

MARIA KARŁOWSKA-JASEK, EWA RZEWUSKA*

ON THE OCCURRENCE OF CARCINOGENIC POLYCYCLIC AROMATIC COMPOUNDS (PAH) IN SOME POLISH RIVERS

Polycyclic aromatic hydrocarbons PAH are exhibiting, carcinogenic effects of varying intensity according to the number of rings. Benzo (*a*)pyrene was found to be the most active species. The PAH compounds such as 3,4-benzo (*b*)fluoranthene and 10, 11-benzo (*j*) fluoranthene were found to be average in activity. Other compounds were found either weakly carcinogenic or not exhibiting any activity. The classification has been conducted experimentally and based on the degree of influence of the substance on the skin of mouse.

1. INTRODUCTION

Polycyclic aromatic hydrocarbons (PAH) are colourless water insoluble substances. Some of them are potent carcinogens. The representatives of mono and of polycyclic aromatic hydrocarbons listed in table 1.

Table 1

Mono- and polycyclic aromatic hydrocarbons

Benzene	naphthalene	di-phenyl
anthracene	phenanthrene	fluorene
benzo(<i>a</i>)anthracene	chrysene	pyrene
benzo(<i>a</i>)pyrene	benzo(<i>e</i>)pyrene	perylene
anthracene	benzo(<i>ghi</i>)perylene	benzo(<i>k</i>)fluoranthene
	koronene	

In studies on neoplastic diseases carcinogenic activity of various PAH was tested on mouse skin. It has been stated that benzo (*a*) pyrene (B(*a*)P) is the most active, benzo (*b*) fluoranthene (B(*b*)F) and benzo (*j*) fluoranthene are moderately active, and benzo (*a*) anthracene (B(*a*)A), benzo(*e*)pyrene (B(*e*)P), chrysene (Ch) and indeno (1, 2, 3-*cd*)

* Water Pollution Control Department of the Wrocław Division of the Research Institute for Environmental Development, Wrocław, Poland.

pyrene (IP) are the weakest carcinogens. An increasing tendency of carcinogenic activity has been observed with the increasing number of carbon rings until 5 rings are reached, beyond this number this activity decreases. The world-wide studies on polycyclic aromatic hydrocarbons have allowed to find some features common to all the carcinogenic PAH, namely a distinctly local action on human and animal organisms, and the long time from the contact of tissue with the carcinogenic PAH required for neoplastic symptoms to be manifested. The disease itself is explained by the ability of the hydrocarbon molecule to combine with cell acceptor of the living organism.

It is well known that PAH are formed during the high temperature pyrolysis of hydrocarbons (673–773 K), but more recently strong evidence has indicated that PAH may be produced by bacteria and plants (KNORR, SCHENK, HANCOCK et al. [19]. In soil samples from forests and fields BLUMER [2] found the concentrations of PAH of between 40 and 1300 $\mu\text{g kg}^{-1}$. The areas examined were distant from major highways and industry. Sampling was performed under dry weather conditions. According to BORNEFF and KUNTE [7] the average level of carcinogenic PAH in ground water was 0.06 $\mu\text{g dm}^{-3}$. These authors as well as FALK et al. [14] concluded that bituminous road surfaces and car tyre wear both contribute PAH to run-off from roadways. The latter in turn, may contribute substantially to levels of PAH in sewage. Samoilovich and Redkin have found appreciable levels of B (a) P in rivers polluted by oilfield and petroleum industry effluents. These scientists found B (a) P concentrations of 0.05–3.5 $\mu\text{g dm}^{-3}$ 3–4 km downstream from the point of entry of the effluent, and 0.07–1.06 $\mu\text{g dm}^{-3}$ 22 km downstream.

BORNEFF and FISHER [5] have stated that the dried plankton from the Lake Constanca contained about 100 $\mu\text{g kg}^{-1}$ of PAH.

