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Original Research

Preliminary Investigation for Forecasting the Content of Polycyclic Aromatic Hydrocarbons after Soil Fertilization with Sewage Sludge

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Abstract

In addition to the positive effects on the physical, chemical and biological soil properties of the application of sewage sludge into the soil, there is often an increase of both organic and inorganic pollutants in the soil. One of the most popular organic xenobiotics frequently present in sewage sludge are polycyclic aromatic hydrocarbons. The aim of this study is to determine possibilities for forecasting the content of individual PAHs in the soil on the basis of their content in sewage sludge. Two types of sewage sludge with different PAH contents, both in terms of quantity and quality, were examined. The different types of sludge were introduced into the soil in the following doses: 30, 75, 150, 300 and 600 t/ha. The results obtained showed a clear differentiation in the behaviour of individual PAHs in relation to the dose applied and the type of sewage sludge.

Keywords: sewage sludge, PAHs, soil fertilization, persistent organic pollutants

Introduction

About 3.3 million tons of hydrated sewage sludge is produced in Poland every year [1], out of which only a few percent is utilized. Such a situation presents a great danger to the environment. One of the methods of sewage sludge utilization is their application in agriculture. About 1/3 of the sludge generated annually in the countries of the European Union, the United States and Canada [2] is utilized in this way. The application of sewage sludge improves the physical, biological and chemical properties of the soil [3].

However, numerous research reports point to the danger of a permanent introduction of organic pollutants into the soil with the agriculture utilization of sewage sludge [4-9]. Researchers have drawn attention [4, 5, 10-12] to the high content, relative to their origin, of polychlorinated biphenyls (PCB), dioxins and furans (PCDD/F), pesticides and polycyclic aromatic hydrocarbons and their derivatives. The application of sewage sludge in agriculture creates the potential danger of their penetration into plants and then into the human food chain [13, 14].

Despite the above actual danger of environmental pollution with sewage sludge being introduced into the soil, no attempts have been undertaken in Poland to determine the maximum content of individual PAHs - or their sum - in sewage sludge used for fertilization. This is an especially difficult issue which requires a series of studies. The following problems are the most difficult part of the above issue [4, 15]:

(1) the scarcity of information on the content and behaviour of PAHs during sewage treatment (in relation to its origin, method of treatment, etc.), or further PAH behaviour in soil fertilized with sludge (in relation to soil properties and sewage sludge);
(2) the regulation of organic contaminant limits would be extremely costly;

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Table 1. Physicochemical properties of soils and sewage sludge used in research.

Properties		Matrix			
		Soil *P*	Sewage sludge *P*	Soil *W*	Sewage sludge *W*
Soil texture (%)	1-0.1	86	-	86	-
	0.1-0.02	7	-	7	-
	<0.02	7	-	7	-
pH [in KCl]		4.3	6.01	5.8	6.4
H_H (mmol/kg)		46.5	47.3	22.3	24.5
CEC (mmol/kg)		13.4	500.4	48.9	583.2
TEB (mmol/kg)		59.9	547.7	71.2	607.7
BS (%)		22.4	99.4	68.7	96.0
TOC (g/kg)		11.2	210	12.1	237
N (g/kg)		1.4	17.8	1.2	19.8

H_H – hydrolitical acidity; CEC – cation exchange capacity; TEB – total of exchangeable bases; BS – degree of base saturation, TOC – total organic carbon, N – total nitrogen

(3) no uniform opinion on the existence or profitability of using such types of limits.

Despite the difficulties presented above, standards worked out in the European Union [16, 17] relate both to the sum and a few individual PAHs. In the present study, theoretical calculations were carried out in order to determine possible PAH content in the soil after the introduction of sewage sludge.

Materials and Methods

Determination of PAH Content

PAH was determined using the HPLC method with detection UV (254 nm) after optimization of the analytic process with respect to the amount and type of solvent used and extraction time, by means of ultrasonic method [1] and optimization of the extract purification process by the solid phase extraction method [18, 19]. 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 ml/min. Detection was carried out at 254 nm. The column was installed in a thermostated oven at 31°C (LCO 101, ECOM, Czech Republic). All reported concentration values of PAHs are expressed on a dry-wt basis of sewage sludge (determined by drying the sewage sludge for 24 h at 105°C) and are the average of triplicate extraction.

