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# POLYCYCLIC AROMATIC HYDROCARBONS IN AQUATIC ENVIRONMENT OF THE RADOMKA RIVER BASIN. FIELD STUDY IN POLAND

The contamination of the aquatic environment of the Radomka River basin with PAHs was investigated. Ten PAHs were being determined in the river waters and eight PAHs – in the bottom sediments. Some concentrations of PAHs dissolved in water are determined as follows: the lowest – 2 ng/dm$^3$, and the highest – 656 ng/dm$^3$, for the particulate phase – from 19 ng/dm$^3$ to 18 278 ng/dm$^3$. The higher concentrations of PAHs in river occurred in the periods of atmospheric precipitation and rapid snow melting. Their concentrations in the bottom sediments changed periodically from 21 μg/kg to 9500 μg/kg. The predominant compounds were fluoranthene and pyrene.

## 1. INTRODUCTION

Since the early 1980s a number of investigation results have been published. They indicate that aquatic environment is significantly contaminated with polycyclic aromatic hydrocarbons (PAHs). Most of the investigations have dealt with their presence in surface waters, where over 100 compounds of this group have been identified [1]. PAHs get into water primarily in the form of dry and wet deposits from the air [2]. A small amount of PAHs may be washed out from the soil by rainfall, but their main sources are industrial wastewaters, primarily from petrochemical, coking and gasworks plants, accidental spills of petroleum and its products, municipal wastewaters, particularly the raw ones, and road runoffs [3]. There have also been reports discussing the contribution of some natural processes, such as biosynthesis by plants and various aquatic organisms [4]. PAHs are of great interest as some of them have been proved to have strong carcinogenic and mutagenic properties. The studies of surface water contamination with PAHs, due to relatively complicated methods, are carried out for the areas directly subjected to anthropogenic activities in large industrial centres or urban areas in the USA [5], Canada [6] and Poland [7]. The PAHs concentra-

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tions in the rivers flowing in the Katowice [8] and Wrocław [9] regions are determined and the results published, hence the latter can to a certain degree illustrate the contamination of the aquatic environment in Poland with those compounds.

A number of researchers have pointed out that PAHs are found in surface waters in a liquid phase (small amounts) or in the solid phase being adsorbed on the particles forming suspended matter and colloidal solutions [6], [10], [11].

In water, PAHs from various sources, particularly PAHs associated with solid particles, enter bottom sediments via sedimentation and adsorption. The bottom sediments in industrialized areas contain up to as much as several thousand μg/kg of the compounds, e.g. in the River Seine [12]. This can be confirmed by the results of geochemical monitoring of river sediments in Poland [13]. The sediments with very low contents of PAHs (25–100 μg/kg) contain only some compounds of this type, normally those with lower molecular weights. It is assumed that the presence of those harmful substances can be a very good indicator of surface water pollution, as their concentrations in the sediments are thousand times higher than in the water.

In order to improve our knowledge about contamination of the aquatic environment in Poland with PAHs, the aquatic ecosystem of the Radomka River basin has been investigated. This is a poorly industrialized area, where there are no typical sources of PAHs emissions. The Radomka River, whose total length is 107 km, flows into the Vistula River at 432 km of its course. Together with its tributaries the Radomka collects water from primarily agricultural areas. Both the river water and bottom sediments have been investigated in a selected area of the basin.

## 2. SCOPE AND METHODS OF INVESTIGATION

The Radomka from the 57th km of its course to its and the Vistula's confluence as well as its tributary the Mleczna River and the latter's tributary the Pacynka River were selected as the most important for systematic investigation. Two-thirds of the length of the Mleczna is within the city of Radom with its 300 thousand of inhabitants. The investigations lasted one year (1998/1999) and samples were collected every 1 to 2 months. The location of sampling sites is presented in figure 1.

The samples of bottom sediments were collected twice in the autumn of 1997 and in the summer of 1998 (in the close vicinity of the sites of water samples collection); additionally, the samples from the Mleczna were collected in the winter of 1998 and in the spring of 1999.

