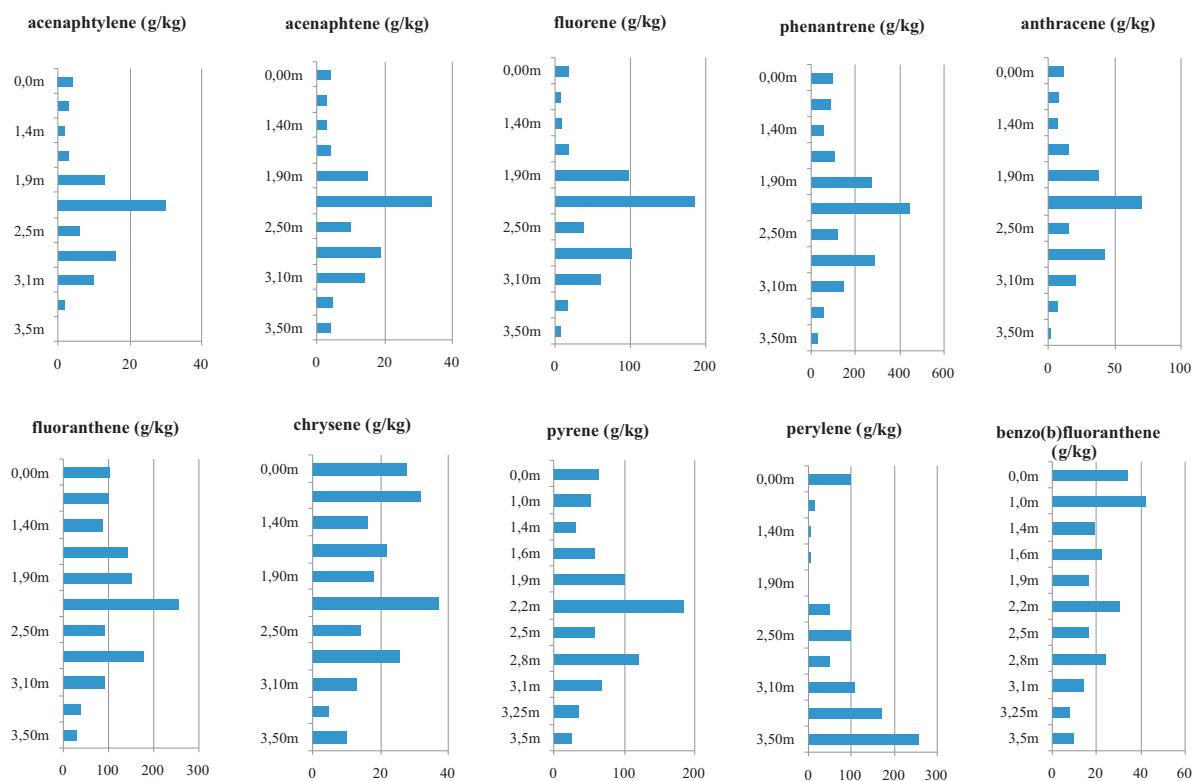


Fig. 7. Content variability of PAHs in vertical profile of Stoczek peatland
 Ryc. 7. Zmienność zawartości WWA w profilu pionowym torfowiska Stoczek