Plot Experiment

The study block consisted of 2 x 6 plots (15 m^2 each), founded on light soils. Plots were located according to increasing doses of sludge as follows: soil without any fertilization – control; sewage sludge 30 t/ha, 75 t/ha, 150 t/ha, 300 t/ha, 600 t/ha. Fermented sewage sludges from mechanical-biological sewage treatment plants were used. The amount of sludge applied was established by taking into account fertilizing (30 t/ha), melioration (75-300 t/ha), and extreme doses (600 t/ha). The choice of extreme doses was aimed at establishing the degree at which soil becomes polluted with PAHs, and what the background is upon which the durability of these compounds is founded. Sludge doses were calculated taking into consideration the sludge's dry mass and the density of the solid soil phase. It was mixed with a surface soil layer up to a depth of 20 cm.

Data Analysis

The calculation was determined on the basis of the following equation:

$$G = \left(\frac{V}{\rho}\right) \cdot WWA_G \ [mg/plot]$$

where:

G - PAH content in soil on the experimental plot (3x5x0.2 m) [mg];

V - volume of soil on the experimental plot (3x5x0.2 m) [m^3];

ρ - bulk density of soil after sewage sludge introduction [$kg \cdot m^{-3}$];

WWA_G - PAH content in soil [$mg \cdot kg^{-1}$].

$$O = D_o \cdot WWA_O \ [mg/sewage\ sludge\ dose]$$

where:

O - PAH amount applied with sewage sludge on experimental plot [mg]

D_o - sewage sludge dose introduced on experimental plot [kg];

WWA_O - PAH content in sewage sludge [$mg \cdot kg^{-1}$].

Table 2. Concentration of PAHs in soils used in research.

PAHs	Soil *P*		Soil *W*	
	Content [µg · kg^{-1}]	RSD [%]	Content [µg · kg^{-1}]	RSD [%]
Naphthalene (Na)	3.5	17	3.0	9
Acenaphthylene (Ace)	15.1	15	13.0	11
Acenaphthene (Ac)	10.4	13	10.6	17
Fluorene (Fl)	0.0	-	3.6	6
Phenanthrene (Phen)	1.2	16	0.8	9
Anthracene (Ant)	0.1	15	0.1	9
Fluoranthene (Fluo)	2.3	15	2.4	11
Pyrene (Pyr)	0.0	-	1.8	8
Benzo[a]anthracene (BaA)	1.2	15	1.4	4
Chryzene (Ch)	1.1	10	1.2	14
Benzo[b]fluoranthene (BbF)	1.6	12	3.6	21
Benzo[k]fluoranthene (BkF)	1.1	13	1.2	18
Benzo[a]pyrene (BaP)	1.5	10	1.7	15
Dibenz[a,h]anthracene (DahA)	1.0	12	2.2	6
Benzo[ghi]perylene (BghiP)	0.6	13	0.7	10
Indeno[1,2,3-cd]pyrene (Ind)	2.3	15	2.3	11
∑ 16 PAH	43.0	14	49.6	8

RSD – relative standar deviation [%]

Results and Discussion

Characteristic of Soils and Sewage Sludge

Tables 1 and 2 present the properties of the soils used in this experiment, and Figure 1 – the content of individual PAHs present in the sewage sludge. The soils were characterized by their very low PAH content (about 50 µg/kg); a predominant content of 2- and 3-ring PAHs was determined (Table 2).

The sludge showed a clear differentiation of PAH content. Sludge *P* was characterized by the PAH sum content at a level of 3894 µg/kg with predominantly "light" (2- and 3-rings) PAHs (Fig. 1). In comparison, sludge *W* was characterized by both a higher content of the PAH sum (5712 µg/kg) and clear differences in their group composition (Fig. 1).

