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INFLUENCE OF FLY ASH ON CHANGES OF POLYCYCLIC AROMATIC HYDROCARBONS CONTENT IN COMPOSTED SEWAGE SLUDGE

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WPŁYW PYŁU LOTNEGO NA ZMIANĘ ZAWARTOŚCI WIELOPIERŚCIENIOWYCH WĘGLOWODORÓW AROMATYCZNYCH W KOMPOSTOWANYM OSADZIE ŚCIEKOWYM

W pracy oceniano zmianę zawartości wielopierścieniowych węglowodorów aromatycznych (WWA) podczas kompostowania osadów ściekowych stabilizowanych pyłami z elektrociepłowni. Oznaczona zawartość sumy wielopierścieniowych węglowodorów aromatycznych w osadzie użytym do kompostowania wynosiła 10385 μg/kg (±830). W pyle stwierdzono jedynie obecność trzech WWA (fenantrenu – 0,9 μg/kg, antracenu – 1,9 μg/kg oraz chryzenu – 2,7 μg/kg). Dodatek pyłu do kompostowanego osadu ściekowego wywarł, w zależności od jego udziału, zróżnicowany wpływ na degradację WWA. Stosunkowo najlepszy stopień rozkładu (66,3%) stwierdzono przy udziale pyłu w ilości 20% (udział wagowy). W kompostowanym osadzie ściekowym oraz osadzie z 30% dodatkiem pyłu stwierdzono obniżenie zawartości WWA odpowiednio o 38 i 32,4%. Największym zakresem mineralizacji charakteryzowały się 3-pierścieniowe WWA. Oznaczone czasy połowicznego rozkładu wszystkich badanych związków w zależności od węglowodoru wahały się od 59 do 1164 dni.

Summary

In the present paper changes of polycyclic aromatic hydrocarbons (PAHs) content were evaluated during composting of sewage sludge stabilized with coal fly ash. The content of PAHs in sewage sludge used for composting was 10385 μg/kg (±830). In fly ash only three PAHs were determined (phenanthrene – 0.9 μg/kg, anthracene – 1.9 μg/kg and chrysene – 2.7 μg/kg). Addition of fly ash to composted sewage sludge had various – dependent on its share – effect on PAHs mineralization. Relatively best degradation (66.3%) was noted when sewage sludge was mixed with fly ash in amount 20% (w/w). In composted sewage sludge and sludge with 30% addition of fly ash a decrease of PAH content was also observed (38 and 32.4% respectively). Relatively "best" mineralization was noted for 3-ring PAHs. Estimated half-lives of all investigated compounds depended on individual PAHs properties and ranged from 59 to 1164 days.

INTRODUCTION

Sewage sludge is a valuable source of nutrients for plants. It also improves physical, chemical and biological properties of soil. Numerous articles have shown that sewage sludge – beside heavy metals and morbid microorganisms [4, 7] can also be loaded with persistent organic pollutants (POPs) [3, 7, 16, 19, 26, 40]. For this reason environmental utilization of sewage sludge in agriculture create potential danger to human health. Contaminants introduced with sewage sludge into the soil, may last in it for many years [1, 2]. Intensity of POPs degradation in sewage sludge-amended soils is clearly dependent on applied sewage sludge dose, content of pollutants in it and degree of homogenization of sludge with soil [2, 9, 17, 23, 34–36].

One of the methods of improving sewage sludge quality is composting. It results in stabilization of organic matter present in sewage sludge and improvement of hygienic properties of sewage sludge itself [11, 25]. Some researches also indicate that composting may lead to the degradation of many organic pollutants [8, 9, 18, 22, 24, 29, 37]. During composting specific conditions, related to the increase of microbiological activity and high temperature, take place and for that reason biochemical reactions are more intense than in soils. These conditions favor growth of microorganisms capable of mineralization of this type of contaminants [8, 29]. However, high organic matter content limits pollutants degradation as a result of sequestration or forming of bound-residue. This way, they become not-available (or very hard available) to microorganisms.

Fly ash from power plants is a waste material created during incineration of hard coal. It contains CaO, MgO, SiO_2 and Fe-Mn oxides. High pH of this waste (up to 12) makes it a good additional stabilizer of sewage sludge, reducing heavy metals mobility and pathogenic microorganisms activity [38, 41, 43]. Moreover, in many papers [14, 15, 39] positive influence of fly ashes on some soil properties has been showed.

