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Application of UV irradiation for polycyclic aromatic hydrocarbon removal from treated wastewater in water reclamation system

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ABSTRACT

Water is one of the most important chemical compounds for humans and other living organisms. These days the problem of its insufficiency, growing pollution and poor management is crucial all over the world. New regulations proposed by the European Commission (EC) encourage water reuse (e.g., in agriculture) which results in attempts to reclaim it from treated wastewater from municipal wastewater treatment plants. Water obtained this way should correspond with microbiological and chemical requirements. Conventional wastewater treatment processes fail to remove some emerging pollutants such as polycyclic aromatic hydrocarbons (PAH). It is important to choose the right method that will allow to achieve the desired effect. Studies show that the efficiency of PAH removal from wastewater using UV irradiation may vary depending on PAH concentration in wastewater samples prior to the process. The removal of PAH is higher in the wastewater samples with an additional amount of those compounds (up to 84%) than from the samples with no added PAH standard mixture (up to 65%). It was also confirmed that the removal efficiency differs according to the number of rings of the PAH compound. The aim of this research was the assessment of PAH removal effectiveness from treated wastewater with the application of UV irradiation using low-pressure lamps with different exposure time as a part of further research considering the removal of emerging pollutants from treated municipal wastewater in order to obtain reclaimed water. The obtained results were compared with the norms standardized by the Council Directive 98/83/EC of 3 November 1998 on the quality of water intended for human consumption. PAH removal varied from 11% to 62% after 0.5 h of UV irradiation, 19%–100% after 1 h of UV irradiation, 41%–100% after 2 h of UV irradiation and 72%–100% after 12 h of UV irradiation. PAH standardized in the Council Directive 98/83/EC of 3 November 1998 on the quality of water intended for human consumption, that is, benzo(a)pyrene, benzo(b)fluoranthene and benzo(k)fluoranthene were removed after 1 h of UV irradiation.

Keywords: Polycyclic aromatic hydrocarbons; Treated wastewater; UV irradiation; Reclaimed water

1. Introduction

Providing people all over the world with drinking water of suitable quality is crucial. The water environment is highly vulnerable to any changes caused by human activity. Therefore, water quality monitoring is a very important matter [1–3]. However, it is strictly associated with the problem of its insufficiency as the growing pollution and poor management make it harder to fulfill the conditions contained in law regulations concerning water quality. The latest regulations proposed by the European Commission provide encouragement to develop the ways of water reclamation, for example, from treated wastewater from municipal wastewater treatment plants and reuse, for example, in agriculture. Although it is essential that the water obtained in the process complies with the microbiological

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and chemical requirements regulated by law. Unfortunately, it is unavailable to reach it with conventional wastewater treatment processes. Especially when it comes to removing some emerging pollutants like polycyclic aromatic hydrocarbons (PAH) [2,4–7].

Current regulations concerning wastewater discharge to the environment do not specify permissible levels of PAH. However, due to their highly toxic, carcinogenic and mutagenic character, PAH should have been monitored and eliminated from the water environment [8–11].

The matter of sewage treatment still needs to be resolved particularly because of European Union directives that are more rigorous and aiming at constantly developing natural environment protection. It also involves a constant increase of costs connected to water supply and wastewater discharge. Obtaining the wastewater purified to a degree as it is possible to reuse it as reclaimed water would be a perfect solution and it seems like the disinfection process is the answer to that [12]. Choosing the method that would allow to achieve the desired effect is essential. UV radiation combined with other disinfection methods may be effective to dispose of pathogens in water. It may also provide an effective reduction of trace organic chemical compounds [11,13]. It is also recommended as the safest and most economical option in view of the technical, financial and environmental merits compared to chlorination and ozonation [14–16].

However, it is important that wastewater is properly treated before the disinfection for the process to be effective. Reuse of treated wastewater is an appealing option for environmental protection and water resources availability extension. Water reuse practices include green space and crop irrigation, urban uses such as toilet flushing and also industrial applications. The safety of a process depends on the sewage disinfection process reliability which brings great importance for public health protection [14,17,18].