The methods used in this study made it possible to determine PAHs in the river water. They were both in a liquid phase and adsorbed on suspended solid particles. The surface water samples collected according to the Polish Standard [14] were filtered through a membrane filter (0.45 μm ) using Baker Separex 47. Both the filtrate (0.6–1.0 $dm^3$) and the suspended particles deposited on the filter were analysed by means of different procedures.

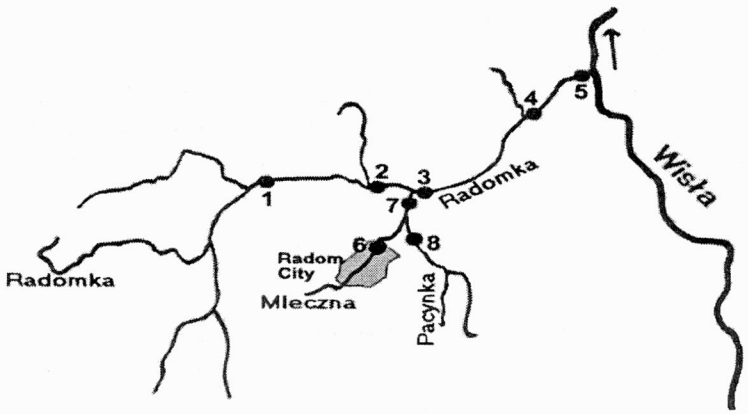


Fig. 1. The location of sampling sites on rivers

Dissolved PAHs were separated from the filtrate using extraction to the solid phase on the extraction disks (EMPORE DISK) [15].

PAHs adsorbed on suspended particles and then retained on the filter were extracted (after drying and weighing) in an ultrasonic field using methanol as a solvent. A similar procedure was used in the case of bottom sediments. After purification in columns with silica gel the extracts obtained were analysed chromatographically by using a HP 1050 liquid chromatograph equipped with a programmable HP 1046 fluorescence detector. The column Vydac 5 PAH 250×4.6 mm with a pre-column Vydac RP $C_{18}$ were used, and the ACN–$H_2O$ mixture was applied as a mobile phase. The separation was performed using a gradient system which allowed qualitative identification being carried out based on standard solutions prepared from the mixture PAH Mix 9 produced by Dr. Ehrenstorfer. This solution contained 16 PAHs chosen by the EPA for their determination in environmental samples.

## 3. RIVER WATER EXAMINATION

Ten compounds were determined in the river water samples: fluoranthene, pyrene, benzo(a)anthracene, chrysene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, dibenzo(a,h)anthracene, benzo(g,h,i)perylene and indeno(1,2,3-c,d)pyrene. The comparison of the concentrations of the 10 PAHs in the Radomka (table 1) proves that their contents in the sector between Przytyk (point 1) and Jedlińsk (point 2) are similar with a slight decrease downstream. At Bartodzieje (point 3) above the confluence of the Radomka and the Mleczna there was a significant increase in the PAH content which exceeded 900 ng/dm$^3$. Farther downstream there was a decrease in the concentrations of total PAHs which before its and the Vistula confluence ranged from 30 to 73 ng/dm$^3$ at point 5. The highest and highly diversified values (from 157 ng/dm$^3$ to as much as 18383 ng/dm$^3$) were obtained for the Mleczna waters at point 6, which was located in the centre of Radom. At the next sampling point, 10 km away, the values

were many times lower, with the maximum values being even 30 times lower. This substantial decrease in PAHs in the water of such a small river resulted mainly from diluting contaminants with water from its tributary, the Pacynka River, and the treated effluents discharged from the municipal treatment plant. PAHs concentrations in the Pacynka did not show significant variations and ranged from 20 to 50 ng/dm$^3$.

