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Assessing the concentration of chosen PAH in drinking water

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ABSTRACT

Many of the pollutants present in water are highly toxic to living organisms and hardly biodegradable. Legal acts regulate the water quality defining the type and the acceptable concentrations of particular pollutants in drinking water. Organic micropollutants such as polycyclic aromatic hydrocarbons (PAH) have negative impact on health and life of humans, what makes it necessary to monitor and assess their concentration in drinking water. The aim of this research was the assessment of concentration of chosen PAH in drinking water taken from different places of technological system of water treatment plant located in Silesian Voivodeship in the fall, and later in the spring. Solid phase extraction method has been used to separate PAH from water samples. To analyze the concentration and composition of PAH, the method of capillary gas chromatography has been used. The concentration of chosen PAH has been changing according to sample type and the time it was taken. The results obtained during the research has been compared with norms contained in Council Directive 98/83/EC of 3 November 1998 on the quality of water intended for human consumption and Regulation of the Minister of Health of 7 December 2017 on the quality of water intended for human consumption.

Keywords: Polycyclic aromatic hydrocarbons; Water treatment plant; Drinking water

1. Introduction

Water is one of the most important chemical compounds on earth. The water quality has big impact on all of the living organisms. The concentration of organic micropollutants in water intended for drinking purposes is the problem in many countries. Awareness of danger which comes with water contaminated with organic substances causes the actions to stop the water quality from deterioration [1–5]. Organic micropollutants present in surface water are a wide group of chemical compounds, which are toxic and dangerous for living organism. The group includes substances such as PAH [4]. There are about 300 polycyclic aromatic compounds but the most frequently analyzed is the group of 16 PAH listed by the Environmental Protection Agency (EPA). The substances on the EPA list are highly toxic and must be monitored in the environment [2,6–9].

PAH are widely present in the environment which is the main effect of pyrolytic processes. The main sources of those compounds might be natural (wildfire, volcanic activity) or anthropogenic (industrial operations: coke plants, refineries, steel mills, power plants) [6,10–12]. The presence of PAH in surface waters intended for human consumption is connected to anthropogenic sources, such as industrial and municipal wastewater [4,6,9,10].

PAH are composed of multiple aromatic rings (2–13) in which the electrons are delocalized [6,13–15]. They are present in the environment as a mixture, they do not occur separately [10–12]. The concentration of PAH in natural waters is very low, but nevertheless they are still considered dangerous for living organisms because of their toxicity, carcinogenic and mutagenic character and the bioaccumulation ability. The most harmful and very extensively studied substance from this group is benzo(a)pyrene [6,10–12,16–19].

Documents that regulate the permissible concentration of PAH in drinking water are Council Directive 98/83/EC of 3 November 1998 on the quality of water intended for human

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consumption and Regulation of the Minister of Health of 7 December 2017 on the quality of water intended for human consumption. The permissible level of benzo(a)pyrene concentration in water amounted to 0.01 μg/L (10 ng/L). In case of the sum of PAH (benzo(b)fluoranthene, benzo(k) fluoranthene, benzo(ghi)perylene and indeno(1,2,3-cd) pyrene) the permissible level of concentration amounts to 0.1 μg/L (100 ng/L) [5,20,21].

The aim of this research was to evaluate the concentration and composition of PAH in water intended for human consumption, taken from water treatment plant located in Silesian Voivodeship in different stages of water treatment process.

2. Materials and methods

2.1. Materials

For the research the water from water treatment plant in Silesian Voivodeship was used. Water treatment plant belongs to the Upper Silesian Water Supply Company. The water for the company is supplied from the reservoir which was built over the period of 1935 to 1939, and then adapted to water supplying in the years 1948 to 1951. Nowadays the area of the reservoir cover approximately 6 km^2 and its volume is approximately 17 mln m^3 [20,22–24].