It is still hard to imagine people would be inclined to drink reclaimed water, even if it is pure enough and completely safe. However, there are ways that reclaimed water can help humans not including drinking it directly. The use of reclaimed wastewater might contribute to better water management as if it supplies the water needed for some purposes, at the same time it may free up freshwater somewhere else, for example, for drinking purposes. The East Bay Municipal Utility District in California has an implemented water reclamation project that conserves drinking water, reduces pollution of San Francisco Bay and provides water for irrigation and industrial purposes. Its main direction is to save enough water so it is possible to provide drinking water to 83,000 households [19].

The aim of this research was the assessment of PAH removal effectiveness from treated wastewater with the application of UV irradiation using low-pressure lamps with different exposure time as a part of further research considering the removal of emerging pollutants from treated municipal wastewater in order to obtain reclaimed water pure enough to safely use it again, for example, in agriculture. The obtained results were compared with the norms standardized by the Council Directive 98/83/EC of 3 November 1998 on the quality of water intended for human consumption, as they are very strict and it is a great challenge to reclaim water quality good enough for possible reuse as a drinking water [20].

2. Materials and methods

2.1. Materials

For the research treated wastewater samples from the wastewater treatment plant located in Silesian Voivodeship in Poland were taken. Wastewater treatment plant technological system consists of mechanical and biological treatment methods with the use of activated sludge. Over the period of 2013 to 2014, the treatment plant was modernized and adapted to the current legal and technological regulations. Septic tank sewage collection point is on site. The capacity of the wastewater treatment plant equals 2,505 m^3/d.

Wastewater samples were then exposed to UV light for 0.5, 1, 2 and 12 h respectively in order to determine the optimum contact time [14]. Changes in PAH concentration were determined based on the analysis of these compound's concentration in samples before and after UV irradiation. Low pressure, 10 W, WWF 409-00 flow-through lamp by Heissner Company was used with the wavelength of 254 nm.

2.2. Analytical procedure

For PAH extraction from the wastewater samples, the solid phase extraction (SPE) method was applied, using 500 mg Octadecyl C18 6 mL Bakerbond Extraction Columns and SPE Apparatus. For the purpose of PAH extraction, 0.5 L of wastewater was taken. Isopropyl alcohol was added to the samples. The wastewater was passed through SPE extraction columns preconditioned using 6 mL of methanol and 6 mL of isopropyl alcohol and distilled water mixture in the ratio 85:15. After that, the columns with PAH trapped on sorbent were dried in a vacuum for approximately 30 min. PAH was then eluted from the columns filling using 3 mL × 1 mL of dichloromethane. Extracts were condensed to the volume of 1 mL and analyzed with the use of gas chromatography and mass spectrometer (GC-MS) [21–23]. A standard mixture of 16 PAH by RESTEK company was used and 0.002 mL of extract was injected on the DB-5 column using a microsyringe. Helium was used as a carrier gas. Determination of all analyzed compounds was performed in triplicate. To eliminate the error as a result of sample taking method, the wastewater samples were averaged. These representative samples were put under further procedures. The detection limits varied from 0.3 to 1.8 ng/L for PAH. The recoveries of PAH are shown in Table 1. Analytes identification confirmation relied on GC retention time and qualifier ions [23,24]. Peaks with acceptable ion ratios were integrated to be later quantified. Because of this, the main error impacting the variation of concentration of analyzed compounds in the samples is analytical error [23].

3. Results and discussion

The results of chosen PAH concentrations in wastewater samples and their standard deviations are presented in Table 2.