Table 1

Total PAHs contents in the river waters [ng/dm$^3$]

| Samplig points | Sampling sites | Course of river [km] | Σ10 PAHs mean | Σ10 PAHs range | Σ6 PAHs mean | Σ6 PAHs range |
|---|---|---|---|---|---|---|
| 1 | the Radomka (Przytyk) | 58 | 223 | 15–625 | 171 | 12–481 |
| 2 | the Radomka (Jedlińsk) | 45 | 139 | 18–452 | 110 | 14–452 |
| 3 | the Radomka(Bartodzieje) | 33 | 243 | 34–973 | 122 | 19–420 |
| 4 | the Radomka ( Brzóza) | 16 | 107 | 31–260 | 65 | 17–144 |
| 5 | the Radomka ( Kłoda) | 3 | 55 | 31–73 | 37 | 18–51 |
| 6 | the Mleczna (Radom) | 16 | 6226 | 157 – 18383 | 3499 | 87–10375 |
| 7 | the Mleczna (Owadów) | 5 | 283 | 49–632 | 204 | 22–554 |
| 8 | the Pacynka (Lesiów) | 1 | 38 | 20–50 | 30 | 30–31 |

10 PAHs – F, P, B(a)A, Ch, B(b)F, B(k)F, B(a)P, B(ghi)P, I(cd)P, D(a)A.
6 PAHs – F, B(b)F, B(k)F, B(a)P, B(ghi)P, I(cd)P.

The total contents of the six PAHs: fluoranthene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, benzo(g,h,i)perylene and indeno(c,d)-pyrene, which according to the World Health Organization [16] should be monitored, in the Radomka River (the villages of Przytyk (point 1), Jedlińsk (point 2) and Bartodzieje (point 3)) are close to those detected in the Vistula (13–365 ng/dm$^3$) at its source (the village of Strumień) and in the Pszczynka River (18–408 ng/dm$^3$) (the village of Łąka). The other values are considerably lower than those found in the rivers of the Katowice region [8], even at the end of their courses. This comparison justifies describing the Radomka River as lightly polluted.

PAH concentration in the Mleczna (the village of Owadów, point 7) was comparable to this detected in the Pszemsza River in Chełmek [8], which varied from 19 to 586 ng/dm$^3$, and to those obtained in the late 1990s for the Widawa and the Barycz Rivers [9], where the total contents of six PAHs were 300 ng/dm$^3$ or lower and 600 ng/dm$^3$ or lower, respectively. This justified classifying both rivers as the least polluted ones in the Wrocław region. The maximum concentrations determined in the Mleczna were as much as three times higher than those detected in the Oława River (3500 ng/dm$^3$) [9]. This comparison justifies including the water of the Mleczna in the group of heavily polluted rivers in the city of Radom (point 6) [17].