GRAPH and DIEHL [17] who analysed the relation between PAH and plants have stated that in vegetables used for human consumption the levels of B (a) P amount to 10–20 $\mu\text{g kg}^{-1}$ dry wt, and in other plants concentrations of individual PAH ranged within 5–110 $\mu\text{g kg}^{-1}$ of dry weight. Their later experiments have shown that the rate of growth of a number of plants and the output of grain (rye) can be increased by feeding carcinogenic PAH. In works of Russian scientists it has been emphasized that the plants which absorb CO_2 , expell oxygen and retain mechanically some impurities on their leaves, can assimilate various compounds (aromatic compounds included) binding them and neutralizing. Some of them, e. g. grape-vine, peach-tree etc. do not absorb aromatic hydrocarbons almost at all, privet assimilates some tenths of miligram, poplar, cherry-tree and others intake miligrams, while maple-trees and others absorb some tens of miligrams. The investigations conducted on behalf of the World Health Organization allowed to state the presence of PAH in drinking water. It has been assumed than even trace amounts of these compounds — if present in water for human consumption — require further studies. According to standards accepted by W. H. O. the allowable amounts of carcinogenic PAH (benzo (a) pyrene, fluoranthene, benzo (k) fluoranthene, benzo (b) fluoranthene, benzo (ghi) perylene and indeno (1, 2, 3, c–d) pyrene) must not exceed 0.200 $\mu\text{g dm}^{-3}$ [12]. This 0.200 $\mu\text{g dm}^{-3}$ standard is the reference level for the concentrations found in various rivers. Some selected examples are given in table 2. This table presents also some examples of concentra-

tions of PAH stated in other sources. Some examples of PAH removal by water and sewage treatment processes are given in table 3.

The investigations conducted have confirmed the importance of the problem studied bearing in mind that man is exposed to a permanent contact with carcinogenic substances occurring in his natural environment, i. e. in soil and plants, water and air.

Table 2

Carcinogenic benzo(a)pyrene levels

Medium monitored	Concentration $\mu\text{g dm}^{-3}$ $\mu\text{g dm}^{-3}$	Literature item
River Gersprenz at Munster	0.0096	[8, 9]
River Danube at Ulm	0.0006–0.0013	[8, 9]
River Main at Selingenstadt	0.0011–0.043	[8, 9]
River Aach at Stockach	0.004–0.016	[8, 9]
River Schussen	0.001	[8, 9]
Thames River, Kew Bridge	0.130	[1]
Thames River, Albert Bridge	0.160	[1]
Thames River, Tower Bridge	0.350	[1]
Rivers in Soviet Union	0.05–3.5	[26]
Ground water	0.045–0.06	[7]
Wastewater		
domestic effluents during dry weather	0.038–0.368	[9]
domestic effluents during heavy rain	1.840	[9]
factory effluents	0.1–0.368	[9]
Marine sediments		
Greenland	5	[22]
Italia	1.4–3000	[12]
France	15–1500	[11]
Forest soil	40–1300	[2]
Plankton		
Greenland	5	[22]
Italy	5–21	[12]
France	400	[23]
Algae		
Greenland	60	[22]
Italy	2	[12]
Fish		
Greenland	15–16	[22]
Italy	65	[12]
Plants		
Edible	0–20	[17]
Other	1110	[17]

Table 3

PAH removal in some water and wastewater treatment processes

Treatment method	% Reduction	Kind of PAH	Literature
Primary treatment	20-80	total	[10]
Primary and biological	75	B(a)P	[15]
Soil not inoculated with bacteria (control)	0	B(a)P	[24]
Soil + N5 bacterial strain	53	B(a)P	[24]
Soil + N13 bacterial strain	66	B(a)P	[24]
Soil + N13 bacterial strain*	82	B(a)P	[24]
Extraction with butyl acetate	90	B(a)P	[27]
Percolating filters and activated sludge	97-99	total	[10]
Synthetic sewage dosed with B(a)P, aluminium sulphate used as a coagulant with addition of synthetic flocculation agents	50	B(a)P	[21]
	90	B(a)P	[21]
Addition of flocculating agent (5 mg/dm ³)	96	B(a)P	[21]
Carbon adsorption bed	99	total	[6]
Water ozonization after 30 min. contact time	99	B(a)P	[7]
Water ozonization after 1 min contact time	39	B(a)P	[7]
Chlorination of drinking water at 0.3-0.5 g/m ³ of chlorine and level of B(a)P 1-5 µg/dm ³ ; contact time 0.5-2 hrs	50-100	B(a)P	[18]

* Before the experiment this strain was cultured in a medium containing B(a)P for 110 days.

2. RESULTS OF INVESTIGATIONS

The water quality surveys carried at our Institute in the years 1975 and 1976 encompassed PAH determinations in 10 monitoring cross-sections along the Oława and Nysa Kłodzka Rivers and their influents as well as in the Odra River.

The ranges of concentrations for the selected PAH determined in the controlled rivers are given in table 4. The values of carcinogenic B(a)P concentrations determined in the water of the rivers controlled are of the same order as in European rivers (see table 2).