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

Table 1
PAH concentration in wastewater samples

Compound	Recovery (%)
Naphthalene	52.5 ± 4.7
Acenaphthylene	68 ± 5.6
Acenaphthene	77.5 ± 7.1
Fluorene	55 ± 5.4
Phenanthrene	90 ± 6.4
Anthracene	120 ± 2.8
Fluoranthene	72.5 ± 7.1
Pyrene	52.5 ± 6.0
Benzo(a)anthracene	67.5 ± 7.0
Chrysene	67.5 ± 6.4
Benzo(b)fluoranthene	72.5 ± 4.4
Benzo(k)fluoranthene	72.5 ± 4.2
Benzo(a)pyrene	75 ± 5.8
Indeno(1,2,3,-cd)pyrene	65 ± 8.8
Dibenzo(a,h)anthracene	51 ± 6.0
Benzo(ghi)perylene	54 ± 5.6

- PAH standardized in the Council Directive 98/83/EC of 3 November 1998 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 [20],
- 2-ring PAH: naphthalene,
- 3-ring PAH: acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene,
- 4-ring PAH: fluoranthene, pyrene, benzo(a)anthracene, chrysene,
- 6-ring PAH: dibenzo(a,h)anthracene.

In the treated wastewater sample before UV irradiation, the concentration of benzo(a)pyrene amounted to 174.1 ng/L, which is much more than the acceptable concentration (10 ng/L) standardized in the Council Directive 98/83/EC of 3 November 1998 on the quality of water intended for human consumption. The concentration of the sum of PAH included in the regulations was below the permissible level (100 ng/L). It amounted to 11.8 ng/L but from four compounds in this group only the presence of two was noted, which were benzo(b)fluoranthene (5.9 ng/L) and benzo(k)fluoranthene (also 5.9 ng/L). The concentration of 2-ring naphthalene amounted to 29.7 ng/L. Analyzing the group of 3-ring PAH, the highest concentration was detected in the case of anthracene. It amounted to 379.3 ng/L and it is at the same time the highest concentration noted out of all analyzed compounds. The concentration of other compounds in this group such as acenaphthylene, acenaphthene, fluorene and phenanthrene amounted to 17.7, 5.5, 7.2 and 6.1 ng/L respectively. Analyzing the group of 4-ring PAH the concentrations of fluoranthene and pyrene, amounted to 26.2 and 8.9 ng/L respectively and 6.1 ng/L in the case of both benzo(a) anthracene and chrysene. The presence of 6-ring PAH, that is, dibenzo(a,h)anthracene in the analyzed sample was not detected. Standard deviations varied from 0.14 to 0.89 ng/L.

In the treated wastewater sample after 0.5 h of UV irradiation, the concentration of benzo(a)pyrene amounted to 137.8 ng/L, which is also much more than the acceptable concentration. The concentration of the sum of PAH included in the regulations amounted to 8.0 ng/L. Similar to the previously discussed sample only the presence of benzo(b)fluoranthene (4.0 ng/L) and benzo(k)fluoranthene (also 4.0 ng/L) was noted in the case of this group of PAH. The concentration of 2-ring naphthalene amounted to 20.6 ng/L. In the group of 3-ring PAH, the highest concentration detected was again in the case of anthracene. It amounted to 305.2 ng/L. The concentration of other compounds in this group such as acenaphthylene, acenaphthene, fluorene and phenanthrene amounted to 15.7, 4.9, 5.0 and 4.9 ng/L respectively. In the group of 4-ring PAH the concentrations of fluoranthene and pyrene, amounted to 10.1, 6.8 respectively 4.9 ng/L in the case of both benzo(a)anthracene and chrysene. The presence of dibenzo(a,h)anthracene in the analyzed sample was not detected similarly to the previous sample. Standard deviations varied from 0.07 to 0.57 ng/L.

In the treated wastewater sample after 1 h of UV irradiation, the concentration of benzo(a)pyrene was not detected. Also, none of the four compounds in the group of the sum of PAH included in the regulations was noted. The concentration of naphthalene amounted to 10.3 ng/L. The 3-ring PAH concentrations, that is, acenaphthylene, acenaphthene, fluorene, phenanthrene and anthracene amounted to 7.0, 4.5, 4.7, 3.8 and 3.9 ng/L respectively. Analyzing the group of 4-ring PAH, the concentrations of fluoranthene and pyrene, amounted to 6.2, 5.8 and 2.5 ng/L in the case of both benzo(a)anthracene and chrysene. The presence of dibenzo(a,h)anthracene in the analyzed sample was not detected similarly to the previous samples. Standard deviations varied from 0.02 to 0.51 ng/L.

