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POLYCYCLIC AROMATIC HYDROCARBONS UPTAKE BY SELECTED PLANTS IN THE VICINITY OF HIGHWAY

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WIELOPIERŚCIENIOWE WĘGLOWODORY AROMATYCZNE W WYBRANYCH ROŚLINACH W POBLIŻU AUTOSTRADY

Streszczenie

W pracy przedstawiono badania zawartości wielopierścieniowych węglowodorów aromatycznych w glebie i próbkach wybranych roślin. Szczególną uwagę zwrócono na sorpcję WWA z gleby przez rośliny uprawiane w pobliżu dróg publicznych. W badaniach uwzględniono zarówno rośliny rosnące na poboczu jezdni jak i na polu uprawnym. Jako biologicznych wskaźników jakości środowiska użyto: kapusty, pietruszki, marchwi, ogórka, mniszka lekarskiego, babki szerokolistnej i podbiału. Próbki pobierano w określonych odległościach od jezdni w pobliżu Siedlec, przy trasie E8 (Moskwa – Berlin). Sumaryczna zawartość WWA w próbkach gleby i kapusty zebranych 5 m od jezdni wynosiła odpowiednio 1,51 μg/kg (zawartość WWA kancerogennych = 556,03 ng/kg) i 358,90 ng/kg (zawartość WWA kancerogennych = 101,17 ng/kg). Natomiast w próbkach zebranych w odległości 15 m od jezdni sumaryczna zawartość WWA wynosiła odpowiednio 136,46 ng/kg w glebie (kancerogennych = 27,30 ng/kg) i 87,20 ng/kg w kapuście (kancerogennych = 12,17 ng/kg).

Summary

The presents paper determination of 16 PAHs in collected samples of soil or plants and investigation of sorption process dynamics. The investigation included plants growing on shoulder of road and on cultivated field. As accumulations of quality of environment cabbage, parsley, carrot, cucumber, dandelion (*Taraxacum officinale*), plantain (*Plantago major*) and colfsfoot (*Tussilago farfara*) were used. The samples were collected at a defined distance to the main road E8 (Moscow – Berlin), near Siedlce. A total PAHs concentration in soil and cabbage samples collected in the distance 5 m to the road was 1.51 μg/kg (total carcinogenic PAHs = 556.03 ng/kg) and 358.90 ng/kg (total carcinogenic = 101.17 ng/kg) respectively. On the other hand, total PAHs in samples collected in 15 m distance to the road was 136.46 ng/kg for soil (total carcinogenic = 27.30 ng/kg) and 87.20 ng/kg for cabbage (total carcinogenic = 12.17 ng/kg).

INTRODUCTION

Over the last several decades, an increased level of environmental pollution by toxic chemical substances, to which belong, among others, polycyclic aromatic hydrocarbons (PAHs). Has been noted the PAH source in the natural environment is industrial development, and mainly the intensive growth of automotive transportation. The upper surface soil layer is particularly susceptible to the accumulation of prevailing quantities of these compounds. Depending on the pH, the time of year and the geomorphologic nature of the soil, PAH accumulation will occur with varying intensity and levels of resultant biodegradation. Among the most important processes causing PAH breakdown in the environment are evaporation, leaching, sorption, solar ray induced photolysis, oxidation reactions, substitution and addition as well as microbiological changes.

The use of biological methods for evaluation of contamination degree is one of trends in contemporary analysis. It is connected with numerous advantages of these methods compared with classical analytical procedures. As an example can serve biomonitoring – carried out with the help of natural bio monitors. In particular, worth attention are biosensors, bio tests, immunosorbents used for preliminary preparation of investigated sample or as a filling of chromatographic column or as an element of a micro reactor in Flow Injection Analysis (FIA) or bio columns [8, 19, 24, 26]. Biosensors are particularly suitable as detector of xenobiotic concentration in a selected element of ecosystem [10, 11, 15, 28]. The introduction of biomonitors to an everyday analytical practice must be preceded by investigations of occurrence correlation of potential xenobiotics in selected species of plants and in selected elements of environment [9, 16, 20].

Soil contamination with petroleum hydrocarbons is a serious problem. In the soil in the location of highways, fuel pumping station and airfields high pollutant concentrations are found. It is necessary to pay attention to the fact that in many places vegetables are grown in the nearest neighbourhood of a road and then directly or indirectly are used as human food. Emission of PAHs is connected with road transport [5, 17, 18]. The increase of PAHs concentration level in the air can be the reason of numerous diseases connected with interaction of this group of compounds on process of replication and mutation of DNA or carcinogenesis [12].