Table 4

PAH concentrations found in three rivers

Source	Range of concentrations in µg dm ⁻³		
	B(a)P	Fl	B(ghi)P
Nysa Kłodzka	0.0031-0.4826	0.0222-2.0148	0.0161-0.4821
Oława	0.0026-3022	0.0311-2.4024	0.0045-0.4827
Odra	0.018-0.8234	0.1911-1.420	0.0284-0.4727

Maximum levels of B (a) P, stated in water of the rivers Nysa Kłodzka and Odra reached or slightly exceeded the value found in the Thames River at the Tower Bridge ($0.350 \mu\text{g}/\text{dm}^3$). It should be emphasized that the highest concentrations of B (a) P were found for cross-sections loaded with sewage of the origin other than petrochemical and coke industries, which are generally considered as the main source of benzopyrenes. Direct or indirect influence of small gas works through the ground water cannot, however, be excluded. Typical branches of the industry — which according to literature data chiefly contribute to levels of PAH in sewage — are not localized within the area of Oława and Nysa Kłodzka drainage basins, except for small gas works.

The highest concentrations of PAH have been found in the monitoring cross-section of the Oława River, below the town of Ziębica, where the water was polluted considerably with insufficiently treated domestic sewage and factory effluents from fruit and vegetable processing plants, sugar plant, slaughter houses etc. Physico-chemical parameters of this water far exceeded the upper standard limits, e. g. the average BOD_5 value was $157 \text{ mgO}_2 \text{ dm}^{-3}$ and the average concentration of N_{NH_4} amounted to 12.1 mgdm^{-3} (tables 4 and 5). The same has been found in the river basin of Nysa Kłodzka, where the maximal concentrations of benzopyrenes were found in the Cielnica River, whose water was characterized by parameters corresponding to those of sewage, with the average BOD value $59.1 \text{ mgO}_2 \text{ dm}^{-3}$ and the average concentration of N_{NH_4} amounting to 7.4 mgdm^{-3} .

In the Nysa Kłodzka waters elevated levels of PAH have been found in the cross-sections distinctly influenced by domestic sewage, effluents from food industry and various workshops, such as laundries, chemical cleaning etc. High concentrations of PAH were found in the Byczeń cross-section, where the average value of BOD_5 amounted to $5.9 \text{ mgO}_2 \text{ dm}^{-3}$ and concentration of suspended solids equaled 42 mgdm^{-3} . The levels of PAH are compared in figs. 1–3.

The highest concentrations of carcinogenic B (a) P were observed in cross-sections loaded with large amounts of sewage, i. e. in the Oława River, below the town of Ziemlice, in the Cielnica River (influent of the Nysa Kłodzka River) and in the Nysa Kłodzka River at Byczeń.

In the remaining cross-sections the ranges of B (a) P for both the rivers, were similar, varying from 0.002 to $0.060 \mu\text{g dm}^{-3}$. The concentrations of PAH in the Odra River were higher than in the rivers just discussed, ranging from 0.040 to $0.250 \mu\text{g dm}^{-3}$, and in single cases they amounted to 0.400 and even exceeded $0.800 \mu\text{g dm}^{-3}$.

In the rivers Oława and Nysa Kłodzka not only the ranges of B (a) P concentrations were similar, but even the cycles stated in separate seasons of the year were analogous. The highest concentrations were observed in summer, they decreased in spring and autumn, and were the lowest in winter.

In the Odra River the contents of PAH were not determined in spring. The highest concentrations occurred in summer, lower in autumn, and the lowest were noted in winter.

In the Cielnica River charged with effluents from malt industry, characterized by periodic operation, the highest levels occurred in autumn.