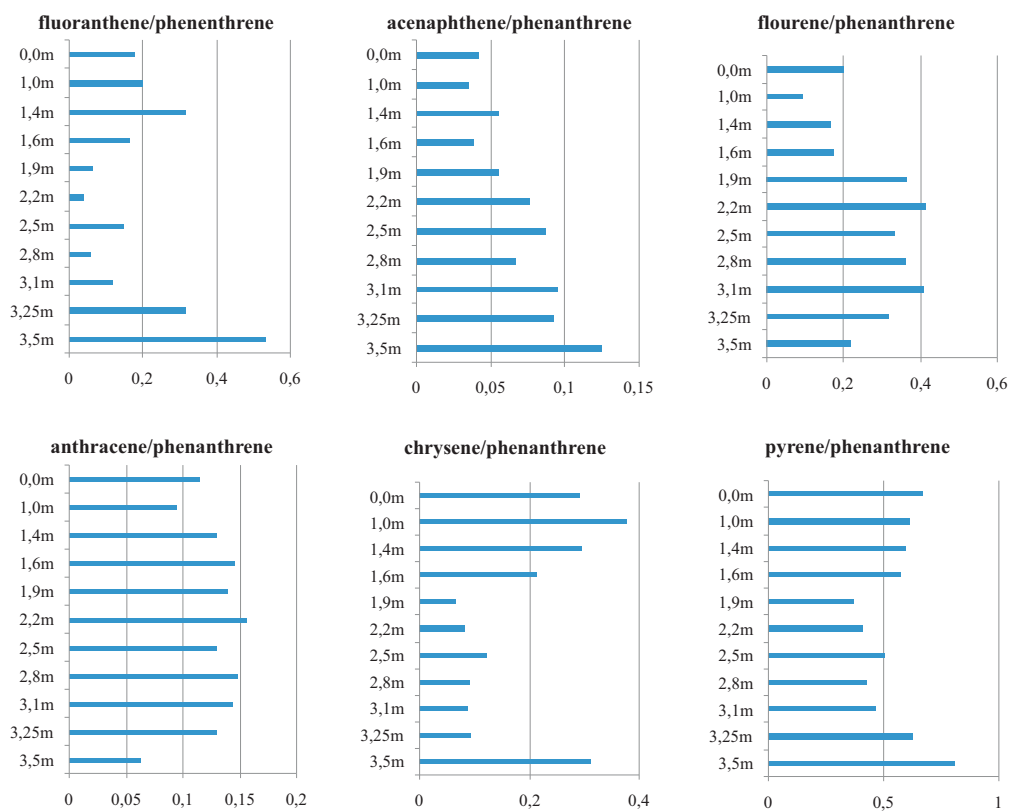


Fig. 8. Ratio variability of selected PAHs in Stoczek peatland
 Ryc. 8. Zmienność stosunków ilościowych wybranych WWA w torfowisku Stoczek

Tab. 1. Statistical parameters of PAHs in peatlands Otałżyno, Huczwa and Stoczek

Tab. 1. Parametry statystyczne WWA w torfowiskach Otałżyno, Huczwa i Stoczek

Parameter Parametr	Otałżyno				Huczwa				Stoczek			
	M	GM	Min	Max	M	GM	Min	Max	M	GM	Min	Max
	$\mu\text{g}\cdot\text{kg}^{-1}$											
acenaphtylene	5	5	3	13	11	10	3	28	8	5	1	30
acenaphthene	39	35	15	78	28	26	8	55	10	7	3	34
fluorene	53	49	27	99	58	54	18	123	51	29	7	185
phenanthrene	104	101	63	181	276	261	123	582	153	115	32	447
anthracene	5	5	3	9	26	23	7	70	21	14	2	70
fluoranthene	20	18	9	49	157	151	75	293	115	99	31	255
pyrene	10	9	5	27	90	85	40	204	73	62	26	184
benzo(a)anthracene	3	3	1	10	18	17	9	43	16	14	5	33
chrysene	7	6	3	25	28	27	14	58	20	18	5	37
benzo(b)fluoranthene	8	7	4	23	20	19	12	46	22	19	8	42
benzo(k)fluoranthene	3	2	2	12	8	8	5	21	8	7	3	18
benzol(e)pyrene	10	9	6	24	14	14	8	36	17	15	6	34
benzo(a)pyrene	3	2	2	10	9	8	5	27	7	6	2	16
perylene	241	7	2	1568	15	10	2	127	79	37	4	259
indeno(1,2,3-cd)pyrene	6	4	3	16	9	9	6	22	10	9	3	27
dibenzo(ah)anthracene	3	3	3	7	3	3	3	3	3	3	3	5
benzo(ghi)perylene	51	36	8	182	15	14	7	26	13	12	7	27
Sum of PAH	570	436	209	2050	785	754	402	1464	628	567	281	1448
Acenaphtene/phenanthrene	0.41	0.34	0.13	0.78	0.11	0.10	0.06	0.18	0.07	0.06	0.04	0.13
fluoranthene/phenanthrene	0.18	0.18	0.11	0.30	0.59	0.58	0.36	0.83	0.92	0.86	0.55	1.65
fluorene/phenanthrene	0.55	0.49	0.25	1.24	0.21	0.21	0.13	0.31	0.28	0.25	0.09	0.41
anthracene/phenanthrene	0.05	0.04	0.03	0.07	0.09	0.09	0.06	0.13	0.13	0.12	0.06	0.16
chrysene/phenanthrene	0.07	0.06	0.03	0.14	0.11	0.10	0.05	0.18	0.18	0.15	0.07	0.38
pyrene/phenanthrene	0.09	0.09	0.06	0.16	0.33	0.33	0.21	0.49	0.55	0.54	0.37	0.81