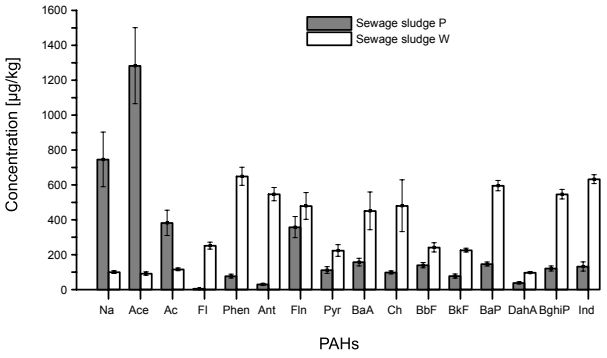

Fig. 1. The concentration of individual PAHs in sewage sludge used in the research.

PAH Concentration after Sewage Sludge Application

Figure 2 presents PAH content in soils after the introduction of sewage sludge. An increase in the added sludge mass caused an increase in PAH content in the fertilized soil. An introduction of the lowest sludge doses caused a more than double or triple increase in PAH sum in relation to the control soil, in the experiment with sludge *P* and sludge *W,* respectively (Fig. 2). An application of consecutive doses caused a further, systematic increase in PAH content, clearly related to the sludge dose applied and its type. A statistically non-significant increase in relation to a dose was observed only in the case of sludge *W*, at its highest dose of 300 t/ha (Fig. 2), and a possible explanation of this phenomenon was presented in another study [20]. An explanation of this phenomenon can be found in the sorption properties of the soils studied in relation to polycyclic aromatic hydrocarbons. Soil with a sewage sludge content at a dose of 20% (and hence with a higher value of organic matter content) can show higher

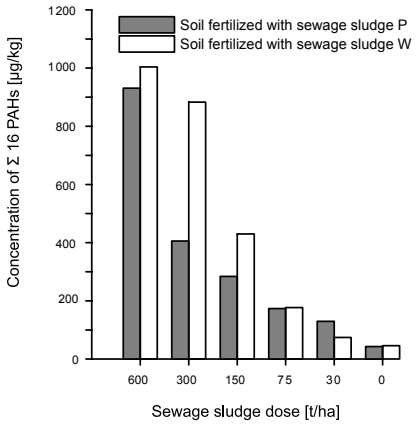

Fig. 2. The concentration of ∑ 16 PAHs in sewge sludge--amneded soils.

sorption abilities in relation to PAH than soil with a 10% sludge addition. Hence, it can be gathered that sorption processes at an increased amount of organic matter are more intense and clearer. Moreover, with an increase in sewage sludge mass, there is an increase in the amount of organic matter of anthropogenic origin (dust, ash and soot) which exerts a stronger influence on PAH than the influence exerted by natural matter. This is due to the "highly" aromatic character of this type of matter and its considerable specific surface area as compared to natural organic matter [20].

The 3-ring PAHs had the highest share in both variants of the present experiment. However, both in the soil with the addition of sludge *P* and sludge *W* with increasing sludge doses, a decrease was observed in the share of 3-ring PAHs and an increase was observed in 4- and 5-ring PAHs.

In the case of sludge *P*, the above phenomenon should be related rather with the properties of the xenobiotics analyzed, whereas in the case of sludge *W* this phenomenon should be explained by the content of the above compounds in the sewage sludge. In *P* sludge, 3-ring PAHs were predominant (Fig. 1). However, these latter compounds are highly volatile. Moreover, they undergo very quick biological and photochemical degradation in the soil [21, 22]. Hence, it should be assumed that during the introduction of sewage sludge into the soil, they quickly dispersed in the environment.

In sludge *W*, 2- and 3-ring PAHs had a small share only; 4-ring hydrocarbons and 5-ring benzo[a]pyrene were predominant. Hence, the increase in the level of these latter compounds is directly proportional to the sludge share in the soil.

The Criteria of Soil Contamination by PAHs

While evaluating the PAH content according to the criteria proposed by IUNG (The Institute of Soil Science, Fertilization and Plant Cultivation in Puławy) [23] which take into consideration the organic carbon content in the soil, both in the experiment with sewage sludge P and W, an introduction of the lowest sludge doses (30 and 75 t/ha) resulted in an increase in PAH content; however, the natural level of these compounds in the soil was not exceeded. Further increases of the sludge dose, 150-600 t/ha, in both experimental variants, still categorized the soils as unpolluted; however, increased values had already been noticed.