In this paper, changes of PAHs content during composting of sewage sludge stabilized with coal fly ash were evaluated. Research was focused on effect of fly ash on disappearance of PAHs, as well as intensity of this process.

MATERIALS AND METHODS

Composting and samples preparation

Composting of sewage sludge was carried out in plastic pots (100 dm^3 each). Sewage sludge (100%) and sewage sludge with addition of 20 and 30% of fly ash (from combustion of hard coal) was placed in them. Sewage sludge and fly ash doses were calculated taking into consideration their dry mass. Composting process was conducted in a closed room with limited access of sunlight. Humidity was kept at constant level (55–60%). Samples for the analyses were taken at the beginning of experiment, after 7 days and later in 14 day periods. Composting was conducted for 231 days. In Table 1 some of physicochemical properties of composted materials are presented and in Figure 1 – shaping of temperature during described process is shown.

Table 1. The physico-chemical properties of composted materials

Properties	Sewage sludge	Fly ash
H_h [cmol(+)/kg]	47.3	–
CEC [cmol(+)/kg]	500.4	98.8
TEB [cmol(+)/kg]	547.7	–
BS [%]	99.4	–
pH in KCl	6.2	9.8
N_t [%]	1.40	0.2
TOC [g/kg]	19.9	2.58
Σ16 PAHs [μg/kg]	10385	5.4

H_h – hydrolytic acidity; CEC – the cation exchange capacity; TEB – the total of the exchangeable bases; BS –the degree of the base saturation; N_t – the total amount of nitrogen; TOC – total organic carbon content

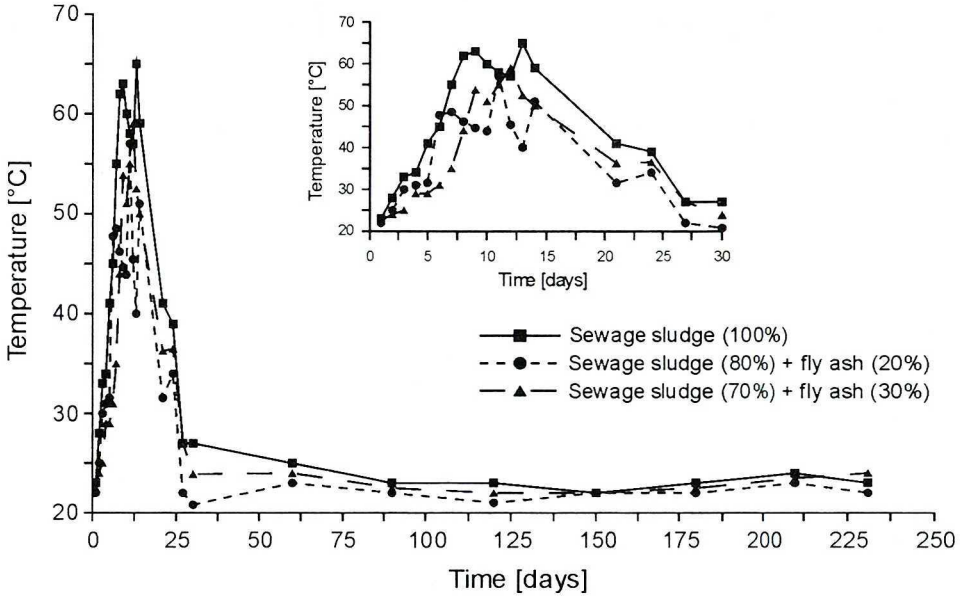

Fig. 1. Temperature during composting process

PAHs analysis

The samples (10 g) were extracted in an ultrasonic bath (Sonic-3, Polsonic, Poland) with two batches of dichloromethane (2 x 40 cm^3). The extract was centrifuged and evaporated to dryness. The residue was then dissolved in a 4 cm^3 mixture of acetonitrile : water (1:1 v/v) and purified by solid phase extraction (SPE) using C_{18} Octadecyl columns (JT Baker-Mallinckrodt, Germany) [31, 32]. PAH was determined using the HPLC-UV method. A Spherisorb-PAH (Schambeck SFD GmbH, Germany) was used for PAH separation.