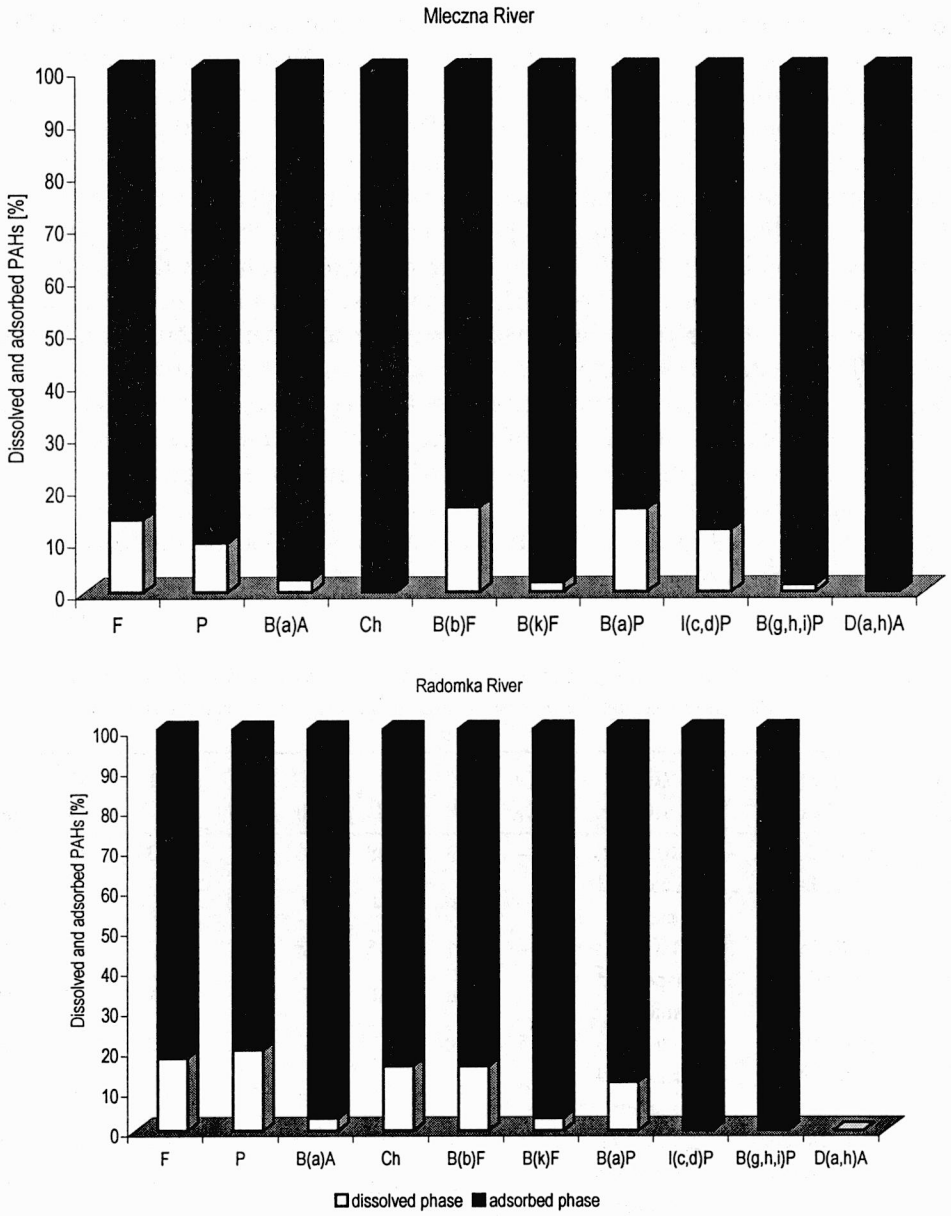


Fig. 2. Dissolved and adsorbed PAHs in river water [%]

Analytical methods involving the determination of PAHs in the liquid and particulate phases showed that those contaminants occur in the river waters tested mostly in the adsorbed phase. Only fluoranthene, pyrene and benzo(b)fluoranthene had a higher share of the dissolved phase in the samples with lower total PAHs contents reaching 16% to 20% of the total content (figure 2). In vast majority of samples, the

share of those compounds in the dissolved phase in the waters of the Radomka and the Mleczna was slightly higher than 2% of the total content. Hydrocarbons characterized by low water solubility, e.g. dibenzo(a,h)anthracene or benzo(g,h,i,)perylene, were not identified in that phase. The concentrations of PAHs in the dissolved phase found by GUSTAFSON [10] usually were higher for compounds of lower molecular weight, such as naphthalene, fluorene or phenanthrene. The ratio of both phases varied from 50% : 50% for fluoranthene to a considerably higher content of the adsorbed phase for such compounds as B(b)F, B(k)F, B(e)P, B(a)P or I(c,d)P, although their concentrations were by an order or even two higher than those determined in the Radomka. THAO PHAM et al. [6] observed in Canadian river waters that the compounds from benzo(a)anthracene to indeno(c,d)pyrene occurred primarily in the adsorbed phase.

Weather conditions had a certain effect on the occurrence of PAHs in the Radomka and the Mleczna. The analysis of the results obtained showed a considerable increase in their concentrations in the periods of atmospheric precipitation and rapid snow melting. PAHs variations in the Radomka ranged from several to over 900 ng/dm$^3$ for the total of the ten PAHs, whereas the variations in the Mleczna was up to 15 000 ng/dm$^3$ (table 2).