Based on the classification of the allowable levels of the sum concentration of 10 PAHs in the soil - further to the law on soil and earth quality standards [24], the soils analyzed having been fertilized with sewage sludge can be classified as group A. However, when the content of individual PAHs is evaluated (further to the above law), standards were exceeded (Group A) in the case of the highest sewage sludge doses (600 t/ha) for naphthalene and benzo[a]pyrene (experiment with sewage sludge *P*), and for fluoranthene, benzo[a]pyrene (experiment with sewage sludge *W*).

Prediction of PAH Concentrations

Figure 3 presents the actual (determined in the plot) (G) and forecast (introduced with a specific sewage sludge dose) (O) content of the PAH sum in the soils fertilized with sewage sludge. Data presented in Figure 3 clearly shows the differences in the "behaviour" of PAHs in relation to the sewage sludge dose applied. A relatively good reflection of the calculations made (in the case of PAH sum), both in the plot with sewage sludge *P* and *W*, was obtained for the sewage sludge dose of 75 t/ha, and in the experiment with sewage sludge *P* at doses of 150 and 75 t/ha. When the sewage sludge dose was the highest, forecasting based on the formulae presented above was the least precise. In the experiment with sewage sludge *P*, the actual PAH content determined in the soil was over 40% lower than the amount introduced with sludge (theoretically calculated). In the experiment with sewage sludge *W*, the above difference was twice as high (Fig. 3B). The above was probably related to PAH losses

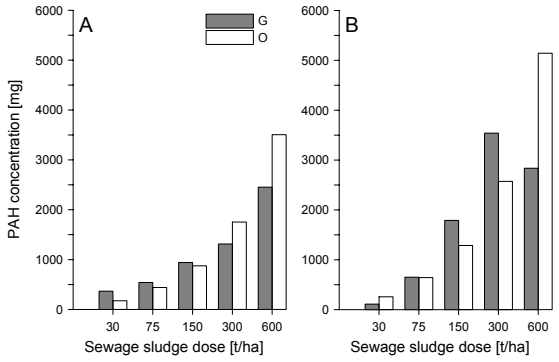

Fig. 3. The actual (determined in the plot) (G) and forecast (introduces in with a specific sewage sludge dose) (O) content of the PAH sum in the sewage sludge-amended soil. A – sewage sludge *P*; B – sewage sludge *W*.

Table 3. The actual (determined in the plot) (G) and forecast (introduces in with a specific sewage sludge dose) (O) content of individual PAHs in sewage sludge (P) -amended soil.

PAHs	Sewage sludge dose									
	30 t/ha		75 t/ha		150 t/ha		300 t/ha		600 t/ha	
	G	O	G	O	G	O	G	O	G	O
Na	44	34	50	84	101	168	138	335	384	671
Ace	115	58	171	144	215	288	352	577	691	1154
Ac	69	17	59	43	95	86	136	172	192	343
Fl	33	0	52	1	69	1	82	2	119	4
Phen	6	3	11	9	16	17	21	34	52	69
Ant	2	1	8	3	14	7	15	13	30	27
Fluo	30	16	51	40	79	80	103	161	233	321
Pyr	15	5	59	13	98	25	112	50	174	100
BaA	9	7	16	18	52	35	57	71	102	141
Ch	5	4	12	11	28	22	32	44	62	88
BbF	6	6	11	16	23	31	67	63	101	126
BkF	10	3	16	9	41	17	43	35	54	69
BaP	4	7	10	16	57	33	65	66	96	131
DahA	-	2	-4	4	-4	8	21	17	24	34
BghiP	10	5	6	14	30	27	38	54	62	108
Ind	8	6	13	15	27	30	31	59	76	118

as a result of volatilization and the biological or photochemical degradation process. Similar conclusions can be drawn in the case of sludge doses of 300 t/ha (Fig. 3A) and 30 t/ha (Fig. 3B).

In the case of the highest dose of sludge *W*, the differences observed can be explained by the fact that samples were collected for analysis two days after the experiment was set, and the degradation processes could have progressed to a higher degree.