The mobile phase (acetonitrile : water, 82:18, v/v) flow was set to 1 cm^3/min (in isocratic condition). Detection was carried out at 254 nm. The column was installed in a thermostated oven at 31°C (LCO 101, ECOM, Czech Republic) [6, 31, 32].

All reported concentration values of PAHs are expressed on a dry-wt basis of samples (determined by drying the samples for 24 h at 105°C) and are the average of triplicate extraction.

Data analysis

The rate of individual PAHs losses was determined on the basis of the kinetic equation of the pseudo-first order [27, 33]:

$$\ln \frac{C_0}{C_t} = {}^{*}k \cdot t \tag{1}$$

where: *k – apparent constant reaction rate of the pseudo first order (1/day),
t – time (days),
C_o – initial PAHs content in soil,
C_t – PAH content in soil after time t.

Half-life ($T_{1/2}$) was determined from equation (1) [26]:

$$T_{\frac{1}{2}} = \frac{\ln 2}{{}^{*}k} \tag{2}$$

Statistical significant differences between the results were evaluated on the basis of standard deviation determinations and analysis of variance method (ANOVA, 95% confidence intervals test). Pearson correlation coefficient was calculated. Significance was set at $P \geq 0.05$.

The relationships between content of polycyclic aromatic hydrocarbons and the physico-chemical properties were determined by correlation analysis with Statistica 6.0. Significance was set at $*P \leq 0.05$ and $**P \leq 0.001$. Statistically significant differences between the results were evaluated on the basis of standard deviation determinations and analysis of variance method (ANOVA). Data with normal distribution were analyzed by the t-test for independent samples ($P \leq 0.05$) (Statistica 6.0 Pl, ArStat, MS Excel 2000). In statistical analyses, estimate of equations significance used to determine half-life was included. $T_{1/2}$ values in the case of which confidence interval for k constants did not cover themselves ($P \leq 0.05$) were treated as significantly different.

RESULTS AND DISCUSSION

Characteristics of materials used to composting

The sum of content of polycyclic aromatic hydrocarbons in sewage sludge used for experiment was 10385 μg/kg (±830) and was similar to PAHs content in sewage sludges from south-eastern Poland [3], and comparable with data presented by other authors [7, 19, 36, 40].

Dominance of 3-ring PAHs (48.7%) – mainly acenaphthylene (12.4%) and acenaphthene (38.7%) was observed. The contribution of mutagenic and carcinogenic 5- and 6-rings polycyclic aromatic hydrocarbons was 17.9 and 7.4% respectively (Tab. 2). The contribution

Table 2. The initial and final content [μg/kg] and half life [days] of PAHs in investigated materials during composting process

PAHs	Sewage sludge (100%)			Sewage sludge (80%) + fly ash (20%)			Sewage sludge (70%) + fly ash (30%)		
	I	II	$T_{1/2}$	I	II	$T_{1/2}$	I	II	$T_{1/2}$
Naphthalene	n.d.	n.d.	–	n.d.	n.d.	–	n.d.	n.d.	–
Acenaphthylene	128 7 ±12	488 ±15*	165	2553 ±10	306 ±11*	75	668 ±12	299 ±11*	199
Acenaphthene	298 2 ±14	1194 ±13*	175	1709 ±12	853 ±10*	230	1240 ±10	881 ±9*	469
Fluorene	597 ±11	446 ±9*	549	415 ±10	336 ±8*	754	463 ±10	341 ±12*	521
Phenantrene	141 ±11	137 ±11	–	95 ±14	89 ±11	–	94 ±12	110 ±8	–
Anthracene	53 ±10	65 ±12	–	37 ±11	39 ±9	–	45 ±11	40 ±8	–
Fluoranthene	775 ±8	581 ±15*	558	297 ±9	373 ±12*	–	443 ±9	363 ±7*	805
Pyrene	906 ±8	655 ±10*	493	151 ±9	366 ±12*	–	438 ±13	296 ±12*	407
Benzo[a]anthracene	459 ±12	642 ±8	–	174 ±12	101 ±9*	295	165 ±8	51 ±10*	137
Chryzene	555 ±14	38 ±8*	59	130 ±12	33 ±13*	117	175 ±11	15 ±11*	65
Benzo[b]fluoranthene	119 6 ±11	1043 ±12*	1164	597 ±11	441 ±10*	529	242 ±12	290 ±9*	–
Benzo[k]fluoranthene	n.d.	n.d.	–	n.d.	n.d.	–	n.d.	n.d.	–
Benzo[a]pyrene	573 ±13	444 ±13*	628	249 ±7	200 ±9*	721	163 ±10	98 ±9*	317
Dibenz[ah]anthracene	90 ±12	77 ±10	–	51 ±9	n.d.	–	23 ±12	n.d.	–
Benzo[ghi]perylene	392 ±10	331 ±13	–	228 ±11	179 ±12*	650	105 ±13	93 ±12	–
Indeno[1,2,3-cd]pyrene	377 ±10	303 ±12*	733	176 ±12	187 ±8	–	96 ±9	69 ±11*	493

