



Persistence of two-, three- and four-ring of PAHs in sewage sludge deposited in different light conditions

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ABSTRACT

The aim of the investigations was to determine the changes in the concentration of PAHs in sewage sludge stored under various light conditions. The sewage sludge samples were stored under aerobic conditions in dark, in laboratory conditions and exposed to UV rays. The changes in the concentration of PAHs were analysed in three series: in sludge samples taken from treatment plant, in sludge with the addition of a standard PAH mixture and in sewage sludge with the addition both a standard mixture and with added sodium azide (abiotic samples). Changes in 10 PAHs concentration in sewage sludge samples were studied at seven-day intervals for four weeks. The concentration of PAHs was determined using gas chromatograph–mass spectrometry. The losses of 10 hydrocarbons after four-weeks of storing were in the range of 12–99%. The half-life of hydrocarbons was in the range of 5–953 d in sewage sludge taken from treatment plant. In sewage sludge amended with standard mixture, half-life of PAHs was not exceed 137 d.

Keywords: Sewage sludge; PAHs; NaN_3 ; UV rays; Darkness; Half-life; GC–MS

1. Introduction

In Poland about 40% of sewage sludges originating from municipal treatment plants is used in agriculture. Sewage sludges support the structure of soil and enrich it with microcompounds. The Polish legislation demands control of eight heavy metals as well as pathogen organisms only in sewage sludges to be applied in the agriculture [1]. However, the presence of toxic organic micropollutants (PAHs, PCBs and AOX), in sewage sludges should also be taken into account [2–5]. Proposed changes to UE directive demand the control of toxic organic micropollutants in sewage sludges applied in agriculture including

PAHs, PCBs, PCDD/PCDF, DEHP, NPE, AOX and LAS [6]. PAHs are considered as pollutants relatively resistant to the process of decomposition which means that under certain conditions, they may undergo destruction under physical–chemical and biological factors and as a consequence they may form new chemical compounds [7–9]. PAHs present in sewage sludges stored in the environment may also be leached from the soil together with rainfalls and/or they can volatilise into the atmosphere [10]. The mentioned conditions do not occur in the natural environment. The sludges applied in agriculture are exposed to the changing environmental conditions. The level of

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decomposition depends on physical–chemical hydrocarbon properties, environment (water, soil and air) and environmental conditions (humidity, temperature, light and pH) [11–14]. The changes in the concentration of PAHs in sewage sludges stored under various temperatures are presented earlier. The most persistence of PAHs was in temperature 5°C compared to 20 and 35°C [15]. Adsorption onto organic matter particles, biodegradation and, at a lower level, volatilisation is regarded as the processes responsible for the changes in PAHs concentration during composting. It was found that the intensity of the above-mentioned processes depends on the number of rings in the molecule [16]. The processes of PAHs destruction are mainly related to chemical oxidation, volatilisation and photodegradation. Photodegradation is one of the dominant processes of PAHs degradation in water environment. In the process of photolysis, for example, the destruction of benzene rings, PAHs destruction and formation of diols, quinones and aldehydes may occur. The mechanisms pathways of photolysis consist of the absorption of light by hydrocarbons and return to the ground form or being transformed into a radical cation [17,18]. In the literature, there is rather scarce information regarding the mechanism of photodegradation. Furthermore, the described experiments were carried out using pure matrices (water) to which one aromatic compound is usually added, treated as the indicator of the whole group of polyaromatic hydrocarbons. Therefore, it is important to determine whether the susceptibility of PAHs on UV radiation is identical for the matrix containing other organic compounds as well as in the case of adding additional amount of PAHs (as a standard mixture). Stability of PAHs is defined as half-life of their decomposition [11,19]. The available investigations take into account mainly soils or the mixture of soil and sewage sludges. It was found that half-lives of PAHs were in the range of 3–3,111 and of 15–408 d in the mixture of soil with sewage sludge and in soil, respectively [11]. In the earlier investigation, in biotic samples, the half-time of carcinogenic hydrocarbons was in the range of 17–126 d. Half-life of these compounds in abiotic sewage sludges was in the range of 32–2,048 d [19].

The aim of this investigation was to determine the changes of PAHs in sewage sludge stored under various light conditions. The objective of this work was to find out what impact various light conditions had on PAHs persistence in sewage sludges stored under aerobic conditions (with standard mixture of PAHs and without standard mixture of PAHs). Parallely, the changes in concentration of PAHs in abiotic sewage sludge (with sodium azide) were studied (with

standard mixture of PAHs and without standard mixture of PAHs). Under experimental conditions, the half-lives of individual compounds were diversified.

2. Materials and methods

2.1. Materials

In the experiment, sewage sludges originating from a municipal treatment plant were used. The process of digestion of sewage sludges in wastewater treatment plant consists of double-stage fermentation and dewatering. Sludges were primarily analysed for: humidity, pH, alkalinity, acidity and contents of organic compounds. The sludges originating from a municipal treatment plant had a low water content (79%) associated with dewatered sludges formerly biochemically stabilised. The same was found for alkalinity (59 mval/L) and pH 8.5. The organic matter content of 49% proved that the sludge was well digested and stored afterwards in the landfill. The determinations of PAHs were made in sludge samples taken from a treatment plant. The results of these determinations were treated as the initial concentration.

2.2. Experimental procedure

Sludge samples were homogenised by quartering to select a representative sample. Seventy-two samples of 10 g each were prepared, and were put into 200 mL glass flasks. The sodium azide was added to 24 samples to inhibit microbial activity (abiotic samples). A standard mixture of 10 PAH compounds in benzene and dichloromethane in the concentration of 2,000 µg/mL each was added to 48 samples. That standard mixture was added to 22 biotic and to 22 abiotic samples. The standard mixture dose was 10,000 µg/kg dm. The samples without the addition of sodium azide as well as without the addition of the standard mixture were treated as control samples.

All the samples were incubated for four weeks at a temperature of 20°C in the laboratory conditions. The samples were stored under various light conditions. Sixteen sewage sludge samples were stored at darkness (without the access to light). The further 16 sewage sludge samples were kept under the access to natural day light. The remaining sludge samples (16) were stored in the laboratory conditions and exposed to UV rays periodically (4 h/d). Humidity was kept constant at this time. The whole volume of each sludge sample was used to determine PAHs. PAH samples were taken at the beginning of the experiment (the initial concentration) and three times