Table 2

Contents of ten PAHs in the adsorbed phase in the Radomka River and Mleczna River water (ng/dm$^3$) and the amount of suspended matter (mg/dm$^3$) during the investigation period

| Samplig points | Samplig sites | Total contents | III 98 | IV 98 | V 98* | VII 98 | X 98 | XII 98** | II 99 | III 99 |
|---|---|---|---|---|---|---|---|---|---|---|
| 3 | the Radomka (Bartodzieje) | Σ 10 PAHs | – | 33 | 298 | 46 | 41 | 973 | 40 | 76 |
| | | Suspended matter | – | n.d. | n.d. | 9 | 3.5 | 52 | 4 | 4 |
| 4 | the Radomka (Brzóza) | Σ 10 PAHs | – | 27 | 135 | 28 | 41 | 222 | 38 | 260 |
| | | Suspended matter | – | n.d. | n.d. | 7 | 4 | 20 | 4.5 | 15 |
| 6 | the Mleczna (Radom) | Σ 10 PAHs | 157 | 235 | 7524 | – | – | 14225 | 3004 | 160 |
| | | Suspended matter | 9 | 12 | 78 | – | – | 165 | 36 | 5 |
| 7 | the Mleczna (Owadów) | Σ 10 PAHs | 132 | – | 519 | 49 | – | 632 | 200 | 164 |
| | | Suspended matter | 7 | – | n.d. | 9 | – | 84 | 12 | 10 |

*During the rain.
**During the rapid snow melting.

Figure 3 shows the changes in the contents of the adsorbed phases of individual PAHs in the waters of the two rivers, depending on atmospheric precipitations. The concentrations of B(k)F and I(c,d)P were substantially increased in more polluted sector of the Radomka, while in the Mleczna (point 6), where it collected the precipitation runoffs from a larger part of Radom, there was even a 50-fold increase in the