In the case of arable area, high level of impurities can cause retardation of plant growth and unfavourable changes in crops [5, 14, 21].

PAH in the soil can be affected by many various physicochemical (oxidation, sorption, chemical degradation) and microbial transformations. The most important are processes resulting in total mineralization. The rate of decomposition of PAHs in the soil is not directly dependent on general number of soil microorganisms but on the presence of specific microorganism populations capable of decomposing PAH [6, 14, 22, 27]. The aim of this study was to determine the concentration of 16 PAHs in samples of soil and in herbs and vegetables destined directly or indirectly for consumption.

MATERIALS AND METHODS

The aim of this study was to determine the concentration of 16 PAHs: naphthalene (Na), acenaphthylene (Ace), acenaphtene (Acn), fluorene (Flu), phenanthrene (Fen), anthracene (An), fluoranthene (Fl), pyrene (Pir), benzo(a)anthracene (B(a)A), chrysene

(Ch), benzo(b)fluoranthene (B(b)F), benzo(k)fluoranthene (B(k)F), benzo(a)pyrene (B(a)P), dibenzo(a,h)anthracene (D(ah)A), benzo(g,h,i)perylene (B(ghi)P), indeno[1,2,3-c,d]pyrene (IP) in soil samples and in herbs and vegetables destined directly or indirectly for consumption.

The area of investigation:

- detection and determination of PAHs in collected samples of soil and plants;
- investigation of sorption process dynamics of determined hydrocarbons by soil, over and underground selected plants.

The investigation included plants growing on a road shoulder and on cultivable fields. Samples were collected in the vicinity of Siedlce, along international route E8 (Moscow – Berlin). In each case the analysed samples were taken from six points, symmetrically orientated to an axis of a road. The soil samples were collected in a distance of 5 m and 15 m from the road. The samples of dandelion (*Taraxacum officinale*) – 5 m, samples of plantain (*Plantago major*) – 5 m, samples of colfsfoot (*Tussilago farfara*) – 5 m, samples of carrot (roots and leaves) – 5 m and 15 m, samples of parsley (roots and leaves) – 5 m and 15 m, samples of cabbage – 5 m and 15 m, samples of cucumber – 15 m. The materials after grinding were mixed in order to obtain average samples for the analysis.

The freshly sample (soil or plants) to the contenting weight, then (10 g ± 0.001) extracted in a Soxhlet apparatus for 8 hours with hexane (170 cm^3). The extract was concentrated by evaporation. The residue (2-5 cm^3) was subjected to solid phase extraction (SPE) on a column (3 cm^3) packed with chemically modified silica [10]. SPE column was washed earlier with hexane (3 x 4 cm^3). PAHs were eluted by a mixture of toluene and acetonitrile (1:3) (3 x 3 cm^3). The eluate was evaporated, the residue dissolved in 2 cm^3 of acetonitrile and chromatographed (AN-384, J.T. Baker).

The HPLC analysis was performed using Shimadzu (Kyoto, Japan) instrument equipped with Rheodyne Model 7125 injector with $2*10^{-11}$ m^3 sample loop, a column (250 x 4 mm) packed with Si 60-ODS (particle size 10 μm), UV detector SPD-6 and integrator C-R6A. The mobile phase consisted of aqueous methanol (70%, v/v). The flow rate was 1 cm^3/min. Reagents purity for HPLC: hexane, methanol, toluene, acetonitryle were HPLC-grade (S. Witko – J.T. Baker, Łódź, Poland). As standards a mixture of 16 PAHs (Fluka AG, Busch, Switzerland).

RESULTS AND DISCUSSION

Evaluation of investigated samples of soil and plants is presented in tables 1–5. As accumulations of quality of environment cabbage, parsley, carrot, cucumber, dandelion (*Taraxacum officinale*), plantain (*Plantago major*) and coltsfoot (*Tussilago farfara*) were used (Dandelion, plantain and coltsfoot are used in folk medicine for their anti-inflammatory properties). The comparison of PAHs concentration in various distances from a road showed that the amount of determined compounds decreases with the distance from the main source of contamination, in soil and in whole ecosystem, e.g. in soil, the concentration of phenanthrene was 113.05 ng/kg in a sample collected in a distance of 15 m and 10.26 ng/kg in a sample collected in a distance of 5 m from the road (Tab. 1). In samples of cabbage, the concentration of benzo(a)anthracene decreases from 10.21 to 4.05 ng/kg with the distance from the road (Tab. 2). A similar relationship can be observed for other hydrocarbons in all investigated samples. The obtained results for individual compounds concentration show unambiguously a significant influence of arterial road on the level of PAHs in soil and in