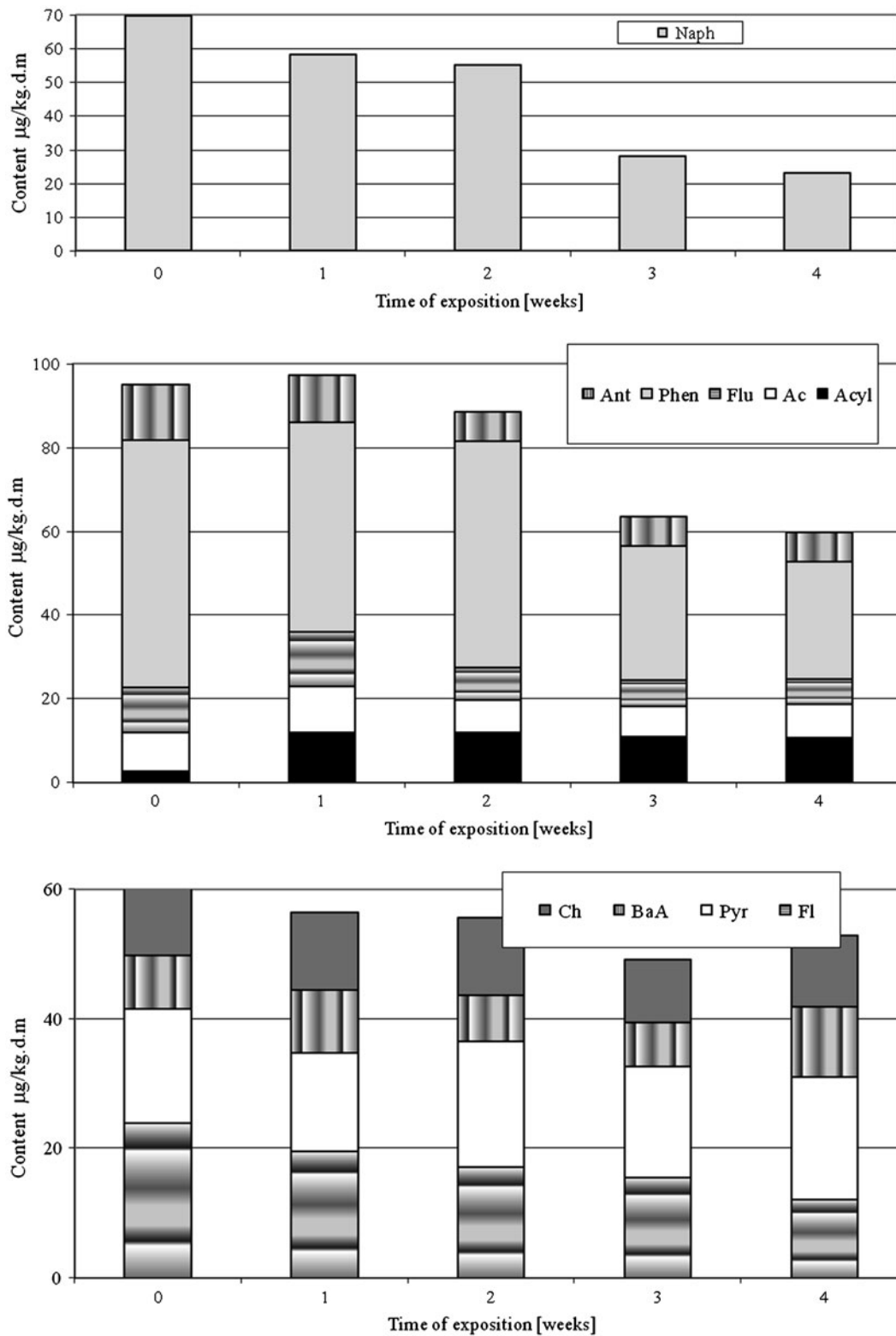


Fig. 1. Changes in the concentration of naphthalene, three- and four-ring of PAHs in sewage sludge (biotic samples) during four weeks of incubation in the dark.

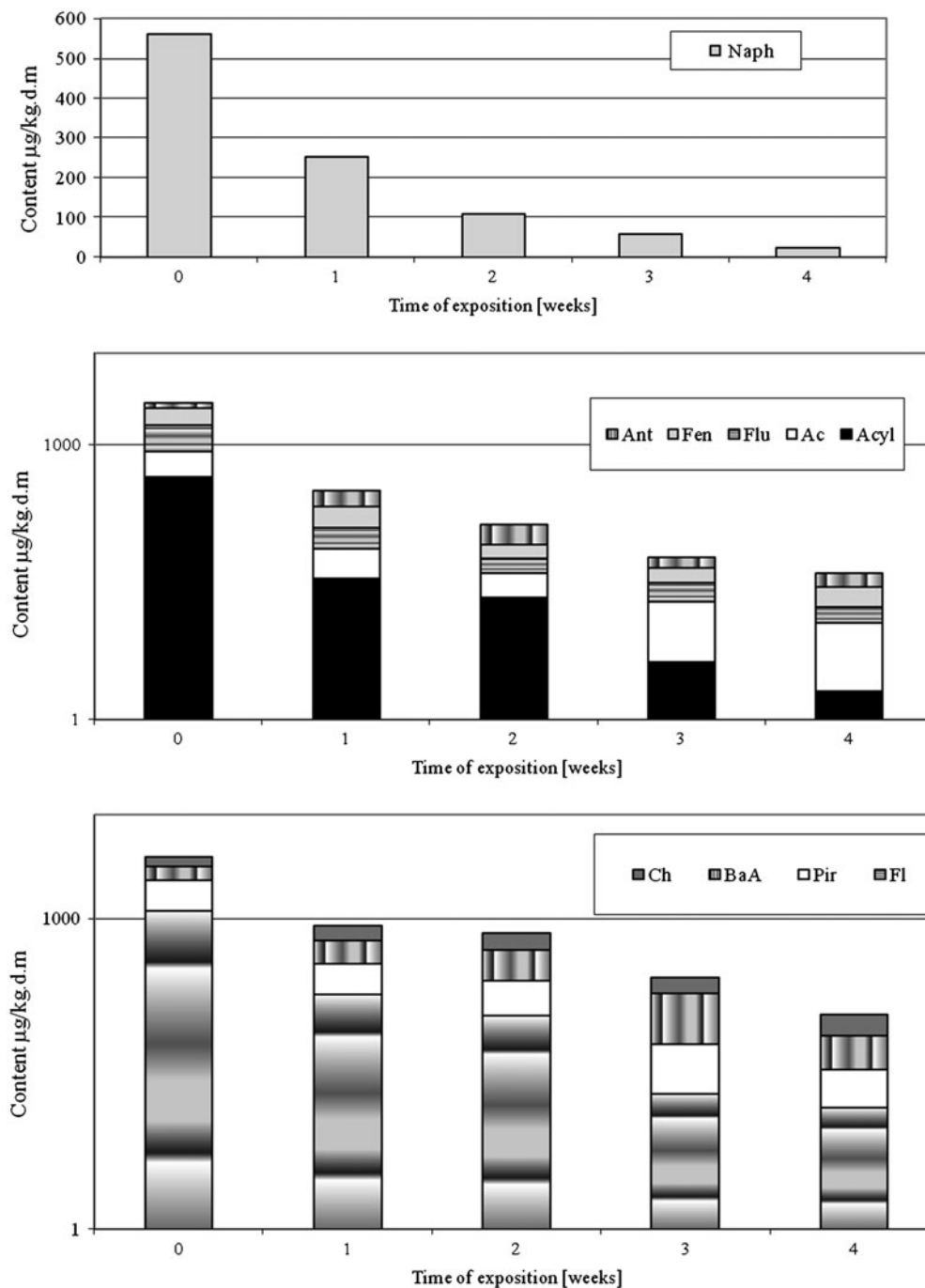


Fig. 2. Changes in the concentration of naphthalene, three- and four-ring of PAHs in sewage sludge amended with standard mixture during four weeks of incubation in darkness.

at one-week interval (after 1–4 weeks). The following investigations were made:

- (1) determination of the changes in PAH concentration in sewage sludge samples taken from municipal treatment plant (biotic-control samples),
- (2) determination of the changes in PAH concentration in sewage sludge samples taken from a municipal treatment plant and supplemented with a standard mixture of these compounds,
- (3) determination of the changes in PAH concentration in sewage sludge samples taken from