Water samples from water treatment plant were taken in the fall and then in the spring, in chosen places of the technological system of the plant:

- water sample from the reservoir,
- water sample after primary ozonation (only in the spring),
- water sample after final ozonation,
- water sample from the pumping station,
- water sample from the consumer (water pipes length is approximately equal to 1.6 km).

Water travel time to reach each places of sample taking was considered. The depth of water sampling was approximately 0.4 m. The temperature of water samples varied from 4°C to 8°C in the fall and 6°C to 10°C in the spring.

Abbreviated scheme of the technological system of water treatment plant with the places of sample taking pointed out is shown in Fig. 1.

Water from the reservoir undergoes processes:

- primary oxidation using ozone,
- coagulation in flash mixing and slow mixing tanks,
- rapid anthracite-sand filtration,
- secondary oxidation using ozone,
- adsorption on filter with granular activated carbon,
- disinfection using ozone [23].

The water samples were taken in different places of water treatment plant technological system to check if the organic micropollutants such as PAH are efficiently removed in conventional water treatment processes. To check if the changing seasons have essential impact on PAH concentration in surface water, the samples were taken in the fall and later in the spring.

2.2. Analytical procedure

For PAH extraction from the water, the solid phase extraction method was applied, using 500 mg Octadecyl C_{18} 6 mL Bakerbond columns. For this purpose 0.5 L of water was taken. Petroleum ether and dichloromethane were added to the water samples in the ratio 3:2 (52.5 mL of petroleum ether and 35 mL of dichloromethane). The samples were passed through preconditioned SPE extraction columns. To prepare the columns for PAH extraction 6 mL of hexane were passed through the columns filling, then the columns were dried in vacuum for 2 min. After that 6 mL of methanol and 6 mL of distilled water were passed through the column filling. After the water samples were passed through them, the columns with PAH trapped on sorbent were dried in vacuum for 30 min. PAH were then eluted from the columns filling using 3 × 1 mL of hexane. Extracts were condensed to the volume of 1 mL. Prepared samples were analyzed with the use of gas chromatography and mass spectrometer (GC-MS) [19,20]. Standard mixture of 16 PAH by RESTEK company was used. 0.002 mL of extract were injected on DB-5 column. Helium was used as a carrier gas. PAH determination was performed in triplicate. To eliminate the error as a result of sample taking method, the water samples were averaged and those representative samples were put under further procedures. Because of this, the main error impacting the variation of concentration of chosen PAH in the samples is analytical error. Standard deviations of PAH concentration in water samples are presented in Tables 1 and 2.

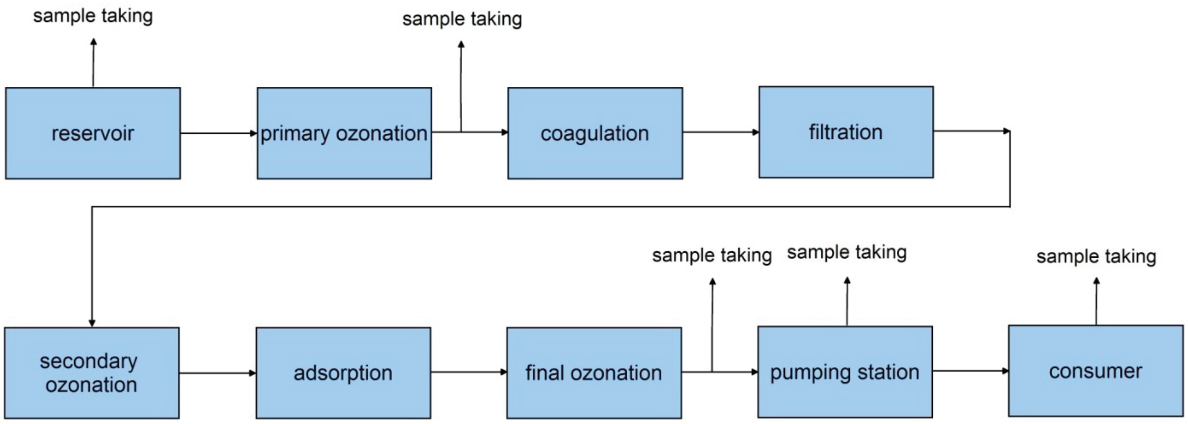

Fig. 1. Abbreviated scheme of the technological system of water treatment plant with the places of sample taking pointed out.