I i II – beginning and end of composting respectively; $T_{1/2}$ – half life of PAHs calculate on the basis of equation (1) and (2); ± – relative standard deviation (n = 3) [%]; * indicates statistically significant differences ($P \leq 0.05$) in relation to beginning of experiment

of benzo[a]pyrene, considered to be representative of all 16 PAH, was around 5.5%. It is characteristic for most of sewage sludges [19].

Taking into consideration the directive of the European Union [13] concerning the maximum content of a total of 11 PAHs (6 mg/kg) in sewage sludge, we can say that - this limit in sewage sludge used in the experiment was exceeded by 16%.

In coal fly ash used in the experiment only trace content of PAHs (5.4 μg/kg) was found. Among all 16 determined compounds only phenanthrene (0.9 μg/kg), anthracene (1.9 μg/kg and chrysene (2.7 μg/kg) were present in fly ash.

The fly ash addition had a significant effect on decrease of investigated compounds content in prepared for composting materials (Tab. 2). A sum of 16 PAHs content decreased about 34 and 58% for 20 and 30% addition of fly ash, respectively. "Dilution" of sludge with fly ash has also resulted in the lowering of the content of PAHs, taken into consideration when evaluating sewage sludge for agricultural purposes [13]. The only exception was acenaphthylene, for which, when compared with its content in sewage sludge, a significant increase was observed (Tab. 2). Further increase of fly ash addition (from 20 to 30%) caused gradual decrease of only 5- and 6-ring PAHs. In the case of the other PAHs, an increase of fly ash addition to 30% did not cause further changes of their content. Only in the case of fluoranthene, pyrene and chrysene significant ($P \leq 0.05$) an increase of their content – when compared with sludge with 20% addition of fly ash – was observed (Tab. 2). It is difficult to explain a significant increase of acenaphthylene share after fly ash addition to the sewage sludge. An increase in the content of 4-ring PAHs (fluoranthene, pyrene and chrysene) was also surprising when fly ash share increased from 20 to 30% in the materials prepared for composting. Coal fly ash is characterized by a relatively low content of PAHs, in connection with it, one could expect that the effect of "dilution" of PAH present in sewage sludge (related to decrease of their concentration) will take place, rather than their content increase. Observed phenomenon is most probably related with PAHs sorption/ desorption processes, and explanation of it requires additional research, especially on PAHs bioavailable forms and their sorption on materials subjected to the composting process. Higher contents of the compounds studied could be related to pollution of materials during sludge homogenization process with fly ash.

Changes of PAH content during composting

In soils losses of semi-volatile organic compounds (e.g. PAHs) take place in two stages. Compound dissipation from soils has been conceptualized using biphasic decay curves, i.e. a preliminary short period of rapid loss, followed by a subsequent longer period of slower loss. In the first stage, volatilization and leaching processes of mostly 2 and 3-ring PAH prevail, while in the second stage major role is played by microbiological degradation, slow sorption (e.g. sequestration or forming of bond-residue) and physical or chemical degradation [20, 21, 27, 30, 34, 35]. Both, first and second stages are determined to a large extent by mechanisms of sorption/desorption of contaminants by soil organic matter [34, 35]. Processes of PAHs content changes during composting have not been precisely investigated yet. The results on the changes in the content of semi-volatile organic contaminants [10, 18, 22, 42] obtained so far showed that during composting they underwent (similarly with the soils) sorption by the components of the compost and became incorporated in the stable structures of the macro-particles (such as humus substance), degradation to their metabolites, and then to CO_2 and H_2O. A higher content of organic

matter in the sewage sludge than in the soil meant that sorption significantly influenced PAH degradation. This can lead to the limiting of bioavailability (a decrease in the PAH degradation range), or else to a decrease in the toxicity of pollutants (a process favoring biodegradation). Moreover, high content of nutrients may significantly increase degradation of persistent organic pollutants through stimulating activity of microorganisms capable of their mineralization [20, 21, 33].