Table 1. Content of PAHs [ng/kg] in soil samples from cultivated fields near the international route (n = 7)

Compound	Distance			
	*5 m	*15 m	#5 m	#15 m
Na	124.85 ±0.02	23.12 ±0.01	98.23 ±0.04	12.19 ±0.01
Ace	135.04 ±0.03	18.32 ±0.03	102.17 ±0.03	16.08 ±0.02
Acn	110.76 ±0.01	31.45 ±0.02	123.41 ±0.03	17.67 ±0.01
Flu	114.76 ±0.03	28.97 ±0.02	89.34 ±0.04	13.08 ±0.01
Fen	113.05 ±0.01	15.36 ±0.02	78.67 ±0.02	10.26 ±0.02
An	116.66 ±0.03	27.39 ±0.03	94.58 ±0.03	16.07 ±0.03
Fl	114.13 ±0.02	21.23 ±0.03	79.42 ±0.02	13.46 ±0.02
Pir	113.26 ±0.04	17.45 ±0.02	108.12 ±0.02	10.35 ±0.02
B(a)A	117.35 ±0.02	15.34 ±0.02	93.24 ±0.02	10.43 ±0.01
Ch	115.17 ±0.01	14.22 ±0.01	82.58 ±0.03	8.16 ±0.01
B(b)F	110.54 ±0.01	8.09 ±0.02	94.27 ±0.02	3.25 ±0.01
B(k)F	-	-	2.59 ±0.01	1.14 ±0.01
B(a)P	112.45 ±0.03	6.51 ±0.02	74.32 ±0.02	4.32 ±0.01
D(ah)A	-	-	-	-
B(ghi)P	-	-	-	-
IP	110.52 ±0.03	2.72 ±0.01	13.67 ±0.01	-
Total PAHs	1508.54 ±0.29	230.17 ±0.26	1134.61 ±0.34	136.46 ±0.19
Total carc. PAHs	556.03 ±0.08	31.54 ±0.06	267.43 ±0.09	27.3 ±0.04

- not detected

* - samples collected June 15, 2001

- samples collected September 8, 2001

Table 2. Content of PAHs [ng/kg] in samples of cabbage growing in a distance of 5 m or 15 m from the international route (samples collected September 8, 2001, n = 7)

Compound	Distance	
	5 m	15 m
Na	66.01 ±0.02	6.48 ±0.01
Ace	41.91 ±0.02	31.86 ±0.01
Acn	44.22 ±0.03	31.58 ±0.02
Flu	66.53 ±0.01	-
Fen	-	-
An	18.64 ±0.01	2.75 ±0.01
Fl	9.22 ±0.02	2.36 ±0.01
Pir	10.67 ±0.01	-
B(a)A	10.21 ±0.01	4.05 ±0.01
Ch	18.02 ±0.02	-
B(b)F	15.04 ±0.02	-
B(k)F	16.15 ±0.02	5.01 ±0.02
B(a)P	-	-
D(ah)A	11.01 ±0.01	-
B(ghi)P	21.12 ±0.01	2.02 ±0.01
IP	10.15 ±0.01	1.09 ±0.01
Total PAHs	358.90 ±0.22	87.20 ±0.11
Total carc. PAHs	101.17 ±0.09	12.17 ±0.04

- not detected

vegetation.

A total PAHs concentration in soil and cabbage samples collected in a distance of 5 m from the road was 1.51 μg/kg (total carcinogenic PAHs = 556.03 ng/kg) and 358.90 ng/kg (total carcinogenic = 101.17 ng/kg), respectively. On the other hand, total PAHs in samples collected in a distance of 15 m from the road was 136.46 ng/kg for soil (total carcinogenic = 27.30 ng/kg) and 87.20 ng/kg for cabbage (total carcinogenic = 12.17 ng/kg, sea Tab. 1, 2, Fig. 1, 2). The cited papers reported comparable data [5, 12, 18, 19].