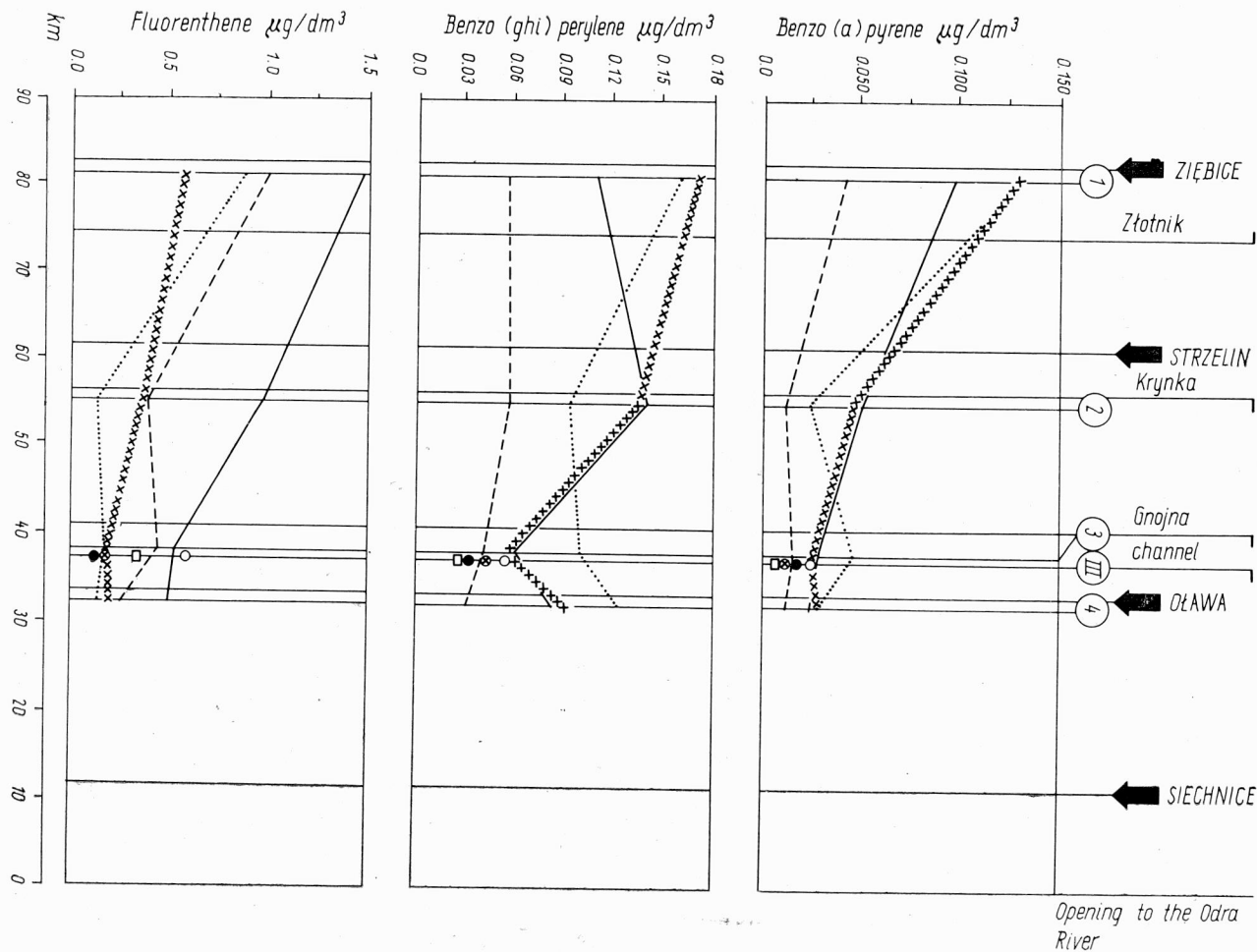


Fig. 1. Levels of average PAH concentrations stated in the Oława River

-- ○ spring, ○ summer, × × × × ○ autumn, - - - □ winter

Rys. 1. Poziom średnich stężeń związków WWA w wodach Oławy

-- ○ wiosna, ○ lato, × × × × ○ jesień, - - - □ zima

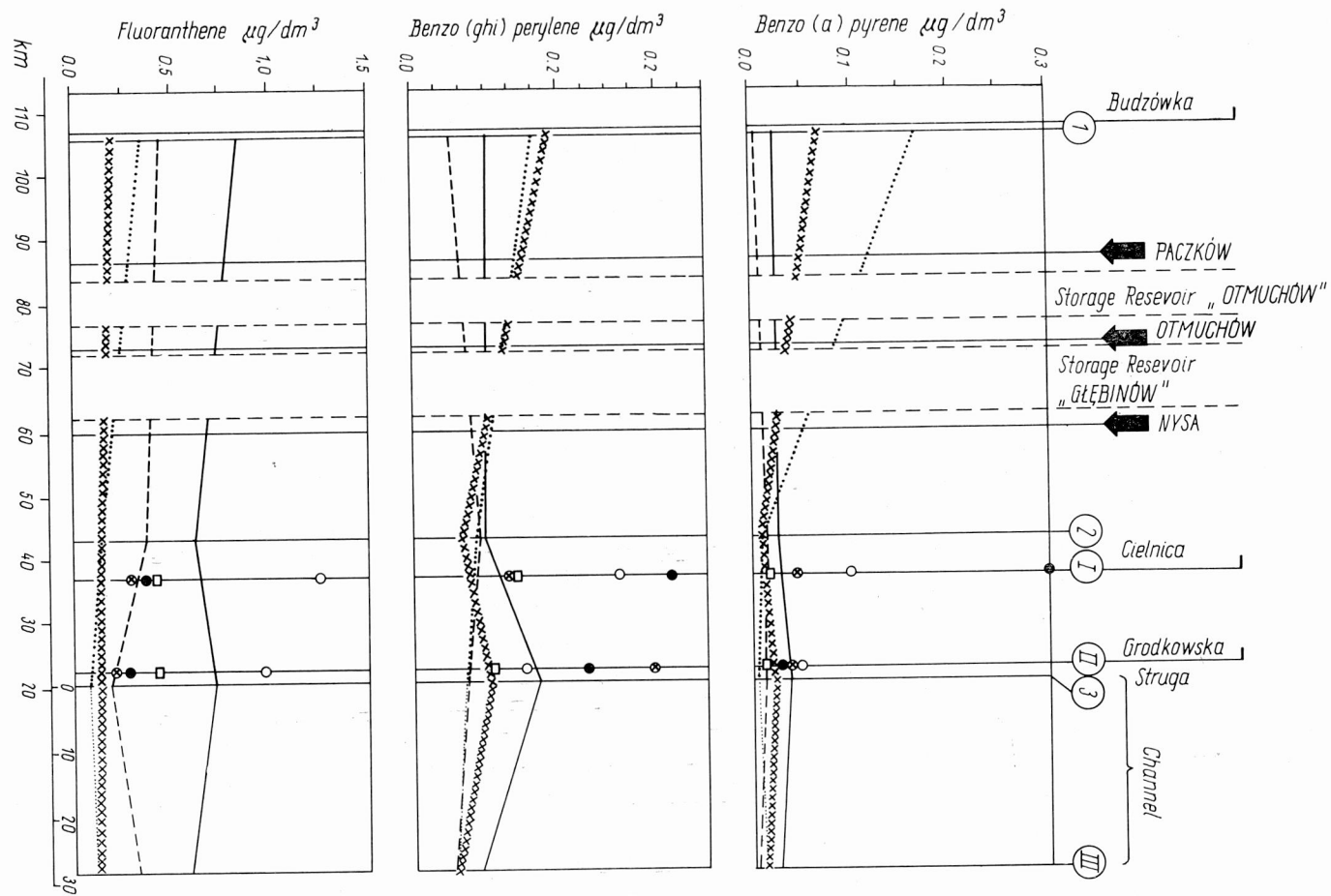


Fig. 2. Levels of average PAH concentrations stated in Nysa Kłodzka River

--- ○ spring, ○ summer, × × × × ○ autumn, - - - - □ winter

Rys. 2. Poziom średnich stężeń związków WWA w wodach Nysy Kłodzkiej

--- ○ wiosna, ○ lato, × × × × ○ jesień, - - - - □ zima

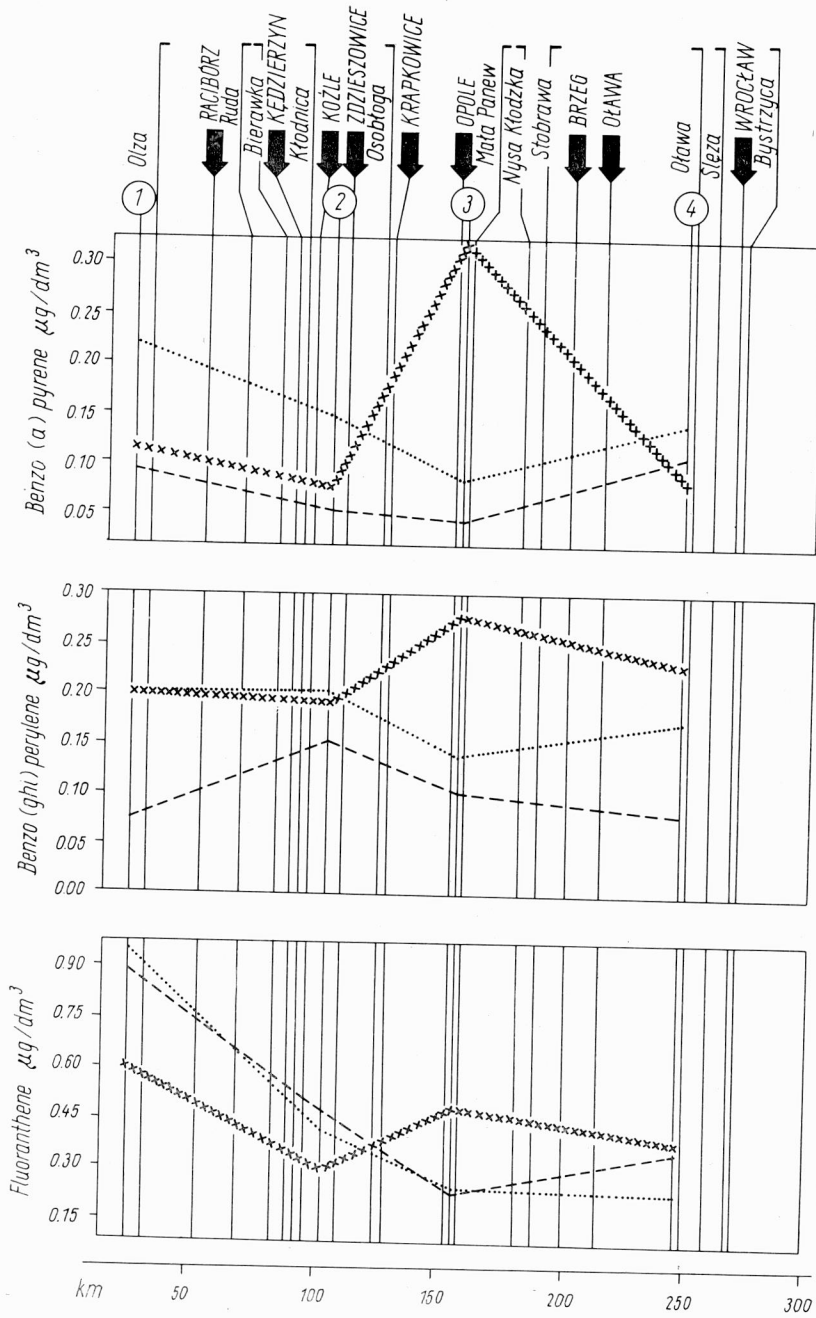


Fig. 3. Levels of average PAH concentrations stated in Odra River

..... summer, x x x autumn, - - - winter
 Rys. 3. Poziom średnich stężeń związków WWA w wodach Odry
 lato, x x x jesień, - - - zima

The investigations on fluoranthene concentrations showed similar seasonal variations of concentrations in river waters, i. e. the highest concentrations were found in spring, lower to winter and the lowest ones in summer and autumn. The highest concentrations: 2.000 and 2.400 $\mu\text{g dm}^{-3}$, were found in two cross-sections — in the Oława River and in the Cielnica River. In Odra River seasonal changes