Changes in the content of 16 PAHs during composting process were characterized by clear dynamic in all variants of experiment (Fig. 2). When analyzing changes of 16 PAHs sum content it was noted that they were dependent on amount of fly ash addition.

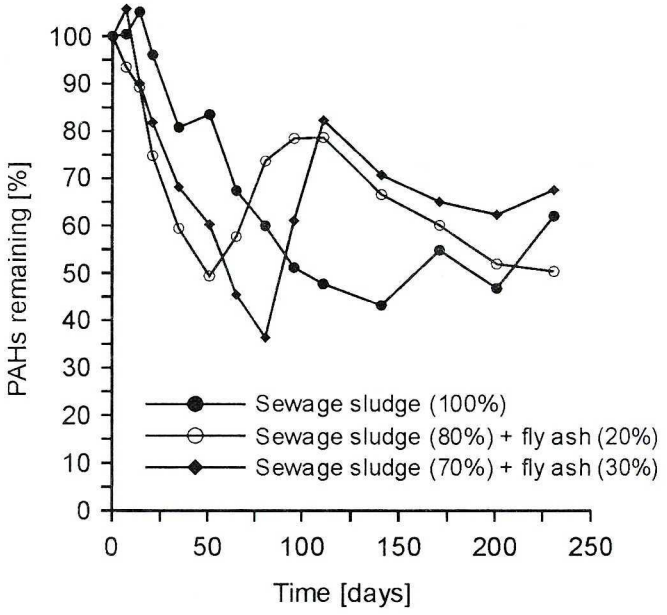

Fig. 2. Changes [%] of Σ 16 PAHs content in materials during composting process

In sewage sludge composted for 150 days, gradual decrease of PAHs content was noted. An increase of investigated compounds content was observed about 51st day of process, but it had no significant influence on it's course. A decrease in the sum of 16 PAHs was determined primarily by all changes in the content of 3- and 5-ring compounds and, to a lesser degree, 4-ring PAHs (Fig. 3). The content of 6-ring PAHs remained unchanged almost through a whole period of composting. An insignificant decrease was noted in the second part of the experiment, although an increase of their content following it indicates (Fig. 3 A) that it was related to the process of strong sorption of investigated contaminants by compost components. Organic matter present in sewage sludge was subject of continuous changes and transformations [2], what could affect its ability to adsorb PAHs. In Figure 4 the content of total organic carbon (TOC) changes in composted materials is presented. Our earlier paper [2] showed that in sewage sludge-amended soils, after initial decrease of PAHs content, an increase followed. Moreover, clear dependence on total organic matter content was noted. In the present studies, significant relations were observed in the