M – mean, GM – geometric mean

by tetra- and pentacyclic compounds, particularly by tetracyclic fluoranthene, the PAH spectrum in the peats investigated is dominated by tricyclic compounds (Bojakowska et al. 2012). Of the PAHs found in the peats investigated, tricyclic phenanthrene is the domi-

nant one; it is present in its highest concentrations in the atmosphere as a gaseous phase. The studies carried out have shown that peats extracted from the Otałżyno, Huczwa and Stoczek sites are characterised by low pyrogenic PAH content (4-, 5- and 6-ring compounds,

except for perylene). What is especially interesting about fen peats extracted from the Otałżyno site is the higher acenaphthene content compared with bog peats and relatively scant amounts of fluoranthene relative to phenanthrene if compared with peats from the Huczwa and Stoczek deposits. Low fluorethene content relative to phenanthrene is especially noticeable in woody peats.

4. Conclusions

1. Peats from the Otałżyno deposit (a raised bog) are characterised by lower PAH content – 436 $\mu\text{g. kg}^{-1}$ compared with peats from the Huczwa and Stoczek (fens), where PAH content is 754 and 567 $\mu\text{g. kg}^{-1}$, respectively.
2. The PAH spectrum of the peats studied is dominated by tricyclic compounds, primarily by phenanthrene, in contrast with the PAH spectra of modern deposits in which tetra- and pentacyclic compounds account for the majority of polycyclic aromatic hydrocarbons.
3. Especially worth mentioning is the fact that bog peats have higher acenaphthene content, but at the same time have relatively low levels of fluoranthene relative to phenanthrene, if compared with peats from the Huczwa and Stoczek deposits.
4. Some of the peat layers have been found to contain increased levels of pyrogenic hydrocarbons relative to phenanthrene.
5. The variations in PAHs profile has been caused by the age and type of peat deposits.

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ZMIENNOŚĆ WWA W PROFILU PIONOWYM W TORFOWISKACH OTALŻYNO, HUCZWA I STOCZEK

Streszczenie

Celem niniejszej pracy było zbadanie zmienności niepodstawionych wielopierścieniowych węglowodorów aromatycznych (WWA) w pionowych profilach pobranych z torfowisk. Do badań zostały wybrane trzy złoża torfowe: Otałżyno obok Kartuz (północna Polska), Stoczek koło Radzyna Podlaskiego (centralna Polska) oraz Huczwa koło Tomaszowa Lubelskiego (południowo-wschodnia Polska). Zawartość wielopierścieniowych węglowodorów aromatycznych była oznaczana chromatograficznie. W trakcie oznaczeń identyfikowano 17 związków (15 spośród nich występuje na liście priorytetowych węglowodorów, które EPA rekomenduje do oznaczania w środowisku naturalnym). Torfy z torfowiska Otałżyno (torfowisko wysokie) charakteryzowały się mniejszą zawartością WWA ($436 \mu\text{g}\cdot\text{kg}^{-1}$) w porównaniu z torfami z torfowisk Huczwa i Stoczek (torfowiska niskie), w których poziom ten wynosił odpowiednio $754 \mu\text{g}\cdot\text{kg}^{-1}$ i $567 \mu\text{g}\cdot\text{kg}^{-1}$. Profil WWA badanych torfowisk jest zdominowany przez związki tricykliczne, przede wszystkim przez fenantren, w odróżnieniu od spektrum WWA we współczesnych osadach, w których dominują związki cztero- i pięciopierścieniowe. Zwraca uwagę wyższa zawartość acenaftenu w torfach wysokich przy jednoczesnym względnym ubóstwie w fluoranten względem fenantrenu w porównaniu do torfów ze złóż Huczwa i Stoczek.