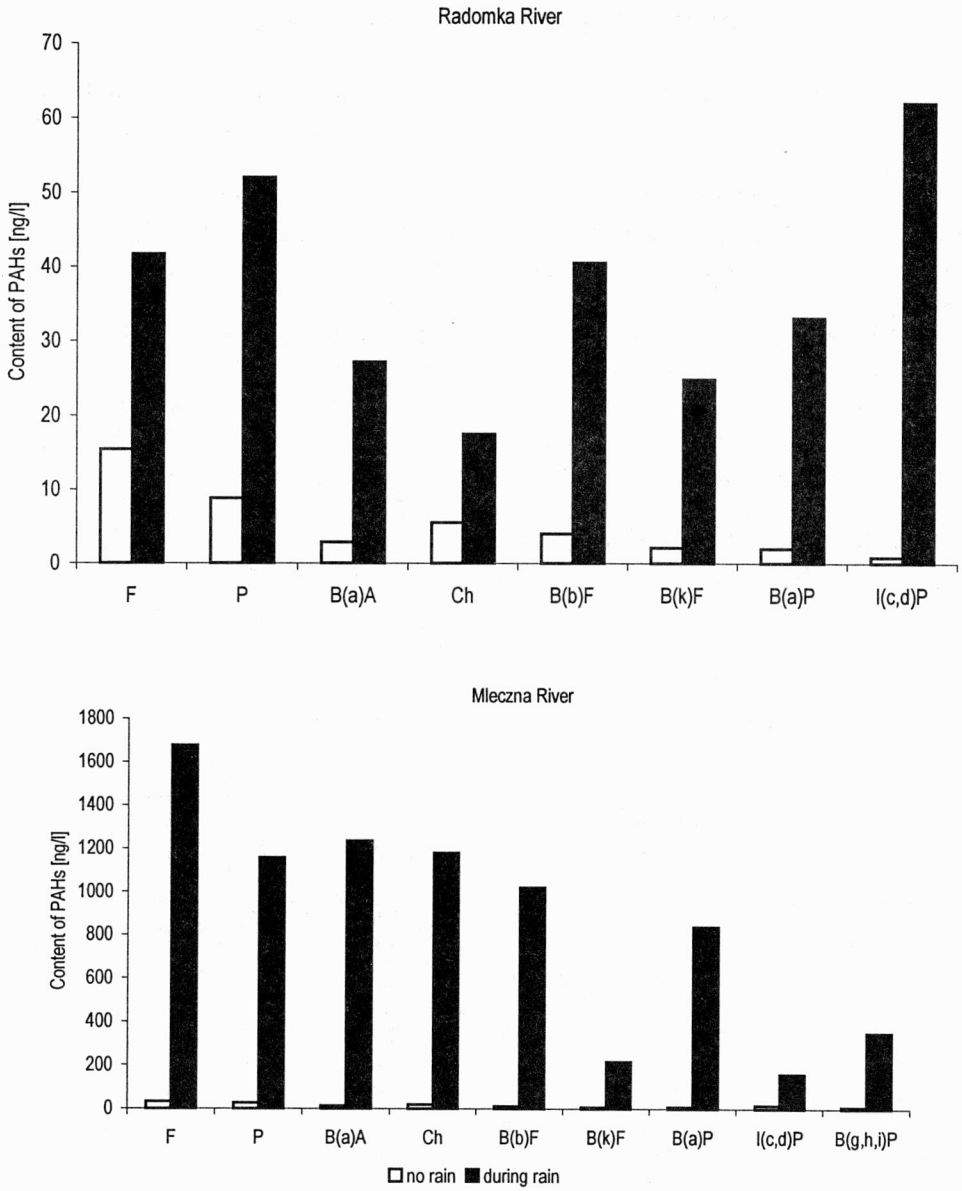


Fig. 3. Contents of PAHs adsorbed in the river water (ng/dm$^3$) in the period with rain and without rain

concentrations of all hydrocarbons. The runoffs carry very large quantities of particles washed out from various surfaces, which leads to a considerable increase in the amount of suspended matter and contaminants in the adsorbed phase associated with it in the Mleczna.

## 4. BOTTOM SEDIMENTS INVESTIGATION

PAHs occurred in various concentrations in the bottom sediments of the Radomka and the Mleczna, depending on the location of sites along the river course. In table 3 the contents of eight compounds normally found in the samples tested are given. The total PAH contents in the Radomka sediments, which had a low content of organic matter (0.17% to 0.45%), ranged from 87 μg/kg of dry matter to 21 μg/kg of dry matter. Lower concentrations of the compounds can be found in the samples collected at the sites farther from the confluence of the Mleczna and the Radomka, i.e. in the villages of Brzóza (point 4) and Kłoda (point 5) (table 3), and also in the sediments from the Pacynka. The low concentrations of PAHs in the Radomka are comparable to those detected in the Bzura River and the Wieprz River in 1993; however, only phenanthrene and fluoranthene were determined there [13]. The sediments in the Mleczna, particularly those collected at point 6 in the centre of Radom, exhibited totally different characteristics. They had a higher content of organic matter, up to as much as 27.5%, a variable particle-size distribution with a higher percentage content of a very fine fraction, below 63 μm, reaching above 4%.

Table 3

Concentrations of individual PAHs in the bottom sediments of the Radomka and the Mleczna (μg/kg)