In the treated wastewater sample after 2 h of UV irradiation, nine out of sixteen analyzed compounds were present. The concentration of naphthalene amounted to 7.5 ng/L. In the case of 3-ring PAH concentrations, that is, acenaphthylene, acenaphthene, fluorene, and anthracene amounted to 6.7, 3.3, 2.8 and 3.1 ng/L respectively. The only compound from this group that was not detected is phenanthrene. Analyzing the group of 4-ring PAH, the concentrations of fluoranthene, pyrene, benzo(a)anthracene and chrysene amounted to 3.7, 4.2, 1.8 and 1.8 ng/L respectively. The presence of dibenzo(a,h)anthracene in the analyzed sample was not detected similarly to the previous samples. Standard deviations varied from 0.10 to 0.45 ng/L.

In the treated wastewater sample after 12 h of UV irradiation, seven out of sixteen analyzed compounds were present. The concentration of naphthalene amounted to 7.0 ng/L. In case of 3-ring PAH concentrations, that is, acenaphthylene, acenaphthene, fluorene, and anthracene amounted to 4.3, 1.5, 1.5 and 2.5 ng/L respectively. Analyzing the group of 4-ring PAH, the concentrations of fluoranthene and pyrene amounted to 2.9 and 1.8 ng/L. The presence of benzo(a)anthracene and chrysene was not detected. Studies show that the efficiency of PAH removal from wastewater using UV rays may vary. It depends on PAH concentration in wastewater samples prior to the irradiation. It was confirmed by the studies of Włodarczyk-Makuła. The removal of PAH was higher in the wastewater samples with an

Table 2
PAH concentration in wastewater samples

Compound	Wastewater		After 0.5 h of UV		After 1 h of UV		After 2 h of UV		After 12 h of UV	
	Concentration (ng/L)	Standard deviation (ng/L)	Concentration (ng/L)	Standard deviation (ng/L)	Concentration (ng/L)	Standard deviation (ng/L)	Concentration (ng/L)	Standard deviation (ng/L)	Concentration (ng/L)	Standard deviation (ng/L)
Naphthalene	29.7	0.42	20.6	0.35	10.3	0.30	7.5	0.31	7.0	0.21
Acenaphthylene	17.7	0.35	15.7	0.28	7.0	0.23	6.7	0.24	4.3	0.06
Acenaphthene	5.5	0.14	4.9	0.07	4.5	0.02	3.3	0.13	1.5	0.27
Fluorene	7.2	0.49	5.0	0.42	4.7	0.37	2.8	0.38	1.5	0.08
Phenanthrene	6.1	0.64	4.9	0.57	3.8	0.51	nd	–	nd	–
Anthracene	379.3	0.42	305.2	0.35	3.9	0.30	3.1	0.31	2.5	0.07
Fluoranthene	26.2	0.28	10.1	0.21	6.2	0.16	3.7	0.17	2.9	0.13
Pyrene	8.9	0.21	6.8	0.14	5,8	0.19	4.2	0.10	1.8	0.20
Benzo(a)anthracene	6.1	0.57	4.9	0.49	2.5	0.44	1.8	0.45	nd	–
Chrysene	6.1	0.49	4.9	0.42	2.5	0.37	1.8	0.38	nd	–
Benzo(b)fluoranthene	5.9	0.34	4.0	0.57	nd	–	nd	–	nd	–
Benzo(k)fluoranthene	5.9	0.34	4.0	0.57	nd	–	nd	–	nd	–
Benzo(a)pyrene	174.1	0.89	137.8	0.42	nd	–	nd	–	nd	–
Indeno(1,2,3,-cd)pyrene	nd[a]	–	nd	–	nd	–	nd	–	nd	–
Dibenzo(a,h)anthracene	nd	–	nd	–	nd	–	nd	–	nd	–
Benzo(ghi)perylene	nd	–	nd	–	nd	–	nd	-	nd	–

[a] nd – not detected

additional amount of those compounds (up to 84%) than from the samples with no added PAH standard mixture (up to 65%). It was also confirmed by the same studies that the removal efficiency differs according to the number of rings of PAH compound [9,13].