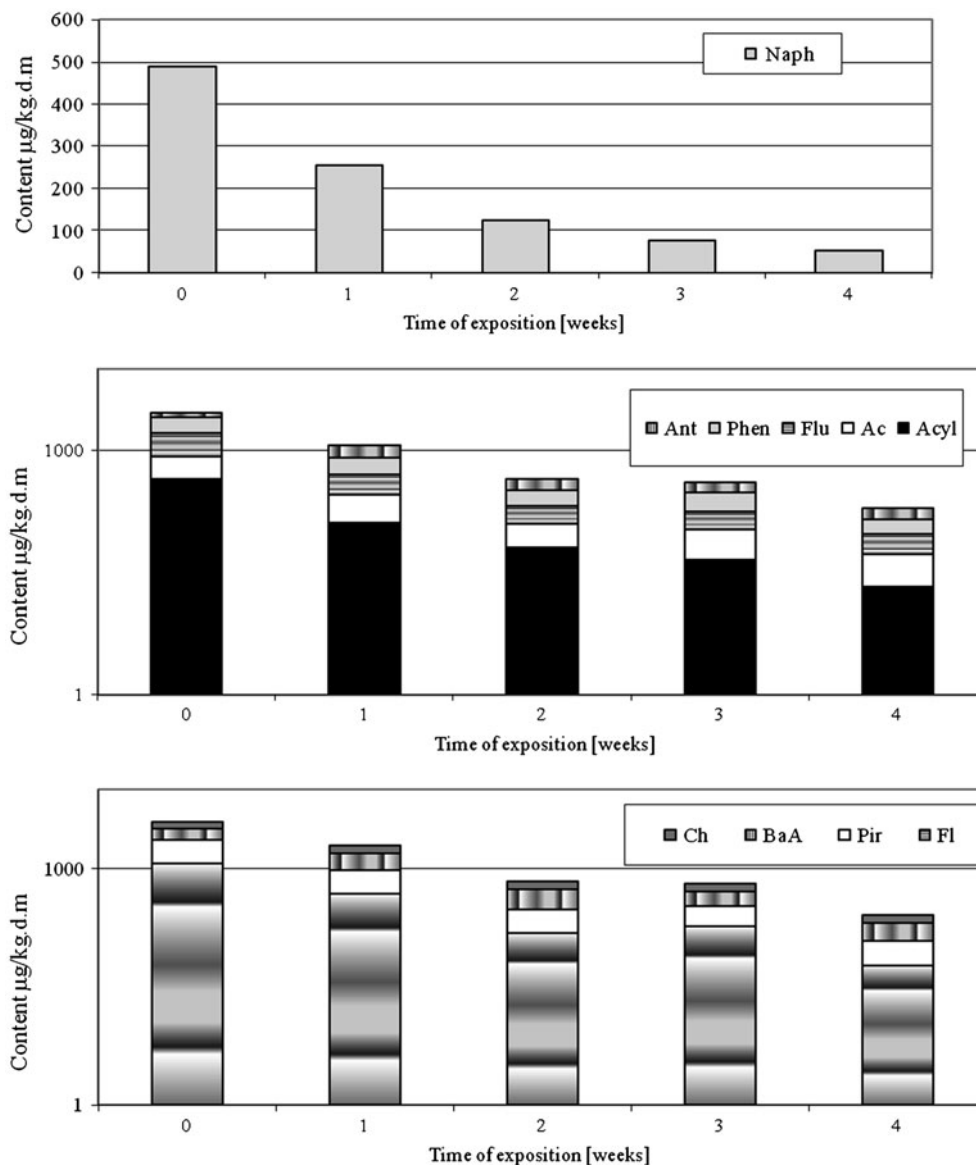


Fig. 3. Changes in the concentration of naphthalene, three- and four-ring of PAHs in sewage sludge amended with standard mixture and sodium azide (abiotic samples) during our weeks of incubation in darkness.

municipal treatment plant and supported with the standard mixture of these compounds as well as sodium azide.

The analyses were made after four weeks determining the concentration of PAHs in sludge samples stored under different light conditions.

2.3. Calculation of PAHs half-life

Assuming that the speed of decomposition of substrate (PAHs) takes place according to the first-order

reaction. Half-life $T_{1/2}$ of hydrocarbons was calculated according to the following equations [10]:

$$T_{1/2} = \frac{\ln 2}{k} \quad (1)$$

and

$$\ln \frac{C_0}{C_t} = k \cdot t \quad (2)$$

where C_0 —initial concentration of PAHs ($\mu\text{g}/\text{kg dm}$); C_t —PAHs concentration in sewage sludge after t days