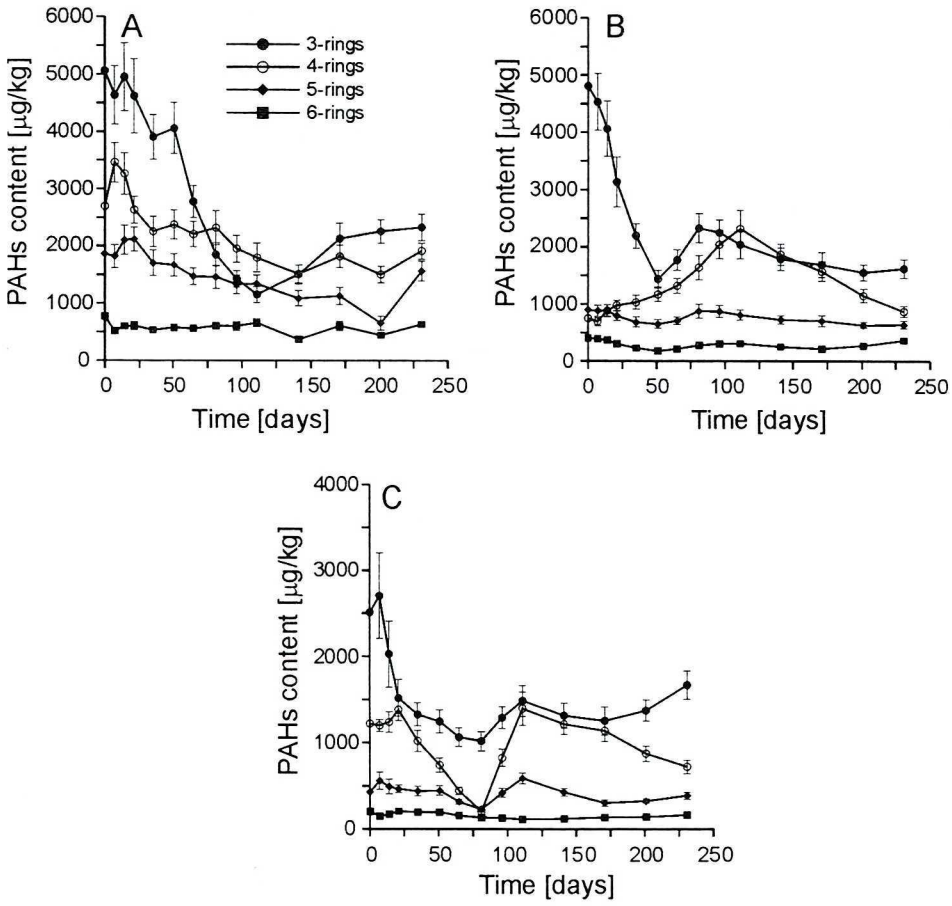

Fig. 3. Changes of polycyclic aromatic hydrocarbons content depending on number of rings
A – sewage sludge (100%), B – sewage sludge (80%) and fly ash (20%),
C – sewage sludge (70%) and fly ash (30%)

experimental variant with sewage sludge only and sludge with a 30% fly ash addition, both in the case of individual PAHs and their sum. In the case of experiment with sewage sludge alone, significant dependencies between TOC content and acenaphthylene ($r = 0.540$), acenaphthene ($r = 0.826$), fluoranthene ($r = 0.628$), pyrene ($r = 0.740$), chrysene ($r = 0.540$), benzo[b]fluoranthene ($r = 0.728$) and sum of 16 PAHs ($r = 0.831$) were observed. A similar number of significant correlations between TOC and acenaphthene ($r = 0.931$), fluorene ($r = 0.887$), anthracene ($r = 0.843$), fluoranthene ($r = 0.666$), pyrene ($r = 0.646$), dibenz[ah]anthracene ($r = 0.788$) and 16 PAH sum ($r = 0.911$) were noted in an experiment with 30% addition of fly ash. As one can see, only in the case of acenaphthene and pyrene presented relationships repeated in both variants of experiment. For remaining compounds correlations were specific for a given variant of experiment. It indicates various directions of relationships between TOC and PAHs. This was also indicated by a different direction of mutual influences between TOC and PAHs. This last observation confirmed differentiated influence of organic matter on PAHs which depends on matrix properties (i.e. pH, CEC, etc.)

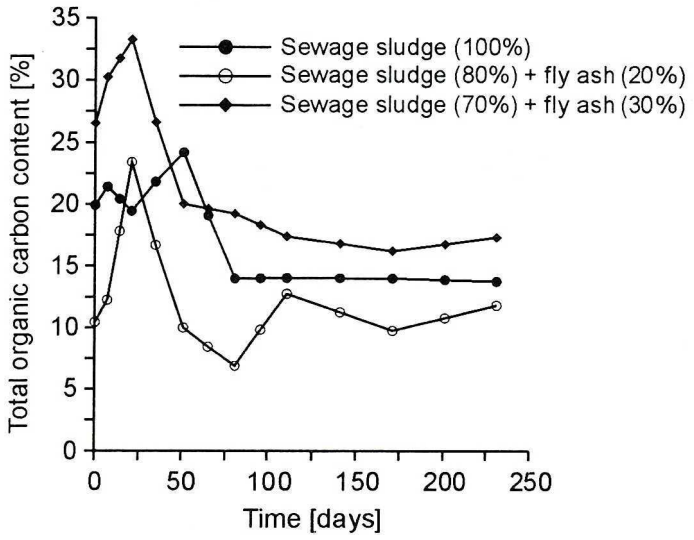

Fig. 4. Changes of total organic carbon (TOC) content during composting process

observed by other authors [12, 26, 34, 35]. In no case in a variant with 20% fly ash addition, significant relationships between TOC and individual PAHs were noted.