Results (Fig. 3A – 30 t/ha; Fig. 3B – 150 and 300 t/ha) in which PAH content in the soil was higher than the amount of PAHs introduced into the soil with sewage sludge (theoretically calculated) were quite surprising. It can be assumed that the above phenomenon was related to the sorption properties of organic matter in relation to polycyclic aromatic hydrocarbons. Sludge is characterized by a high content of organic matter and also other components (dust, soot, biomass, etc.) capable of binding organic xenobiotics strongly. The analytical method applied in the present experiment, even though characterized by a high level of recovery (76% on average) [1], is not able to extract the strongest bound PAHs. Moreover, some authors [25] have drawn attention to the fact that PAH particles can be cumulated by micro-organisms which can decrease the efficiency of extraction. Taking into account earlier studies [15, 20], it can be assumed that the introduction of sewage

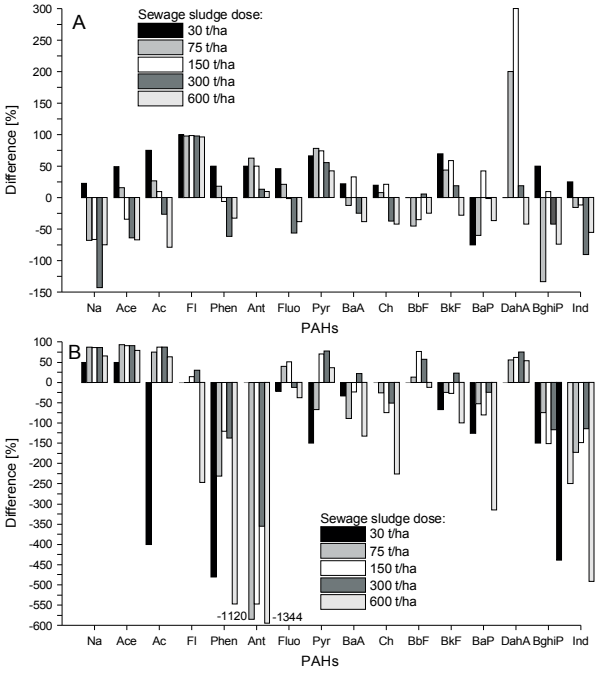

Fig. 4. The difference between forecast and actual PAH content in sewage sludge-amended soil. A – sewage sludge *P*; B – sewage sludge *W*.

Table 4. The actual (determined in the plot) (G) and forecast (introduces in with a specific sewage sludge dose) (O) content of individual PAHs in the sewage sludge (W) -amended soil.

PAHs	Sewage sludge dose									
	30 Mg · ha^{-1}		75 Mg · ha^{-1}		150 Mg · ha^{-1}		300 Mg		600 Mg · ha^{-1}	
	G	O	G	O	G	O	G	O	G	O
Na	10	5	82	11	158	22	325	45	256	89
Ace	8	4	135	10	241	21	440	41	388	82
Ac	1	5	51	13	211	26	401	52	275	103
Fl	-	11	-	28	65	56	160	113	65	226
Phen	5	29	22	73	66	146	123	292	90	583
Ant	0,5	25	5	61	19	123	54	246	34	491
Fluo	18	22	89	54	217	108	191	215	312	430
Pyr	4	10	15	25	170	50	435	100	315	201
BaA	15	20	27	51	82	101	258	202	174	405
Ch	-	22	43	54	62	108	143	216	132	431
BbF	0,0	10	31	27	221	54	249	108	193	217
BkF	6	10	20	25	40	51	130	101	101	202
BaP	12,	27	44	67	74	134	215	268	129	535
DahA	14	4	25	11	57	22	170	43	185	87
BghiP	10	25	35	61	49	123	113	245	91	491
Ind	8	28	26	71	57	142	132	284	96	568

sludge into the soil causes sludge "dispersion," mineralization of part of the organic matter and biomass which weakens the mutual reactions between PAHs and organic matter; hence, PAHs are released and efficiency of extraction increases.

Analyzing data from (Table 3 and 4) on the basis of formulas (1) and (2) together with differences in the actual and forecast content of the individual PAHs in the soils studied as presented in Figures 4A and B, a clear differentiation can be observed relating to the PAH type, sewage sludge dose and experimental variant (experiment with sewage sludge *P* and *W*).