3. Results and discussion

The results of PAH concentration in water samples taken in the fall are presented in Table 1.

For the discussion of the results, PAH were divided into the following groups:

- PAH standardized in the Council Directive 98/83/EC of 3 November 1998 on the quality of water intended for human consumption and Regulation of the Minister of Health of 7 December 2017 on the quality of water intended for human consumption: benzo(a)pyrene, the sum of PAH (benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(ghi)perylene, indeno(1,2,3-cd)pyrene),
- two-ring PAH: naphthalene,
- three-ring PAH: acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene,
- four-ring PAH: fluoranthene, pyrene, benzo(a)anthracene, chrysene,
- six-ring PAH: dibenzo(a,h)anthracene.

In the sample taken from the reservoir, the concentration of benzo(a)pyrene amounted to 123 ng/L, which is much more than the acceptable concentration standardized in the Council Directive 98/83/EC of 3 November 1998 on the quality of water intended for human consumption and Regulation of the Minister of Health of 7 December 2017 on the quality of water intended for human consumption. The concentration of sum of PAH included in the regulations was also above the permissible level. The sum of benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(ghi)perylene and indeno(1,2,3-cd)pyrene amounted to 589 ng/L. The highest concentration, that is, 190 ng/L, was noted in case of benzo(b)fluoranthene and benzo(k)fluoranthene. The research shows that there are high concentrations of benzo(b)fluoranthene and benzo(k)fluoranthene in some surface waters in Poland [25]. The lowest concentration, that is, 89 ng/L, was detected for benzo(ghi)perylene. The concentration of two-ring naphthalene amounted to 79 ng/L. In the group of three-ring PAH, acenaphthylene and acenaphthene were not detected. Concentrations of fluorene, phenanthrene and anthracene amounted to 11, 130 and 61 ng/L. Analyzing the group of four-ring PAH, high levels of benzo(a)anthracene (154 ng/L) and chrysene (158 ng/L) were detected. Fluoranthene and pyrene were not detected. The concentration of six-ring PAH, that is, dibenzo(a,h)anthracene in analyzed sample amounted to 28 ng/L.

In the water taken after final ozonation, the concentration of benzo(a)pyrene was very low, that is, 6 ng/L, which is below the permissible level. The sum of PAH amounted to 43 ng/L, which is also below the acceptable level contained in the Council Directive 98/83/EC of 3 November 1998 on the quality of water intended for human consumption and Regulation of the Minister of Health of 7 December 2017 on the quality of water intended for human consumption. The concentration of naphthalene in the sample amounted to 64 ng/L, and decreased insignificantly compared with the sample taken from the reservoir. In case of three-ring PAH, the presence of acenaphthylene and anthracene was not noted. Acenaphthene which was not present in the water taken from the reservoir, was detected in amount of 11 ng/L. In the sample, the presence of four-ring fluoranthene and pyrene was detected. Their concentrations amounted to 11 and 6 ng/L. They were not present in the sample taken from the reservoir. The concentrations of benzo(a)anthracene and chrysene decreased compared with the water taken from the reservoir. In the sample six-ring dibenzo(a,h)anthracene was not detected. The concentrations of the analyzed PAH mostly decreased after the final ozonation. According to the literature data, PAH degradation using ozone after adsorption is highly effective [25].