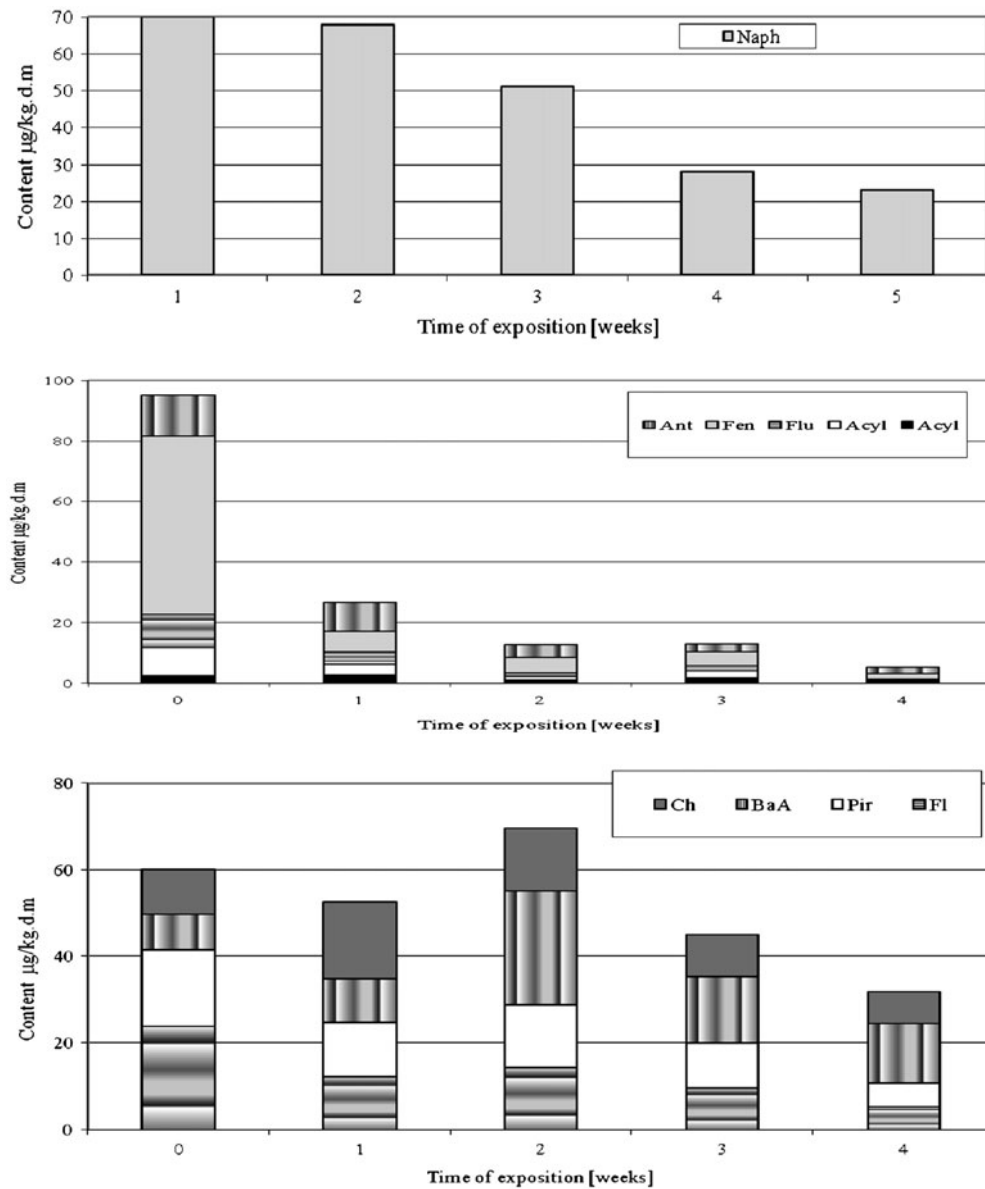


Fig. 4. Changes in the concentration of naphthalene, three- and four-ring of PAHs in sewage sludge (biotic samples) during four weeks of incubation in light conditions.

of sewage sludge incubation ($\mu\text{g}/\text{kg dm}$); t —time of incubation of sewage sludge [d]; k —the reaction rate constant (d^{-1})

2.4. Statistical methods

A Student t -test was used in order to assess the statistical significance of the results. Comparison of affectivities of PAHs degradation in the sewage sludge stored under differed light conditions was calculated according to t -test. The critical value was read from

tables for specified degree of freedom ($n-2$) and at a confidence level of 95%. Theoretical value of decomposition t_d ranged 4,303.

2.5. PAHs analysis

The extraction of the mixture of solvents, cyclohexane/dichloromethane, was used in order to separate organic matrix from sludges (v/v 5/1). The extraction process using ultrasonic batch was carried out. The extracts were separated from sewage sludge samples in

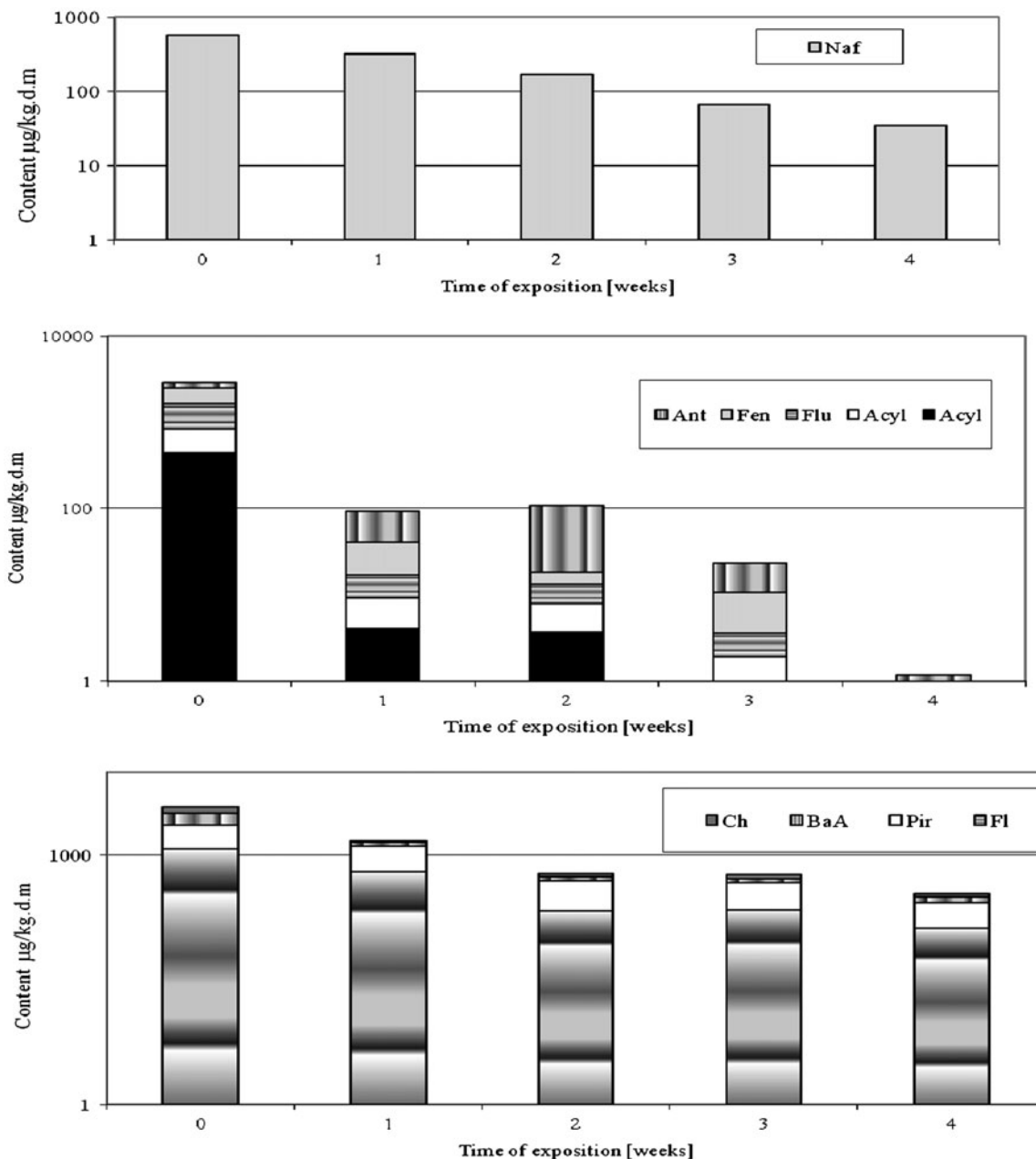


Fig. 5. Changes in the concentration of naphthalene, three- and four-ring of PAHs in sewage sludge amended with standard mixture during four weeks of incubation in light conditions.

the centrifugation process. Prepared extracts were primarily concentrated under the nitrogen stream to a volume of 3 mL. Then extracts were purified on silica gel using SPE chamber under vacuum conditions. Afterwards, the extracts were concentrated to the volume of 1 mL under nitrogen stream and analyzed using gas chromatograph and mass spectrometer (GC–MS) [19,20]. Ten PAHs according to EPA were determined:

(1) two-ring hydrocarbon: naphthalene (Naph),

- (2) three-ring hydrocarbons: acenaphthylene (AcyI), acenaphthene (Ac), fluorene (Flu), phenanthrene (Phen) and anthracene (Ant),
- (3) four-ring hydrocarbons: fluoranthene (Fl), pyrene (Pyr), benzo(a)anthracene (BaA) and chrysene (Ch).

For chromatographic determination of individual PAHs, GC 8000 gas chromatograph Fisons equipped with a mass spectrometric detector MD 800 was used.