| Sampling points | Sampling sites | Sampling period (year) | F | P | B(a)A | Ch | B(b)F | B(k)F | B(a)P | I(cd)P | Σ 8 PAH |
|---|---|---|---|---|---|---|---|---|---|---|---|
| 2 | the Radomka River (Jedliński) | 1997 | 22.5 | 18.3 | 7.2 | 7.6 | 11.4 | 7.05 | 7.6 | 5.3 | 87 |
| | | 1998 | 19.9 | 10.8 | 2.5 | 5.1 | 4.5 | 2.7 | 3.3 | 2.7 | 51.7 |
| 3 | the Radomka River (Bartodzieje) | 1997 | 12.5 | 10.9 | 4.2 | 4.2 | 7.1 | 4.2 | 4.9 | 4.0 | 52 |
| | | 1998 | 3.5 | 4.1 | 2.2 | 2.6 | 3.1 | 2.0 | 2.9 | 2.6 | 23 |
| 4 | the Radomka River (Brzóza) | 1997 | 4.7 | 3.2 | 2.8 | 2.9 | 4.1 | 2.7 | 3.0 | 1.6 | 25 |
| | | 1998 | 4.2 | 7.1 | 3.3 | 3.4 | 4.7 | 2.6 | 2.8 | 1.5 | 29.7 |
| 5 | the Radomka River (Kłoda) | 1997 | 5 | 2.5 | 2.7 | 3.0 | 4.3 | 1.9 | 2.0 | 2.0 | 21 |
| | | 1998 | 4.4 | 3.5 | 2.6 | 5.1 | 4.5 | 2.7 | 3.4 | 2.8 | 29 |
| 6 | the Mleczna River (Radom) | 1997 | 1444 | 1807 | 1104 | 1100 | 1285 | 600 | 1228 | 939 | 9507 |
| | | 1998 | 70 | 60 | 15 | – | 40 | 20 | 50 | 40 | 305 |
| | | 1998 | 1090 | 1080 | 413 | 456 | 446 | 233 | 525 | 255 | 4498 |
| | | 1999 | 425 | 464 | 325 | 106 | 464 | 244 | 319 | 376 | 2720 |
| 7 | the Mleczna River (Owadów) | 1997 | 97 | 91 | 70 | 107 | 85 | 44 | 74 | 69 | 637 |
| | | 1998 | 122 | 100 | 27 | 51 | 74 | 30 | 62 | 32 | 498 |
| 8 | the Pacynka River | 1997 | 5.0 | 3.4 | 2.6 | 3.0 | 4.7 | 2.5 | 2.8 | 1.0 | 25 |

The concentrations of total PAHs in those sediments were at least by one or even two orders higher than those found in the Radomka sediments. The samples taken in Radom (point 6), at a site where surface runoffs were discharged, showed maximum

total contents of hydrocarbons, sometimes above 9500 μg/kg, which could be compared to the heavily contaminated sediments from the Słupia River in the town of Słupsk or the Odra River in the village of Wróblin [13]. Such high concentrations appeared periodically. The concentrations of several hundred μg/kg were also found at point 6. Such a concentration was characteristic of the sediments from the site where the Mleczna was less contaminated and supplied additionally with the waters from the Pacynka.

Fluoranthene and pyrene predominate in all the sediment samples which also is a characteristic feature of the waters tested. The contents of those hydrocarbons range from about 15% to 25% of total PAHs concentration. In most of the samples, B(b)F is the third, in respect of its share, component. It consists 13.1–16.4% of the total content. B(a)P content is slightly lower than that of B(b)F, ranging from 9.5% to 16.4% (figure 4). The analysis of the results for the sediments in the Mleczna shows an extremely large variation in the total content of the compounds in the samples collected at various time intervals at different seasons. Taking into account the above results and the results of geochemical monitoring of river sediments [13] we may conclude that the total content of eight PAHs at the mouth of the Mleczna is then 2885 μg/kg. The predominant compounds are chrysene (27.7%), fluoranthene (20%) and pyrene (16%).