In the water sample taken from the pumping station none of the compounds standardized in the Council Directive 98/83/EC of 3 November 1998 on the quality of water intended for human consumption and Regulation of the Minister of Health of 7 December 2017 on the quality of water intended for human consumption were detected. The concentration of naphthalene, that is, 27 ng/L, decreased compared with the previous samples. In case of three-ring PAH, the presence of acenaphthene and fluorene was detected. Their concentration amounted to 6 and 4 ng/L. None of the compounds of the four-ring and six-ring PAH were detected in the analyzed sample.

In the sample taken from the consumer, the presence of benzo(a)pyrene was not noted. Not detected in the sample taken from the pumping station benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(ghi)perylene and indeno(1,2,3-cd)pyrene were present what might be caused by the waterworks bad condition. Their concentrations amounted to 9, 9, 6 and 8 ng/L. The sum of those compounds (the sum of PAH) amounted to 32 ng/L, which was below the permissible level contained in the Council Directive 98/83/EC of 3 November 1998 on the quality of water intended for human consumption and Regulation of the Minister of Health of 7 December 2017 on the quality of water intended for human consumption. The concentration of naphthalene increased almost twice (from 27 to 51 ng/L) compared with the previous sample. Concentrations of acenaphthene and fluorene were on the similar level compared with the water sample taken from the pumping station. In the water sample, phenanthrene (20 ng/L) and four-ring PAH were detected, but their concentrations were very low. The concentration of six-ring dibenzo(a,h)anthracene amounted to 6 ng/L.

The results of PAH concentration in water samples taken in the spring are presented in Table 2.

In the water sample taken from the reservoir in the spring, the presence of benzo(a)pyrene and the sum of PAH (benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(ghi)perylene, indeno(1,2,3-cd)pyrene) was not detected. The concentration of two-ring naphthalene amounted to 36 ng/L. Three-ring PAH, that is, acenaphthene, fluorene and phenanthrene where present in the amount of 6, 3 and 6 ng/L. The concentration of four-ring fluoranthene and pyrene amounted to 2 ng/L. Dibenzo(a,h)anthracene was not detected in the water sample.

In the sample taken after primary ozonation none of the compounds standardized in the Council Directive 98/83/EC of 3 November 1998 on the quality of water intended for human consumption and Regulation of the Minister of Health of 7 December 2017 on the quality of water intended for human consumption were detected. The concentration of naphthalene decreased compared with the water sample

Table 1
PAH concentration in water samples taken in the fall

Compound	Reservoir		Final ozonation		Pumping station		Consumer	
	Concentration in water sample (ng/L)	Standard deviation (ng/L)	Concentration in water sample (ng/L)	Standard deviation (ng/L)	Concentration in water sample (ng/L)	Standard deviation (ng/L)	Concentration in water sample (ng/L)	Standard deviation (ng/L)
Naphthalene	79	0.6	64	0.3	27	0.6	51	0.5
Acenaphthylene	nd	–	nd	–	nd	–	nd	–
Acenaphthene	nd	–	11	0.2	6	0.2	7	0.2
Fluorene	11	0.7	8	0.1	4	0.1	5	0.2
Phenanthrene	130	0.9	32	0.4	nd	–	20	0.3
Anthracene	61	0.6	nd	–	nd	–	nd	–
Fluoranthene	nd	–	11	0.2	nd	–	3	0.1
Pyrene	nd	–	6	0.1	nd	–	2	0.2
Benzo(a)anthracene	154	0.8	3	0.1	nd	–	2	0.1
Chrysene	158	0.7	8	0.2	nd	–	2	0.3
Benzo(b)fluoranthene	190	0.9	11	0.3	nd	–	9	0.2
Benzo(k)fluoranthene	190	0.9	11	0.4	nd	–	9	0.2
Benzo(a)pyrene	123	0.7	6	0.2	nd	–	nd	–
Indeno(1,2,3,-cd)pyrene	120	0.8	10	0.3	nd	–	8	0.3
Dibenzo(a,h)anthracene	28	0.2	nd	–	nd	–	6	0.2
Benzo(ghi)perylene	89	0.6	11	0.4	nd	–	6	0.3

nd – Not detected.