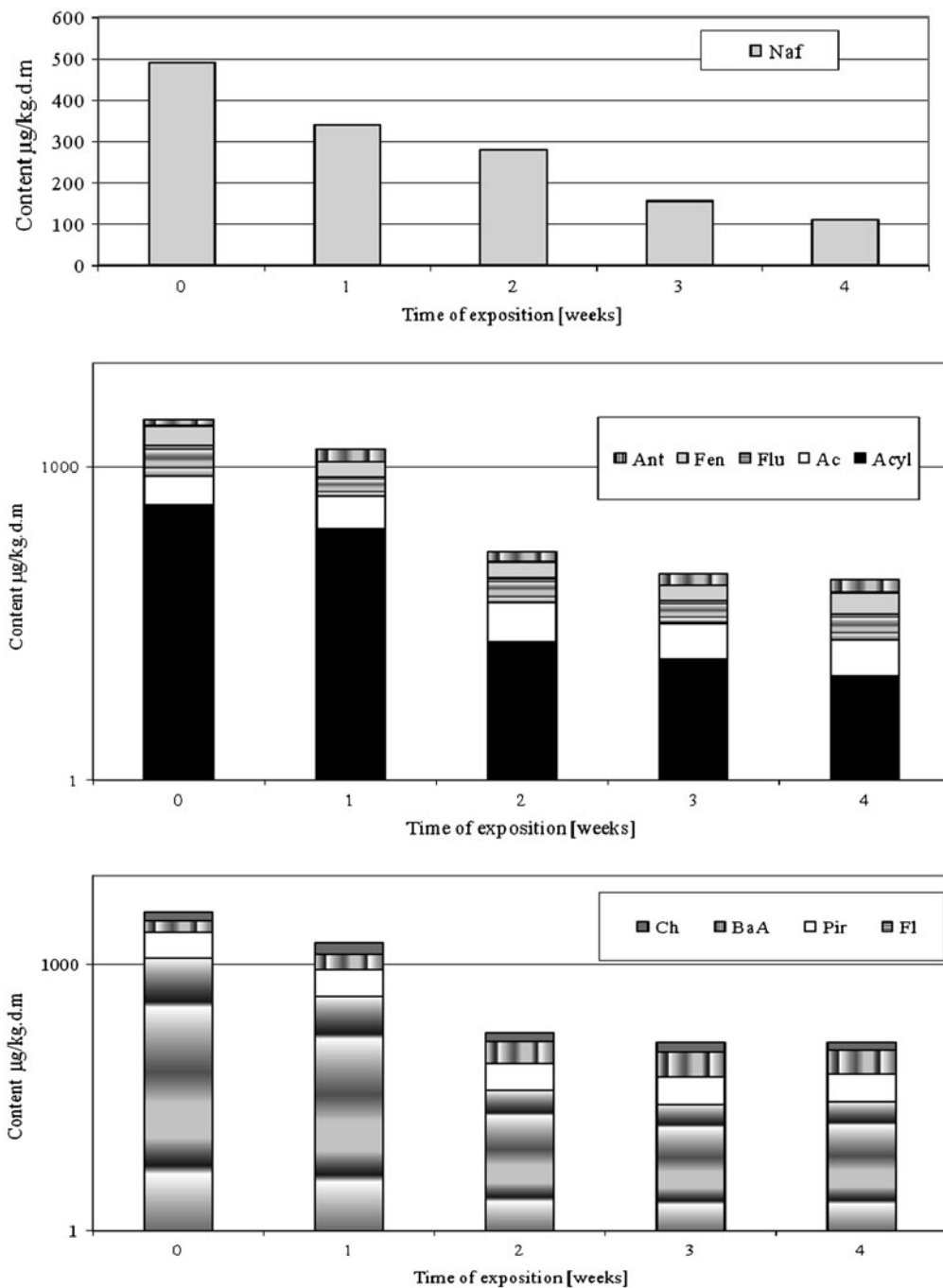


Fig. 6. Changes in the concentration of naphthalene, three- and four-ring of PAHs in sewage sludge amended with standard mixture and sodium azide (abiotic samples) during four weeks of incubation in light conditions.

The parameters of chromatographic analysis were as follow: carrier gas—helium 70 kPa, temperature program 40–120°C (40°C/min) to 280°C (5°C/min) and 280°C for 20 min., volume injection—1 µL, injection system—on column injector, interface temperature—280°C, column—DB-5 (30 m; 0.25 mm; 0.25 µm) and

integration system—MassLab. In order to verify the applied procedure of preparation of sludge samples, the recovery of standard mixture (Accu Standard) 10 PAHs in benzene and dichloromethane in the concentration of 2,000 µg per 1 mL was used. The standard mixture concentration that spiked into the sewage