could be distinguished, but the concentrations varied within a narrower range than in the two remaining rivers. It must be noted, however, that the lower limit was higher, and highest concentration amounted to 1.400 $\mu\text{g dm}^{-3}$.

For benzo (ghi) perylene similar seasonal regularities have been found in the rivers Oława and Nysa Kłodzka, and a different cycle in the Odra River. In the first two rivers the concentrations of B(ghi)P varied from 0.022 to 0.225 $\mu\text{g dm}^{-3}$. In the Odra River the values of B(ghi)P concentrations ranged within 0.050–0.500 $\mu\text{g dm}^{-3}$. The lowest level was noted in winter, and the highest in summer.

Results of chemical and physical examinations of the remaining indices of water pollution (presented in table 5) reveal no direct correlation between the values of the parameters measured and the concentrations of PAH, although in single cases the concentrations of PAH increased considerably with increasing amounts of suspended solid and BOD_5 values. It can, however, be stated that in general the level of PAH in polluted water is higher than in unpolluted waters.

SUMMARY AND CONCLUSIONS

The carcinogenic PAH, i. e. benzo (*a*) pyrene, benzo (*b*) fluoranthene, 10,11 benzo (*j*) fluoranthene, benzo(*a*) anthracene, benzo (*e*) pyrene may be formed during pyrolysis, they are however also produced by bacteria and plants. Petrochemical, coke and oil industries as well roadways are the main sources contributing to the pollution of environment with polycyclic aromatic hydrocarbons. Since these compounds occur in natural environment of man (soil, plants, and water), human organism is permanently exposed to the contact with carcinogenic substances.

In water and sewage treatment processes the removal of benzopyrenes depends on the method applied. Thus primary treatment yields 20–80% reduction of PAH, biological treatment yields 75% PAH removal, while activated sludge followed by coagulation may yield even 97–99%.

The concentrations of PAH determined in the rivers Oława, Nysa Kłodzka and Odra are of the same order as in other European rivers. Maximum concentration of benzo (*a*) pyrene in the Oława River was about 0.350 $\mu\text{g/dm}^{-3}$, thus much higher than the standard value 0.200 $\mu\text{g dm}^{-3}$, and close to that stated in the Thames River at the Tower Bridge. In the remaining rivers maximum concentration of this compound ranged from 0.150 to 0.84 $\mu\text{g dm}^{-3}$.