In Figs. 1–4, the percent of PAH removal from wastewater samples is shown. The removal of naphthalene amounted to 31% after 0.5 h of UV irradiation, 65% after 1 h of UV irradiation, 75% after 2 h of UV irradiation and 76% after 12 h of UV irradiation in comparison to the compound concentration in wastewater before UV irradiation. The removal of acenaphthylene amounted to 12% after 0.5 h of UV irradiation, 61% after 1 h of UV irradiation, 62% after 2 h of UV irradiation and 76% after 12 h of UV irradiation in comparison to the compound concentration in wastewater before UV irradiation. The removal of acenaphthene amounted to 11% after 0.5 h of UV irradiation, 19% after 1 h of UV irradiation, 41% after 2 h of UV irradiation and 72% after 12 h of UV irradiation in comparison to the compound concentration in wastewater before UV irradiation. The removal of fluorene amounted to 31% after 0.5 h of UV irradiation, 35% after 1 h of UV irradiation, 62% after 2 h of UV irradiation and 79% after 12 h of UV irradiation in comparison to the compound concentration in wastewater before UV irradiation. The removal of phenanthrene amounted to 20% after 0.5 h of UV irradiation, 37% after 1 h of UV irradiation and 100% after 2 h of UV irradiation in comparison to the compound concentration in wastewater before UV irradiation. The removal of anthracene amounted to 20% after 0.5 h of UV irradiation, 99% after 1 h of UV irradiation, 99% after 2 h of UV irradiation and 99% after 12 h of UV irradiation in comparison to the compound concentration in wastewater before UV irradiation. The removal of fluoranthene amounted to 62% after 0.5 h of UV irradiation, 76% after 1 h of UV irradiation, 86% after

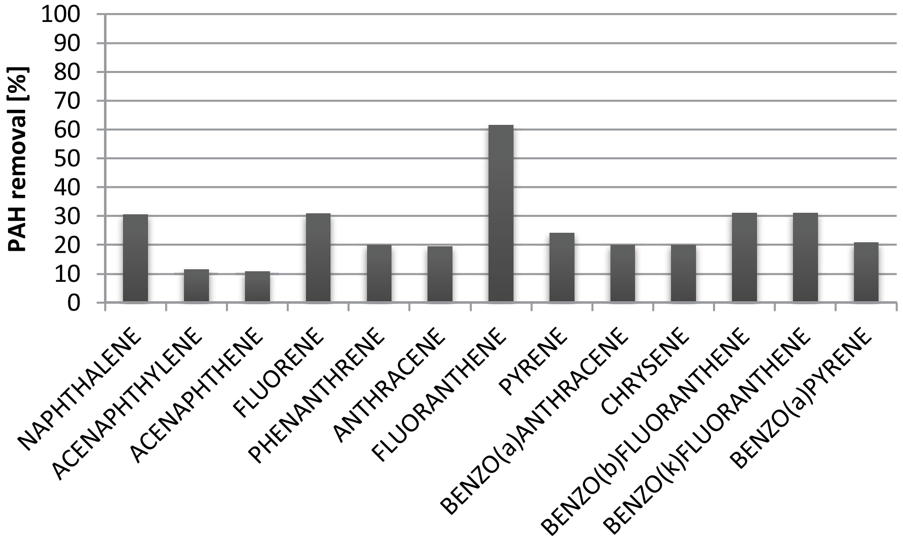

Fig. 1. PAH removal after 0.5 h UV irradiation.