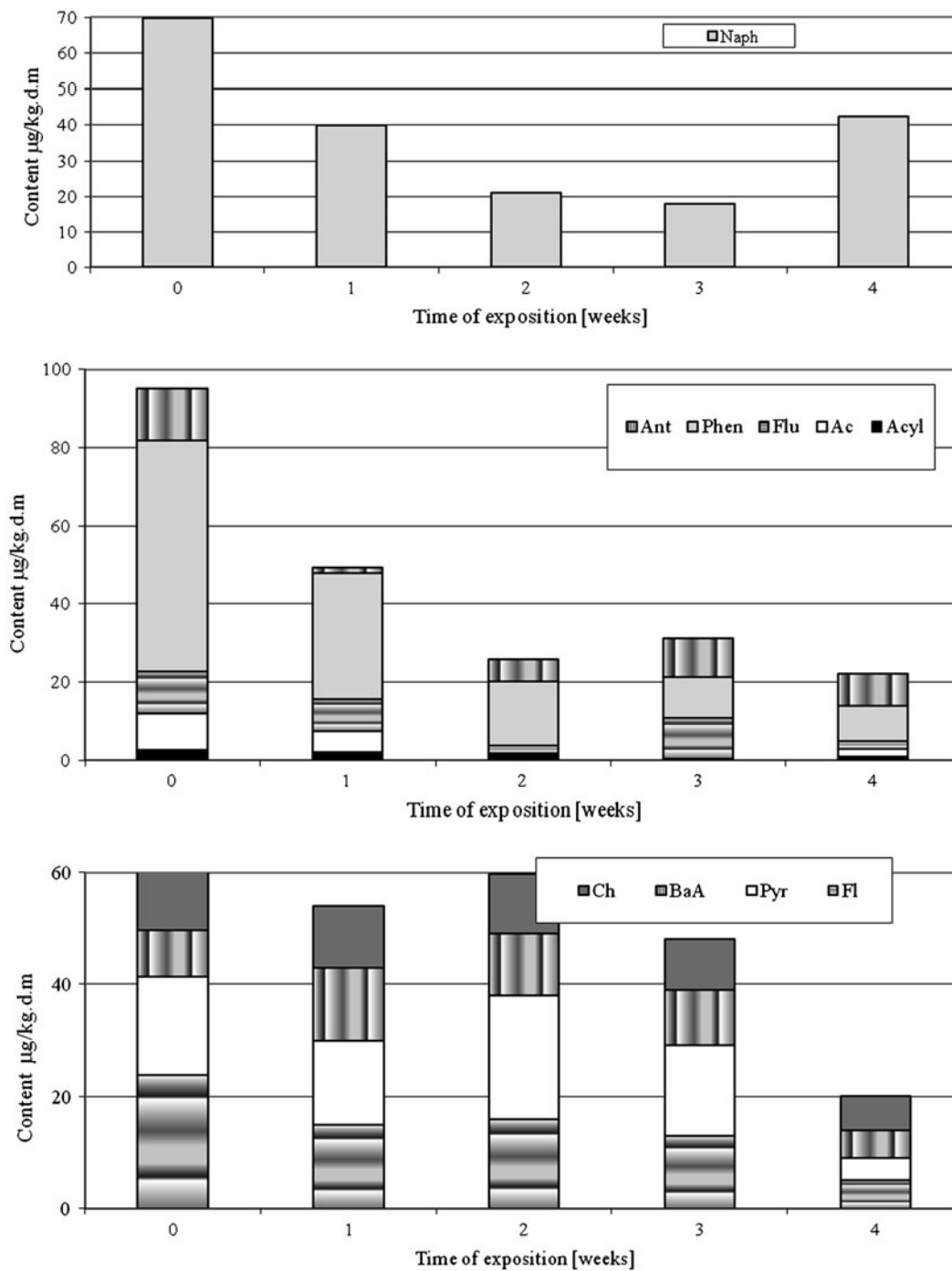


Fig. 7. Changes in the concentration of naphthalene, three- and four-ring of PAHs in sewage sludge (biotic samples) during four weeks of incubation exposed UV rays.

sludge samples was equal to 10 mg/kg dm. The standard mixture was added to samples before adding the solvents and the extraction process. Then, the samples were carefully shaken, extracted and analysed for PAHs according to the procedure described above.

The recoveries of PAHs standard mixture for concentrations in sludges taken from the municipal treatment plant varied from 44 to 92%. The average value was 85% which corresponds to data in the literature [5,20,21].

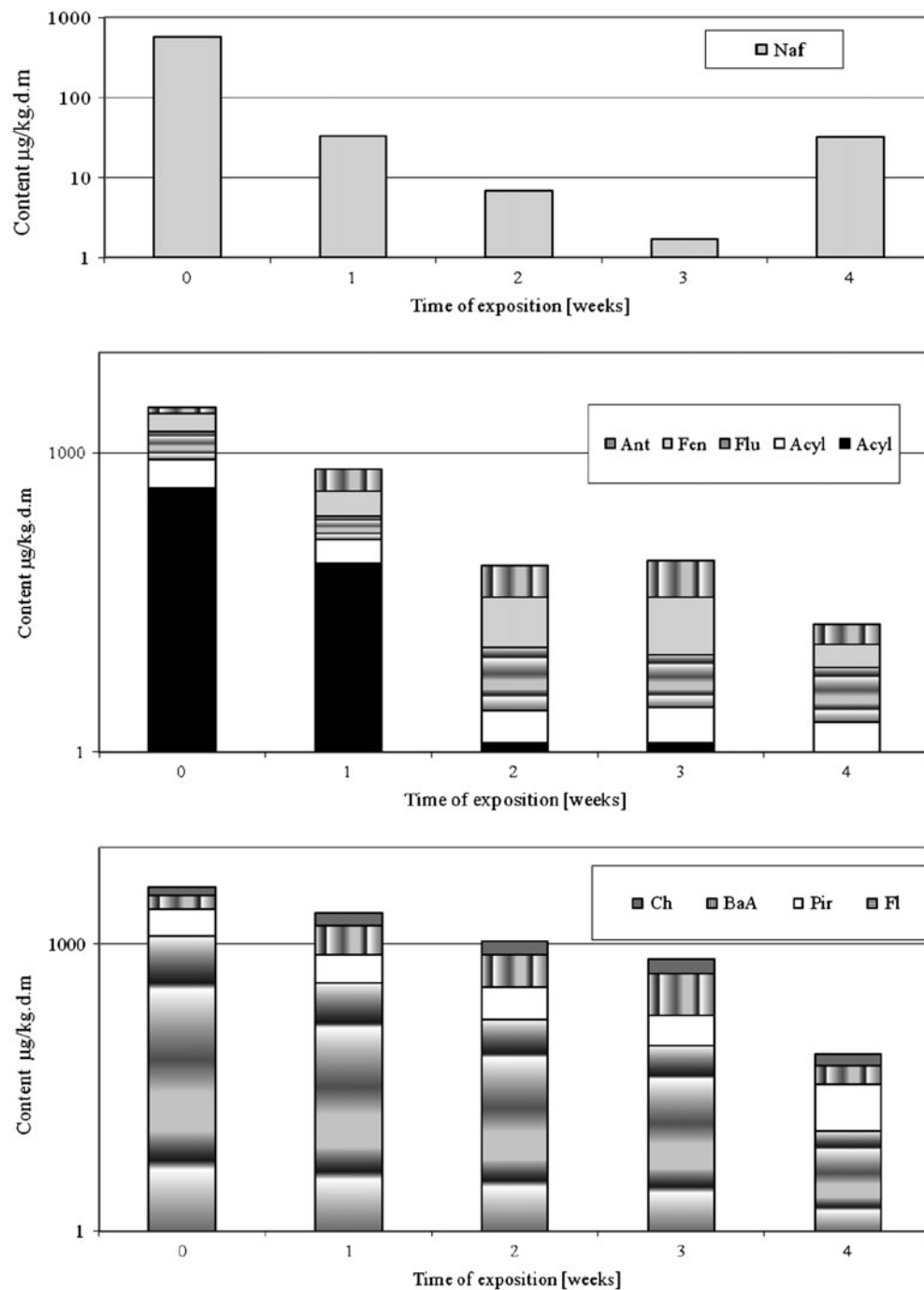


Fig. 8. Changes in the concentration of naphthalene, three- and four-ring of PAHs in sewage sludge amended with standard mixture during four weeks exposed UV rays.

3. Results

In the sludge samples taken from a treatment plant, the initial concentration of PAHs was rather low compared to literary scientific sources and ranged 225 µg/kg of dry matter. The initial concentration of

naphthalene and three-ring hydrocarbons ranged 70–95 µg/kg of dry matter, respectively. The concentration of three-ring compounds was 40% of the total content. The changes in the concentration of naphthalene, three- and four-ring of PAHs in the stored