Maximum concentrations were found for cross-sections receiving large amounts of wastewaters (the rivers Cielnica, Odra and Oława below the town of Ziębice) which did

not originate from the industries recognized as the main sources of PAH, but from the food industry and from inhabitants.

The pollution characterized by physical and chemical parameters is similar for Oława and Nysa Kłodzka Rivers; the seasonal cycles of PAH concentrations are also analogous. For the Odra River — much more polluted than the two remaining rivers — the levels of PAH concentrations were also higher.

It may be concluded that the high concentrations of PAH stated in Polish and European rivers indicate the necessity of further studies to protect man against the contact with carcinogenic substances.

WYSTĘPOWANIE WĘGLOWODORÓW AROMATYCZNYCH W WODACH POWIERZCHNIOWYCH OŁAWY, NYSY KŁODZKIEJ I ODRY

Wielopierścieniowe węglowodory aromatyczne są związkami wykazującymi, w zależności od liczby pierścieni, zróżnicowane pod względem siły działania własności rakotwórcze.

Za najaktywniejszy pod tym względem uznany został benzo (*a*) piren. Do średnio aktywnych związków zaliczono 3,4-benzo (*b*) fluoranten i 10, 11 benzo (*f*) fluoranten. Pozostałe związki uznano za słabo aktywne lub nie wykazujące tendencji rakotwórczych.

Powyższy podział przeprowadzono doświadczalnie, badając oddziaływanie poszczególnych związków na skórę myszy. Badania nad WWA prowadzi wiele ośrodków naukowych w świecie. W Polsce zagadnienia te znajdują się na etapie prac wstępnych.

WWA powstają w wyniku naturalnych procesów, do których można między innymi zaliczyć procesy fizjologiczne niektórych bakterii i roślin, oraz w wyniku gospodarczej działalności człowieka. WWA powstaje między innymi podczas ścierania opon samochodowych na powierzchniach asfaltowych, a także jest wydalany do atmosfery wraz ze spalinami z wylotów kominowych. Związek ten przedostaje się do wód powierzchniowych ze ściekami, np. z przemysłu koksowniczego, petrochemicznego itp. Badania wykazały, że w procesach oczyszczania ścieków WWA można zredukować od 20% przez oczyszczanie mechaniczne do 96% w drodze pełnego oczyszczania. Przy wysokim uzdatnieniu wody przeznaczonej dla celów komunalnych osiągnano prawie 100% redukcji WWA.

W pracach nad oceną jakości wody rzek Oławy, Nysy Kłodzkiej i Odry prowadzonych w latach 1976 i 1977 przez Instytut Kształtowania Środowiska kontrolowano trzy z WWA, stwierdzając następujące zakresy stężeń:

benzo (<i>a</i>) piren	od 0,0031 do 0,8234 gdm ⁻³ ,
fluoranten	od 0,0222 do 2,4024 gdm ⁻³ ,
benzo (<i>ghi</i>) perylen	od 0,0045 do 0,4827 gdm ⁻³ .

Maksymalne stężenia tych substancji stwierdzono w przekrojach znacznie obciążonych tak ściekami komunalnymi jak i pochodzącymi z zakładów przemysłu spożywczego oraz z zakładów usługowych. Wody Odry mające znacznie wyższy poziom zanieczyszczenia związkami organicznymi od dwu pozostałych rzek cechowały również wyższe stężenia WWA.

W wodach Oławy i Nysy Kłodzkiej o podobnym stopniu zanieczyszczenia stwierdzono podobny poziom kontrolowanych WWA. Odnotowano prawidłowości w układach stężeń odpowiadające porom roku — najwyższe wiosną, najniższe latem i jesienią. W wodach Odry zakres stężeń był wyrównany bez względu na porę roku.

Przedstawione wyniki badań stanowią wstępną informację o stopniu zagrożenia wód powierzchniowych w kraju przez WWA. Problem ten wymaga dalszych badań.

DAS AUFTRETEN VON MEHRKERNAROMATEN IM FLUSSWASSER DER OŁAWA, NYSA KŁODZKA UND ODRĄ

Polycyclische aromatische Kohlenwasserstoffe (Mehrkernaromate) weisen je nach Ringzahl eine starke oder schwache cancerogene Aktivität auf. Nach bekannten Literaturangaben, schreibt man die stärkste Wirkung dem 3, 4-Benzpyren, mässige dem 3, 4-Benz-fluoranthen, schwache dem 11, 12-Benz-fluoranthen zu. Mehrkernaromate treten überall auf — in der Luft, im Oberflächengewässer, im Boden und sogar in Nahrungsmitteln; somit kommt den Konzentrationsuntersuchungen größte Bedeutung zu.