Table 2
PAH concentration in water samples taken in the spring

Compound	Reservoir		Primary ozonation		Final ozonation		Pumping station		Consumer	
	Concentration in water sample (ng/L)	Standard deviation (ng/L)	Concentration in water sample (ng/L)	Standard deviation (ng/L)	Concentration in water sample (ng/L)	Standard deviation (ng/L)	Concentration in water sample (ng/L)	Standard deviation (ng/L)	Concentration in water sample (ng/L)	Standard deviation (ng/L)
Naphthalene	36	0.5	23	0.3	18	0.3	26	0.3	23	0.5
Acenaphthylene	nd	–	nd	–	nd	–	nd	–	nd	–
Acenaphthene	6	0.2	4	0.2	4	0.1	6	0.2	6	0.2
Fluorene	3	0.1	3	0.1	2	0.1	nd	–	3	0.1
Phenanthrene	6	0.2	5	0.2	3	0.2	nd	–	3	0.1
Anthracene	nd	–	nd	–	nd	–	nd	–	nd	–
Fluoranthene	2	0.1	3	0.1	4	0.2	nd	–	nd	–
Pyrene	2	0.1	2	0.1	4	0.2	nd	–	nd	–
Benzo(a)anthracene	nd	–	4	0.2	15	0.4	nd	–	nd	–
Chrysene	nd	–	1	0.1	5	0.2	nd	–	nd	–
Benzo(b)fluoranthene	nd	–	nd	–	15	0.5	nd	–	nd	–
Benzo(k)fluoranthene	nd	–	nd	–	15	0.5	nd	–	nd	–
Benzo(a)pyrene	nd	–	nd	–	15	0.4	nd	–	nd	–
Indeno(1,2,3,-cd)pyrene	nd	–	nd	–	13	0.6	nd	–	nd	–
Dibenzo(a,h)anthracene	nd	–	nd	–	nd	–	nd	–	nd	–
Benzo(ghi)perylene	nd	–	nd	–	13	4	nd	–	nd	–

nd – Not detected.

taken from the reservoir, and it amounted to 23 ng/L. Similar dependence has been observed in case of three-ring PAH, that is, acenaphthene and phenanthrene. The concentration of fluorene has not changed compared with the previous sample. In the group of four-ring PAH, the concentration of fluoranthene slightly increased (from 2 to 3 ng/L), while the concentration of pyrene has not changed. The presence of benzo(a)anthracene and chrysene was noted in amount of 4 and 1 ng/L. Similar to the previous water sample, dibenzo(a,h)anthracene was not detected.

In the water sample taken after final ozonation, the concentration of benzo(a)pyrene amounted to 15 ng/L, which was above the permissible level. The concentration of the sum of PAH was below the permissible level. The amount of naphthalene in the sample slightly decreased, similar to the concentrations of three-ring PAH, that is, fluorene and phenanthrene. The concentration of acenaphthene has not changed. The concentrations of four-ring PAH slightly increased compared with the previous sample. Similar to the previous water samples, dibenzo(a,h)anthracene was not detected.

In the water sample taken from the pumping station, the only detected compounds were naphthalene (26 ng/L) and acenaphthene (6 ng/L).

In the water sample taken from the consumer, the detected compounds were naphthalene amounted to 3 ng/L and three-ring PAH, that is, acenaphthene (6 ng/L), fluorene (3 ng/L) and phenanthrene (3 ng/L).

Comparison of the concentrations of PAH in the water samples taken from the reservoir in the fall and in the spring shows that the most of the compounds had higher concentrations in the fall. Acenaphthylene is the only compound not detected in the water samples in the fall and in the spring. Higher concentrations of PAH in the spring were observed in case of acenaphthene, fluoranthene and pyrene. Those compounds were not present in water in the fall. According to the literature data, the concentrations of PAH in the water might depend on a season and on changes in ambient temperature what connects with higher combustion of fossil fuels in fall (e.g., for heating houses) when the temperature is lower [26–28]. The research shows that the higher concentrations of chosen PAH are in the fall compared with the spring [27].