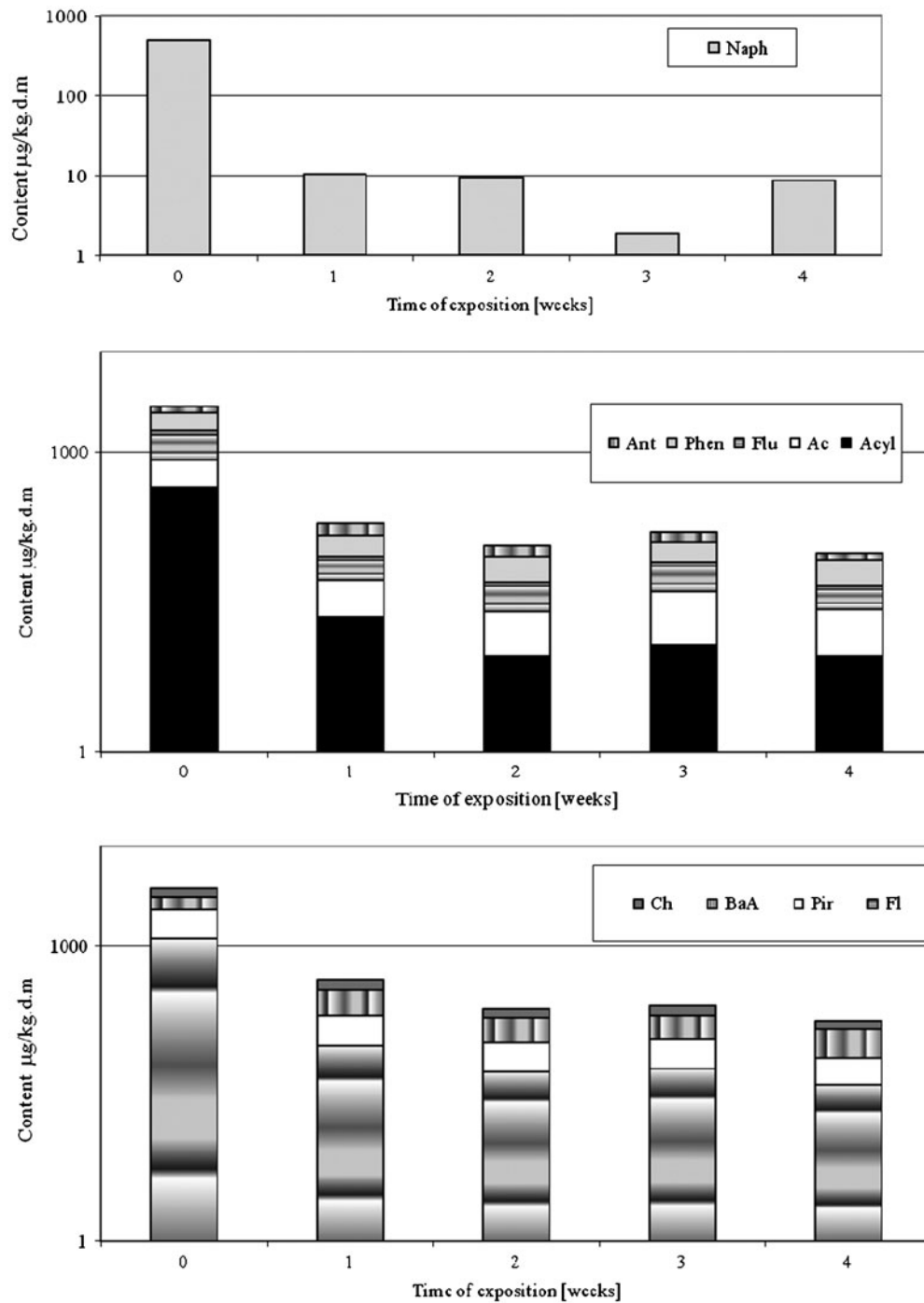


Fig. 9. Changes in the concentration of naphthalene, three- and four-ring of PAHs in sewage sludge amended with standard mixture and sodium azide (abiotic samples) during four weeks exposed UV rays.

sewage sludge during four weeks of incubation in the dark are given in Fig. 1.

In sludge samples incubated in dark, after four weeks of the experiment, the total contents of 10 PAHs was lower than the initial concentration. The

content of these compounds ranged 23 and 60 µg/kg of dry matter. In the sewage sludge stored in the dark, losses of PAHs ranged 40% of the total content, but the most persistence were the four-ring of hydrocarbons. A decrease in these compounds did not exceed

12%. The highest losses were for naphthalene since they are the most volatile compounds. In Fig. 2, changes in the concentration of PAHs in the sewage sludge amended with standard mixture are given.

The concentration of two rings, three rings and four rings of hydrocarbons in sewage sludge spiked with standard mixture reached 559, 2,890 and 3,880 $\mu\text{g}/\text{kg dm}$, respectively. The dynamics of concentration changes of individual hydrocarbon in these samples were different than changes in the concentration of hydrocarbons in control samples. The gradually lower concentration of the 10 studied PAHs was determined during incubation in darkness. The highest losses of hydrocarbons were in the first week of incubation. In the initial step of the investigation (after four weeks), the total PAHs concentration in these sludges was lower than in the initial ones of 98%. The losses of naphthalene, three rings of hydrocarbons and four rings of PAHs were similar and were in the range of 96–99%. After a four-week incubation of samples in sludge, the samples were amended with standard mixture and the average concentration of 10 PAHs was 181 $\mu\text{g}/\text{kg dm}$.

The changes in the concentration of PAHs in the abiotic sewage sludge spiked with standard mixture are presented in Fig. 3. In abiotic samples, the loss of three rings and four rings of PAHs reached 93% and was lower than the loss of these compounds in biotic samples (97–99%).

A Student *t*-test was used in order to assess the statistically significant differences between the initial and final PAH concentrations as well as in order to estimate the statistically significant presence of microorganisms in sludges after 90 d of incubation.

It was estimated that changes in naphthalene, three- and four-ring hydrocarbons concentrations between the initial and final concentration were signif-

icant in the all sewage sludges. The statistical analysis shows that there are significant differences in sludges augmented with the standard mixture among the concentration of hydrocarbons in the both sludges (without and with sodium azide) during deposition of sewage sludges. In Fig. 4, the changes in the concentration of 10 hydrocarbons in sewage sludge stored under light conditions are presented.

The loss of naphthalene was similar to the sewage sludge incubated in dark and did not exceed 67%. In the sludge samples stored under light conditions, a decrease in three rings of PAHs concentration was the highest and ranged 94%. The total contents of 10 PAHs were lower than the initial concentration of 73%. The most persistent were the four rings of hydrocarbons. The changes in the concentration of PAHs in sewage sludge (biotic samples) amended with standard mixture incubated in light condition are given in Fig. 5.

In Fig. 6, the changes in the concentration of 10 hydrocarbons in sewage sludge that amended both standard mixture and sodium azide stored under light conditions are presented.

The average PAHs' initial concentration in sewage sludges biotic and abiotic (with sodium azide) supported with the standard mixture was 7,329 and 7,205 $\mu\text{g}/\text{kg dm}$, respectively. The gradually lower concentration of the 10 studied PAHs was determined during incubation of the samples under light conditions. In the final step of the investigation (after four weeks), the total PAHs concentration in the sewage sludge was lower than in the initial ones of 95% and did not depend on the presence of microorganisms. The highest loss of three-ring of hydrocarbons was noticed (97–99%). At the end of experiment, the total of 10 PAHs concentration did not exceed 400 $\mu\text{g}/\text{kg dm}$. After four weeks of the experiment, in the sewage

Table 1
Removal of PAHs in sewage sludge (%)

Samples	Light conditions	PAHs			
		two ring of PAHs	three ring of PAHs	four ring of PAHs	Total of six PAHs
Sewage sludge—biotic samples (control)	Darkness	67	37	12	40
	Light	67	94	46	73
	UV	82	77	67	76
Sewage sludge—biotic samples with standard mixture of PAHs	Darkness	96	99	97	98
	Light	94	99	91	95
	UV	96	99	99	99
Sewage sludge with both standard mixture of PAHs and sodium azide	Darkness	89	93	93	93
	Light	77	97	97	95
	UV	98	97	96	96

Table 2

Half-life of PAHs, in biotic and abiotic sewage sludges stored in different light conditions (d)