Changes of PAHs content in sludge with both, 20 and 30% fly ash addition were similar. Some differences were noted only between the 50th and 80th composting day, and by the end of experiment (Fig. 2). However, the differences observed did not influence high coefficients of correlation (statistically significant for $P \geq 0.001$), changes in the individual PAHs' content and their sum between experimental variants with the 20 and 30% fly ash share. Similarly, during composting of the sewages sludge only, the least significant influence on the changes of the studied compounds was noted for 6-ring PAHs. Changes in the content of 3, 5 and 6-ring compounds between experimental variants with 20% and 30% fly ash share were very much alike (Fig. 3 B and C). However, distinct changes were observed for 4-ring PAH (Fig 3 B and C). In sewage sludge with 20% fly ash addition, a gradual increase of these compounds up to the 100th day of composting was observed, and then their content fell to the level noted at the beginning of the experiment. In the experiment with 30% of fly ash addition, in initial period (0–21 days) the content of 4-ring PAHs remained constant, after which insignificant increase was observed, followed by a rapid decrease of investigated compounds content (from 1200 to 200 μg/kg) between the 25th and 75th day of composting. During later stages of the present experiment, the content of 4-ring PAHs was slightly below the initial content of these compounds (Fig. 3 C).

It is difficult to explain causes for such shaping of 4-ring PAH content. Most probably, it is related to sorption of investigated compounds by fly ash. An important role of fly ash in this process is suggested by the fact that no such phenomenon was observed when sludge without fly ash addition was composted. Only in initial stage of sewage sludge composting, an increase of 4-ring PAHs, like in experiment with 30% fly ash addition was noted. Solving this issue requires more research, especially on sorption properties of ash as a background for content of bioavailable forms of investigated compounds.

After 231 days the greatest decrease of PAH sum content (49.7% – when compared with the beginning of experiment) was noted in sewage sludge with 20% addition of fly ash. A decrease of determined PAHs content in sewage sludge with highest addition of ash (30%) was significantly lower (32.4%). In the compost obtained from sewage sludge alone, PAHs content was 38% lower than in output material.

When a number of rings is taken in to consideration, highest range of decrease was noted for 3-ring PAHs and it was greatest (66.3%) in compost with 20% of fly ash. Similarly, in the case of 5-ring compounds, their most dissipation was observed in composted mixture of sewage sludge with 20% fly ash (Fig. 5). When evaluating changes of individual PAHs content (Tab. 2), the highest decrease of their content (>50%) was noted for chrysene and acenaphthylene. Moreover, in experiment with 30% fly ash addition, a significant decrease was also observed for benzo[a]anthracene. In the case of remaining PAHs no clear dependencies were observed and changes of their content were clearly depended on variant of experiment (Tab. 2).