Comparison of the concentrations of PAH in the water samples taken after the final ozonation in the fall and in the spring shows that in the water samples taken in the fall, higher concentrations were observed in case of naphthalene, acenaphthene, fluorene, phenanthrene, fluoranthene, pyrene and chrysene. In the water samples taken in the spring, higher concentrations were observed in case of benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, indeno(1,2,3-cd)pyrene and benzo(ghi)perylene.

Comparison of the concentrations of PAH in the water samples taken from the pumping station in the fall and in the spring shows that in those samples the presence of naphthalene, acenaphthene and fluorene (only in the fall) were noted. In case of those compounds, their concentrations were higher in the samples taken in the fall.

Comparison of the concentrations of PAH in the water samples taken from the consumer in the fall and in the spring shows that all of the compounds had higher concentrations in the fall.

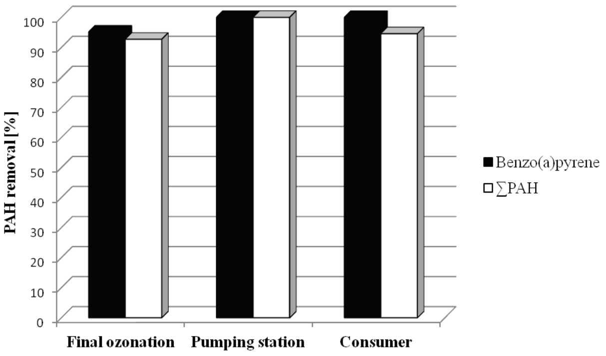

Fig. 2. Percentage of PAH removal from water samples taken in the fall.

In Fig. 2 percentage of PAH removal from water samples taken in the fall is shown. Only PAH standardized in the Council Directive 98/83/EC of 3 November 1998 on the quality of water intended for human consumption and Regulation of the Minister of Health of 7 December 2017 on the quality of water intended for human consumption were considered. The removal of benzo(a)pyrene after final ozonation amounted to 95%. In the water samples taken from the pumping station and from the consumer the removal of benzo(a)pyrene amounted to 100%. The removal of the sum of PAH after final ozonation amounted to 93%. In the water sample taken from the pumping station the removal of the sum of PAH amounted to 100% but it decreased to 95% after the water reached the consumer what might be caused by the waterworks bad condition, as it was previously mentioned.

4. Conclusions

Conducted research allows to draw the following conclusions:

- All the water samples taken from water treatment plant contained PAH.
- Concentration of chosen PAH compounds was depending on the time and place in the technological system of the plant that the water sample was taken.
- The only substance that was present in all the water samples was naphthalene.
- Anthracene was present only in the water sample taken from the reservoir in the fall.
- The only compound not detected in any of the samples was acenaphthylene.
- The highest concentration of PAH was detected in the water sample taken from the reservoir in the fall.
- The highest concentration of the substance from the PAH group in all the taken water samples was noted for benzo(b)fluoranthene and benzo(k)fluoranthene – 190 ng/L.
- In the water sample taken from the pumping station in the spring, the concentration of chosen PAH was the lowest.
- In the water samples taken in the fall, the exceeding of norms contained in the Council Directive 98/83/EC of 3 November 1998 on the quality of water intended for

human consumption and Regulation of the Minister of Health of 7 December 2017 on the quality of water intended for human consumption was not noted.

- Concentration of benzo(a)pyrene in the water sample taken from the reservoir in the fall and the water sample taken after final ozonation in the spring was higher than the acceptable concentration contained in the Council Directive 98/83/EC of 3 November 1998 on the quality of water intended for human consumption and Regulation of the Minister of Health of 7 December 2017 on the quality of water intended for human consumption.

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