Even though in the case of the PAH sum, a quite accurate reflection of the calculations was obtained (differences did not exceed 50% in most experimental variants), when individual PAHs were analyzed, the differences were considerable. First of all, it was seen in the disproportion in the content of the individual PAHs in the experiment with sewage sludge *P* and in the experiment with sewage sludge *W*.

In the experiment with sludge, only in the case of naphthalene (300 t/ha), dibenz[ah]anthracene (30 and 75 t/ha) and benzo[ghi]perylene (75 t/ha), did the differences not exceed 100%; whereas in most of the cases, considerably lower values were noted. In more than half of the PAHs determined, a relatively good reflection of the calculations was obtained that did not exceed a level of 50% (Fig. 4A). Irrespective of the sewage sludge dose applied, differences lower than 50% (in absolute values) between the forecast PAH content and the content actually determined in the soil was noted for benzo[a]anthracene, chrysene, benzo[b]fluoranthene and phenanthrene, fluoranthene (except for the dose of 300 t/ha) and anthracene (except for the dose of 75 t/ha). In the cases described above, the differences observed resulted from the varied imaging of the results obtained in relation to the sewage sludge dose. For all PAHs mentioned above (except benzo[b]fluoranthene and anthracene) at the highest sludge doses (300 and 600 t/ha), a lower content of these xenobiotics was actually determined in the soil than had been forecast.

In the experiment with sewage sludge *W*, more pronounced divergences were observed, since only in 1/4 of the cases, was the difference between the forecast and actual PAH content below 50%. These were clearly higher for most PAHs (above 150%); and the highest disproportions were observed in the case of phenanthrene and anthracene (Fig. 4B). Only in the case of fluoranthene were differences of 50% noted in all experimental variants. For the remaining PAHs, a higher differentiation was noted (Fig. 4B).

Evaluating the frequency of differences (absolute values) which did not exceed 50% (in relation to the

sewage sludge dose), no significant differentiation was observed. Irrespective of the sewage sludge dose applied (in the experiment with sewage sludge *P*) between 9 and 11 such cases were noted (Table 3); whereas in the experiment with sewage sludge *W*, the frequency was more than twice lower (3-4) (Table 4).

On the basis of the correlation coefficients, relations between the difference in the forecast and actual content of individual PAHs and their sum as related to sludge dose were evaluated. Positive, statistically significant ($P \leq 0.05$) relations between the eight PAHs (acenaphthylene, acenaphtene, fluorene, phenanthrene, fluoranthene, chrysene, benzo[k]fluoranthene and indeno[1,2,3-cd]pyrene) and sum of 16 PAHs were observed in the experiment with sewage sludge *P* and between the three PAHs (naphthalene, acenaphthylene, phenanthrene) and the sum of 16 PAHs in the experiment with sewage sludge *W*. The highest frequency (5 cases) of the occurrence of positive relations of an individual PAH in relation to the remaining compounds from this group was found in the case of fluoranthene (the experiment with sewage sludge *P*) and benzo[ghi]perylene (the experiment with sewage sludge *W*). The calculations obtained showed that there were no clear relations, which probably proves that there is idiosyncratic behaviour which is characteristic of each of the xenobiotics during soil fertilization with sewage sludge. The above calls for individual treatment for each compound from this group; frequently encountered generalizations of PAH transformations based on their sum give only an indicative view on the whole issue and cannot be used as the basis for considering the behaviour of individual PAHs.

Conclusion

The results presented in this paper point to the need for further study covering various soil types and also for sewage sludge to be differentiated with respect to the properties and content and share of various PAHs. On the basis of the studies carried out so far [15, 18, 20], it can be assumed that the sum of PAH content cannot be the basis for the evaluation of the usability of sludge for agricultural purposes. It is obvious that even though the sludge applied contained relatively low PAH levels (having complied with EU standards) [17], at the highest dose, the allowable standard for the soil was exceeded (Category A, according to *Regulation on soil quality standards and the standards of earth quality*) [24]. Even though the sludge dose applied was relatively high and not used in real practice, the observed problem points to the need to work out individual standards for specific PAHs. When working on the above problem (PAH content in sewage sludge) attention must be drawn to the phenomenon described in this study, i.e. in the case of many PAHs, determining the higher content of the above compounds after soil fertilization with sewage sludge than the preliminary forecasts had indicated.

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