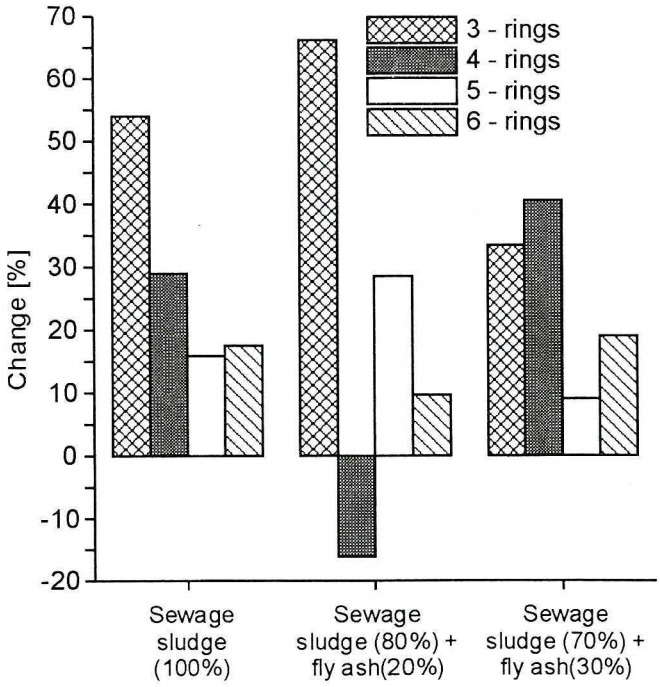

Fig. 5. Changes of PAHs content depending on number of rings in final stage in relation to beginning of experiment

Half-lives of PAHs

Half-lives of investigated compounds determined on the basis of equations (1) and (2) are presented in table 2. Only for acenaphthylene and chrysene in all variants of experiment $T_{1/2}$ was under one year. In the case of acenaphthene, benzo[a]anthracene and benzo[a]pyrene $T_{1/2}$ under one year was noted in chosen variants of experiment only (Tab. 2), while extremely high values ($T_{1/2}$ > 500 days) were obtained for fluorene, fluoranthene

and benzo[ghi]perylene. In the case of phenanthrene and anthracene no statistically significant change of their content was noted (Tab. 2), what may indicate a great persistence of these compounds in investigated materials. However, low persistence of chrysene and relatively high fluorene and phenanthrene is surprising. Presented in literature data concerning $T_{1/2}$ for fluorene and phenanthrene range from 20 to 70 days [27, 28] and from 16 to 123 days [29] respectively. The research show [27, 33] that $T_{1/2}$ values may differ significantly depending on soil properties, and especially soil organic matter content. High $T_{1/2}$ values in presented experiment may be related to a strong interaction of mentioned compounds with sewage sludge and/or fly ash. Previous research [5] indicates a significant influence of sewage sludge on persistence of phenanthrene. Determined half-life of phenanthrene in sewage sludge-amended soil ranged from 360 to 2000 days, depending on an applied sewage sludge dose [5].

Low solubility in water and low volatility of chrysene should rather point to high persistence of this compound in the composted materials. In earlier studies [33] on PAHs decomposition in soils polluted with aircraft fuel, half-lives of chrysene were noted several times greater ranging from 400 to 450 days. In soils fertilized with sewage sludge [5] $T_{1/2}$ values were even higher and ranging from 300 to 2000 days. Determined by Maliszewska-Korybach [27, 28] half-lives times of chrysene in soils polluted with this compound were from 120 to 365 days. Author noted [28] that effect of compost application into soil on chrysene degradation depended on type of soil as well as on a compost dose. In soil originating from strong loamy sand, addition of 3% of compost (w/w) caused over 50% decrease of $T_{1/2}$, while in soil originating from sandy loam addition of compost favored increase of chrysene persistence. Observed in this research, relatively short half-life of chrysene (17 days) may result from favorable conditions, increasing rate of its degradation during composting. This compound could also be strongly entrapped by organic matter (e.g. sequestration or bound residue). In the case of other PAHs, the observed $T_{1/2}$ values were similar or slightly higher than data presented by other authors [5, 20, 21, 27, 33].

CONCLUSION

The results obtained in the present research indicate distinct differences in degradation of investigated compounds. Addition of coal fly ash to sewage sludge had – depending on its amount – various effects on PAHs disappearance. Regardless to the amount of fly ash added it always caused increase of PAHs losses from sludge in the first 50–75 days period and decrease of losses in later period. These differences concerned 3 and 4-ring PAHs mainly. The positive correlation coefficients between content of most PAHs and TOC, when sludge and sludge with 30% fly ash addition are taken into consideration, indicate that transformations of PAHs in these experiments are closely related to organic carbon transformations. Lack of significant dependencies in experiment with 20% fly ash addition, for which relatively best PAHs degradation was observed, does not preclude role of organic carbon in this process. It may only indicate a weaker interaction between PAHs and TOC (expressed by lack of significant dependencies) allowing more efficient degradation of these compounds. However, additional studies, concentrating especially on the determination of the bioavailable PAH fraction share, are necessary to confirm the above observations. It is also important to determine sorption properties of these compounds in relation to composted materials.

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