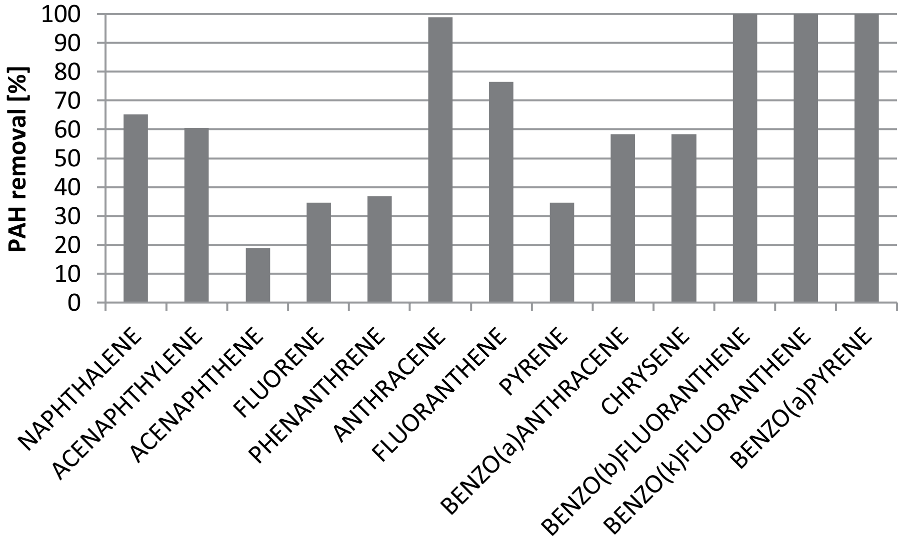

Fig. 2. PAH removal after 1 h UV irradiation.

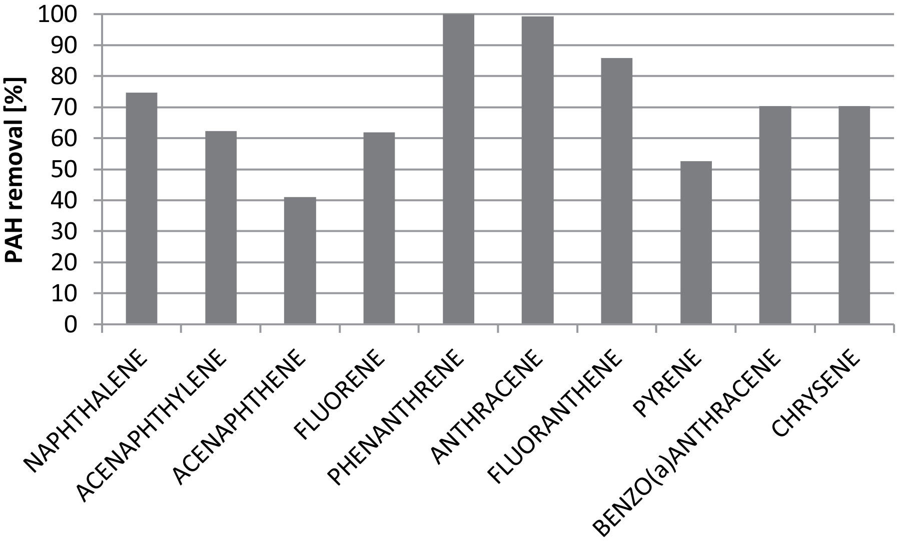

Fig. 3. PAH removal after 2 h UV irradiation.