PAH	Condition	Sewage sludge	Time of stored of sewage sludge (weeks)			
			1	2	3	4
Naphthalene	Darkness	1	55	11	129	46
		2	31	62	19	18
		3	13	32	23	23
	Light	1	476	953	6	9
		2	19	37	26	19
		3	28	56	53	36
	UV	1	19	26	31	105
		2	4	7	7	7
		3	5	5	8	7
Acenaphthylene	Darkness	1	–	–	–	–
		2	4	8	10	9
		3	8	17	16	18
	Light	1	–	–	34	126
		2	3	4	6	6
		3	19	38	10	12
	UV	1	69	138	45	15
		2	6	12	5	7
		3	3	7	8	11
Acenaphthene	Darkness	1	–	–	152	169
		2	5	9	10	14
		3	11	22	17	22
	Light	1	11	22	16	30
		2	2	5	7	7
		3	26	52	12	13
	UV	1	19	38	12	12
		2	5	12	5	7
		3	4	8	10	16
Fluorene	Darkness	1	–	–	98	74
		2	4	7	8	10
		3	8	16	14	17
	Light	1	11	22	13	22
		2	2	5	6	7
		3	10	20	10	11
	UV	1	39	78	17	146
		2	5	10	7	9
		3	3	7	9	14
Phenanthrene	Darkness	1	63	125	352	65
		2	5	11	10	11
		3	10	21	17	21
	Light	1	5	9	13	17
		2	3	6	6	9
		3	11	22	10	11
	UV	1	17	33	24	24
		2	7	14	9	12
		3	4	8	10	14
Anthracene	Darkness	1	62	124	57	67
		2	7	15	15	12
		3	44	89	25	32
	Light	1	30	60	27	24
		2	5	10	21	12
		3	69	137	13	14

(Continued)

Table 2 (Continued)

PAH	Condition	Sewage sludge	Time of stored of sewage sludge (weeks)			
			1	2	3	4
Fluoranthene	UV	1	5	10	36	142
		2	27	54	13	19
		3	5	10	12	16
	Darkness	1	54	109	104	97
		2	6	11	13	10
		3	12	23	15	22
	Light	1	16	31	62	46
		2	17	34	18	24
		3	10	21	9	11
Pyrene	UV	1	22	44	77	68
		2	9	18	15	16
		3	4	8	10	14
	Darkness	1	–	–	–	–
		2	6	11	15	13
		3	12	24	16	21
	Light	1	27	54	155	81
		2	20	40	23	28
		3	11	22	9	11
Benzo(a)anthracene	UV	1	86	171	136	464
		2	10	19	18	17
		3	4	9	10	14
	Darkness	1	–	–	197	263
		2	8	16	25	22
		3	29	58	25	27
	Light	1	–	–	–	–
		2	5	11	12	16
		3	15	30	12	15
Chrysene	UV	1	–	–	–	–
		2	105	209	42	41
		3	6	12	13	18
	Darkness	1	–	–	–	–
		2	10	20	27	19
		3	17	34	20	407
	Light	1	–	–	–	590
		2	5	10	11	16
		3	24	48	10	16
UV	1	–	–	–	308	
	2	37	75	37	32	
	3	5	11	12	16	

1—biotic samples, 2—biotic samples with standard mixture of PAHs, 3—abiotic samples with standard mixture of PAHs.

sludge samples exposed to UV rays, the contents of two- and three-ring compounds was lower than initial concentration of 46 and 99%. The highest loss was typical for naphthalene and three-ring compounds due to the fact that the process of volatilisation occurred. After four weeks of the experiment, in the sludge samples exposed to UV rays, the content of 10 PAHs was lower than the initial concentration of 75 and 98%. Changes in the concentration of naphthalene, three and four-ring of PAHs during four weeks of incubation exposed to UV rays are given in Figs. 7–9.

In sewage sludge exposed to UV rays, fluctuation of PAHs concentration was observed. That may have been due to the release of PAHs from cells of microorganisms or the complex of organic associations during exposure to UV rays [17,18]. The studies proved that the process of degradation of PAHs takes place much more difficult in sewage sludge under UV rays conditions. Therefore, it could be concluded that PAHs may be kept not decomposed in the environment. That is very important in agricultural application of sewage sludge.

In the sludge samples stored under different light conditions, the gradual lower concentrations of naphthalene and three-ring of PAHs were observed. The changes of concentration of four-rings of PAHs were negligible in sewage sludge stored in the dark and under the access of light. The losses of these compounds did not exceed 67%.

The statistical analysis shows that there are no significant differences in sludges augmented with the standard mixture among the concentration of hydrocarbons in both the sludges (without and with added sodium azide) while depositing sewage sludges. There is a significant difference between the total contents of 10 PAHs before and after incubation. The addition of sodium azide had a significant impact on the changes in two and three rings of PAHs concentrations. Therefore, statistical analysis shows the importance of abiotic processes in the loss of PAHs. The differences between the concentrations of PAHs in biotic sewage sludge and the concentration of PAHs in abiotic sewage sludges did not significantly. This indicates the relatively low importance of biological effects in the removal of hydrocarbons. The addition of standard mixture had significant impact on changes of PAHs concentrations in biotic samples.

Percentages of removal of hydrocarbons changes grouped according to ring numbers are presented in Table 1.

In Table 2, the half-life in sewage sludge in different light conditions of investigated hydrocarbons is presented.

The half-life of hydrocarbons was different. The half-life was higher in sewage sludge taken from treatment plant (biotic samples) compared to sewage sludge amended with the standard mixture. The half-time of hydrocarbons was in the range of 5–953 d in sewage sludge. Four rings of PAHs were the most persistent hydrocarbons. The most persistent were the hydrocarbons in sewage sludge stored without light conditions. For example, in the dark, the half-life of phenanthrene and benzo(a)anthracene were 352 and 263, respectively. The half-life of acenaphthylene, acenaphthene, fluorene, pyrene and benzo(a)anthracene were not calculated because the concentration of these compounds were at the same level. The half-life of hydrocarbons content in sewage sludge amended with the standard mixture was lower in biotic samples than in abiotic samples for samples stored in dark and daily light conditions. In the samples exposed to UV rays, the half-life of hydrocarbons was lower in abiotic samples than in biotic samples (in sewage sludge amended with the standard mixture). The half-life of hydrocarbons was in the range of 3–137 d in sewage sludge amended with the standard mixture. Thus, it is

suggested that investigations into the dynamics of changes of PAH concentration in sludges should be carried out without any additional amount of hydrocarbons. The results are similar to the scientific literary sources concerning the behaviour of PAHs in soil and in the sewage sludge during the composting process. It is stated that the stability of individual hydrocarbons could vary and the dynamics of the concentration changes during incubation of the analysed materials could be irregular. Irregular changes in the concentration of PAHs in digestion process (fermentation process, composting process) and in deposited sewage sludge were observed [11,21–25].

4. Conclusions

On the basis of the conducted investigation (under certain conditions) and obtained results, it can be concluded that:

- (1) Significant differences between the initial concentrations and the final concentrations for naphthalene and three- and four-ring of PAHs in sewage sludge were observed.
- (2) The dynamics of hydrocarbons changes in sewage sludges supplemented with the standard mixture was similar to the one observed in the sludges both with and without sodium azide (in biotic and in abiotic samples).
- (3) The most persistent of studied hydrocarbons occurred in the sewage sludge originating from the treatment plant stored without light access.
- (4) The half-life was the highest for hydrocarbons in sewage sludge taken from the treatment plant (in samples without standard mixture of PAHs).
- (5) The results of the investigation regarding sewage sludges indicated that under UV rays, some fluctuations in the concentrations of PAHs were found.

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