Die Verfasser haben das Auftreten der Polycyclischen Aromate im Wasser einiger niederschlesischen Flüsse in den Jahren 1976/77 untersucht und folgende Konzentrationen festgestellt:

3, 4-Benzpyren	0,0031–0,8234 $\mu\text{g}/\text{dm}^3$,
Fluoranthen	0,0222–2,4024 $\mu\text{g}/\text{dm}^3$,
1, 12-Benzperylen	0,0045–0,4827 $\mu\text{g}/\text{dm}^3$.

Grösste Werte hat man — wie erwartet — in diesen Flußquerschnitten gefunden, wo die Belastung mit Abwasser am stärksten war. Das Flußwasser der Odra (Oder) wies höhere Konzentrationen auf als das der beiden anderen Flüsse. Konzentrationsschwankungen während der Jahreszeiten waren unwesentlich. In Nysa Kłodzka Glatzer (Neiße) und Oława (Ohlau) waren die Konzentrationen weit kleiner und jahreszeitabhängig — höchste Werte stellte man im Frühjahr, niedrigste im Sommer und im Herbst fest.

Die Untersuchungen werden fortgesetzt.

НАЛИЧИЕ АРОМАТИЧЕСКИХ МНОГОЯДЕРНЫХ УГЛЕВОДОРОДОВ В ПОВЕРХНОСТНЫХ ВОДАХ НЫСЫ КЛОДЗКСКОЙ, ОЛАВЫ И ОДРЫ

Многоядерные углеводороды ароматического ряда являются соединениями, обнаруживающими, в зависимости от количества циклов, различные по силе действия канцерогенные свойства. Самым активным в этом отношении был признан бенз(a)пирен. К среднеактивным соединениям относят 3,4-бенз(b)флюорантен. Остальные соединения признаны слабоактивными или же не обнаруживающими канцерогенных способностей. Эта классификация была произведена опытным путем, с изучением степени воздействия названных соединений на кожу мышей.

Ввиду наличия ароматических многоядерных углеводородов (AMУ) в основных компонентах среды, то есть в воде, в воздухе, в почве, а также в продовольствии, исследование этих веществ проводится во многих научных центрах мира. В Польше эти вопросы стали, объектом только пока предварительных исследований.

Образование AMУ в среде происходит в результате естественных процессов, к которым можно отнести, между прочим, физиологические процессы некоторых бактерий и растений, а также хозяйственной деятельности человека. К последней нужно отнести образование AMУ в результате трения автомобильных шин об асфальтную поверхность, а также проникновение их из дымоходных труб в воздух или же, вместе со сточными водами, в поверхностные воды, например в районах коксохимической или нефтехимической промышленности, и т.п. Исследования показали, что в процессах очистки сточных вод AMУ восстанавливаются от 20% (при механической очистке) до 96% (при полной очистке). При высокой степени подготовки воды для коммунальных целей достигали до 100% восстановления AMУ.

В ходе работ по оценке качества воды рек Олавы, Нысы Клодзкской и Одры, проведенных в 1976–1977 гг. Институтом формирования среды, были проведены три из обсуждаемых соединений и установлены следующие области концентраций:

бенз(a)пирен	— от 0,0031 до 0,8234 мг дм^{-3} ,
флюорантен	— от 0,0222 до 2,4024 мг дм^{-3} ,
бенз(ghi)перилена	— от 0,0045 до 0,4827 мг дм^{-3} .

Максимальные концентрации этих веществ были обнаружены в профилях, значительно нагруженных сточными водами, выпускаемыми домашними хозяйствами, а также пищевой промышленностью и предприятиями по обдуживанию.

В Одре, в водах которой загрязненность органическими соединениями гораздо выше, чем в остальных реках, концентрации АМУ оказываются тоже более заметными. В водах Олавы и Нысы Клодзской, загрязнение которых по основным физико-химическим показателям одинакова, обнаружена значительная сходимость уровней загрязнения обсуждаемыми соединениями. Здесь отмечены также закономерности в системах концентраций, соответствующих временам года: самые высокие весной, самые низкие летом и осенью.

Область концентраций в водах Одры не обнаруживала каких-то заметных колебаний на протяжении года.

Приведенные результаты исследований представляют собой лишь самую начальную информацию об опасности, вызываемой АМУ, и поэтому проблема нуждается в дальнейшей разработке

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