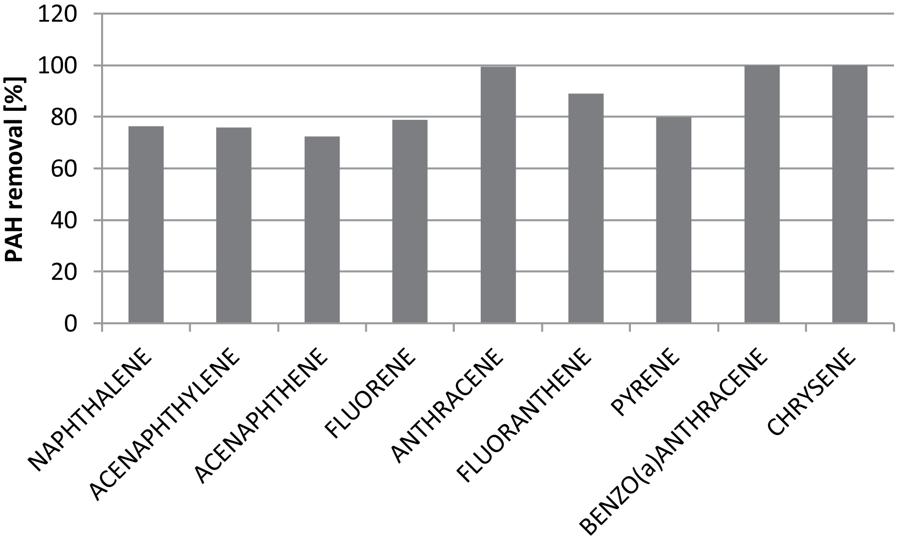

Fig. 4. PAH removal after 12 h UV irradiation.

2 h of UV irradiation and 89% after 12 h of UV irradiation in comparison to the compound concentration in wastewater before UV irradiation. The removal of pyrene amounted to 24% after 0.5 h of UV irradiation, 35% after 1 h of UV irradiation, 53% after 2 h of UV irradiation and 80% after 12 h of UV irradiation in comparison to the compound concentration in wastewater before UV irradiation. The removal of benzo(a) anthracene amounted to 20% after 0.5 h of UV irradiation, 58% after 1 h of UV irradiation, 70% after 2 h of UV irradiation and 100% after 12 h of UV irradiation in comparison to the compound concentration in wastewater before UV irradiation. The removal of chrysene amounted to 20% after 0.5 h of UV irradiation, 58% after 1 h of UV irradiation, 70% after 2 h of UV irradiation and 100% after 12 h of UV irradiation in comparison to the compound concentration in wastewater before UV irradiation. The removal of benzo(b)fluoranthene amounted to 31% after 0.5 h of UV irradiation, 100% after 1 h of UV irradiation in comparison to the compound concentration in wastewater before UV irradiation. The removal of benzo(k)fluoranthene amounted to 31% after 0.5 h of UV irradiation, 100% after 1 h of UV irradiation in comparison to the compound concentration in wastewater before UV irradiation. The removal of benzo(a)pyrene amounted to 21% after 0.5 h of UV irradiation, 100% after 1 h of UV irradiation in comparison to the compound concentration in wastewater before UV irradiation. The removal of PAH after 0.5 h of UV irradiation varied from 11% in case of acenaphthene to 62% in case of fluoranthene. The removal of PAH after 1 h of UV irradiation varied from 19% in case of acenaphthene to 100% in case of benzo(b)fluoranthene, benzo(k)fluoranthene and benzo(a)pyrene. The removal of PAH after 2 h of UV irradiation varied from 41% in case of acenaphthene to 100% in case of phenanthrene. The removal of PAH after 12 h of UV irradiation varied from 72% in case of acenaphthene

to 100% in case of benzo(a)anthracene and chrysene. None of the compounds was totally removed from wastewater after 0.5 h UV exposure. After 1 h of UV rays exposure only 5-ring PAH, that is, benzo(b)fluoranthene, benzo(k)fluoranthene and benzo(a)pyrene were removed. Studies show that the removal rate of heavier compounds is higher than the light ones in case of UV application [25].

4. Conclusions

UV rays exposure time has a significant impact on PAH removal from municipal wastewater. The removal of PAH after 0.5 h of UV irradiation varied from 11% to 62%. After 1 h of UV irradiation, it varied from 19% to 100%. In case of 2 h of UV irradiation, PAH removal varied from 41% to 100%. In case of 12 h of UV irradiation, PAH removal varied from 72% to 100%. The rate of heavier compounds removal is higher than the light ones. All of the compounds standardized in the Council Directive 98/83/EC of 3 November 1998 on the quality of water intended for human consumption were removed after 1 h of UV rays exposure. UV irradiation may be a useful method of removing some emerging pollutants like PAH from treated wastewater and combined with other oxidation methods may be a promising option for the water reclamation process, although further research on the subject is necessary.

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