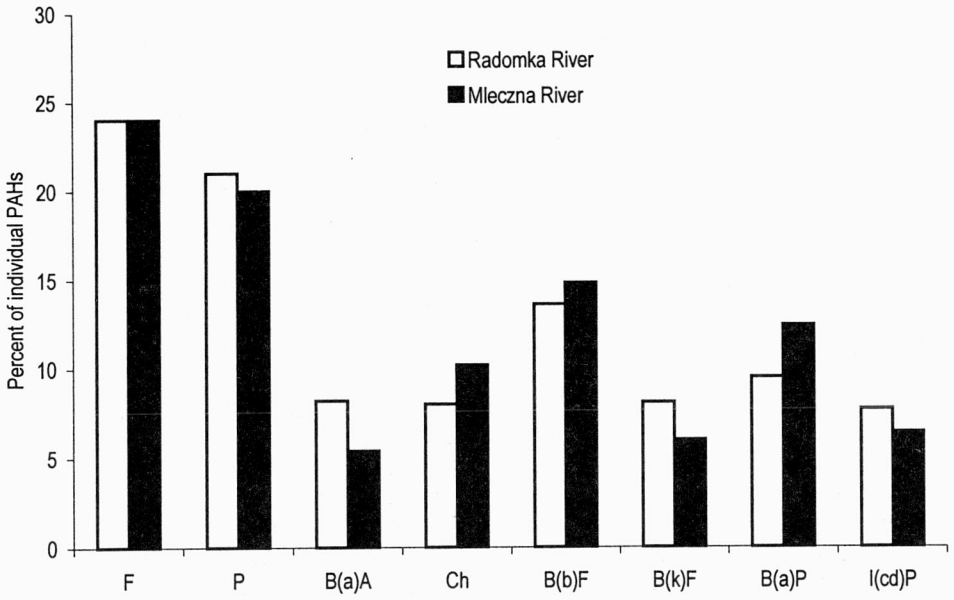


Fig. 4. Distribution of individual PAHs [%] in sediments

Similar changes were observed by KLIMASZEWSKA [18] in the Narew River sediments in which the total content of phenanthrene, fluoranthene, chrysene, pyrene and benzo(a)pyrene was 1296 μg/kg in 1997 and 103 μg/kg a year later.

OLLIVON et al. [12] found that in the bottom sediments of the River Seine fluoranthene and pyrene occurred in the highest concentrations, while the concentration of B(a)P was normally slightly lower than that of B(b)F with the total content of eight PAHs at the level of 4000 to 5000 μg/kg, i.e. close to some of the results for the Mleczna sediments.

## 5. CONCLUSIONS

• The concentrations of PAHs in river water in a poorly industrialized area exhibit variable values – from relatively low values for the Radomka (maximum total contents of PAHs – 973 ng/dm$^3$), which allow us to classify it as slightly polluted with those compounds, to periodically high values for the Mleczna (up to max. 18383 ng/dm$^3$), which justify including it in a group of the rivers heavily polluted with PAHs.

• Fluoranthene and pyrene, both in the soluble and adsorbed phases, occur in the highest concentrations of the ten PAHs investigated. Higher-molecular weight compounds, starting with B(a)P, show considerably lower percentage contents in the total contents being determined. A significant increase in the concentrations of such hydrocarbons as B(a)P, B(b)F and I(c,d)P in river water occurs periodically during precipitations.

• PAHs discharged to rivers together with surface runoffs are intensively accumulated in bottom sediments. As a result, a serious contamination of bottom sediment with PAHs is limited to hundred meters of river length.

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## WIELOPIERŚCIENIOWE WĘGLOWODORY AROMATYCZNE W ŚRODOWISKU WODNYM ZLEWNI RZEKI RADOMKI. BADANIA TERENOWE NA OBSZARZE POLSKI

Przedstawiono wyniki badań dotyczących zanieczyszczenia środowiska wodnego wybranego obszaru zlewni Radomki. Oznaczano 10 wielopierścieniowych węglowodorów aromatycznych (WWA) w wodzie rzecznej, przy czym uwzględniono, że występują one w postaci rozpuszczonej ($C_{min}$ – 2 ng/dm$^3$, $C_{maks}$ – 656 ng/dm$^3$) i w postaci zaadsorbowanej ($C_{min}$ – 19 ng/dm$^3$, $C_{maks}$ – 18 278 ng/dm$^3$). Stwierdzono, że większość węglowodorów występuje w postaci zaadsorbowanej. Uzyskane wyniki wskazują, że w okresie opadów atmosferycznych i intensywnego topnienia śniegu stężenia WWA wyraźnie zwiększają się. W rzecznych osadach dennych wykrywano 8 WWA, których stężenie wahało się od 21 μg/kg do okresowo nawet 9500 μg/kg. Wśród wykrywanych związków zarówno w osadach dennych, jak i w wodach dominowały fluoranten i piren.