In the case of carrot and parsley the concentration of PAHs is higher in aboveground parts, than in underground ones (Tab. 3, 4). A high concentration of hydrocarbons detected

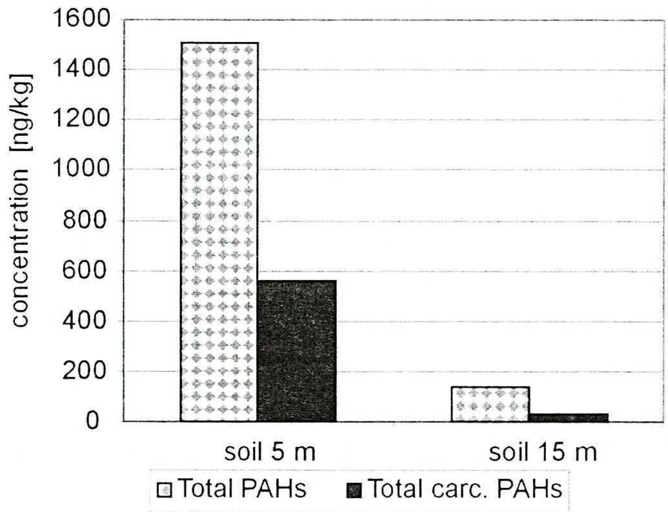

Fig. 1. Concentration of total 16 PAHs and total carcinogenic PAHs in soil

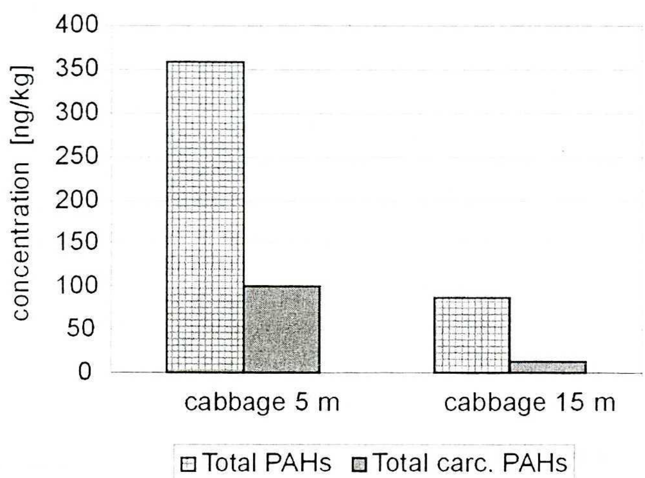

Fig. 2. Concentration of total 16 PAHs and total carcinogenic PAHs in cabbage

Table 3. Content of PAHs [ng/kg] in samples of carrot growing in a distance of 15 m from the international route (samples collected September 8, 2001, n = 7)

Compound	Plant organs	
	Leaves	Root
Na	10.74 ±0.02	10.23 ±0.03
Ace	24.93 ±0.01	17.13 ±0.03
Acn	9.85 ±0.01	-
Flu	13.92 ±0.02	9.51 ±0.01
Fen	18.81 ±0.03	10.52 ±0.02
An	4.46 ±0.01	3.43 ±0.01
Fl	-	1.21 ±0.01
Pir	1.59 ±0.01	3.01 ±0.02
B(a)A	0.25 ±0.02	0.11 ±0.01
Ch	0.23 ±0.01	0.12 ±0.01
B(b)F	0.16 ±0.01	1.11 ±0.02
B(k)F	0.23 ±0.02	-
B(a)P	-	-
D(ah)A	-	-
B(ghi)P	-	0.12 ±0.01
IP	-	0.11 ±0.01
Total PAHs	84.97 ±0.17	56.61 ±0.17
Total carc. PAHs	0.67 ±0.04	1.57 ±0.03

- not detected

Table 4. Content of PAHs [ng/kg] in samples of parsley growing in a distance of 15 m from the international route (samples collected September 8, 2001, n = 7)

Compound	Parsley		Cucumber
	Leaves	Root	
Na	15.15 ±0.03	3.83 ±0.01	4.54 ±0.02
Ace	34.72 ±0.02	1.21 ±0.01	8.67 ±0.01
Acn	7.60 ±0.02	1.00 ±0.02	8.88 ±0.01
Flu	20.85 ±0.01	1.44 ±0.02	8.89 ±0.02
Fen	31.14 ±0.02	6.48 ±0.02	8.54 ±0.03
An	11.82 ±0.01	7.61 ±0.01	2.06 ±0.01
Fl	15.74 ±0.02	2.78 ±0.01	6.22 ±0.03
Pir	-	6.81 ±0.03	8.58 ±0.02
B(a)A	7.57 ±0.01	2.69 ±0.02	4.93 ±0.03
Ch	1.42 ±0.01	1.81 ±0.01	2.35 ±0.04
B(b)F	-	-	1.22 ±0.03
B(k)F	-	-	4.98 ±0.01
B(a)P	-	-	-
D(ah)A	-	-	-
B(ghi)P	-	-	1.44 ±0.03
IP	1.47 ±0.01	-	-
Total PAHs	147.48 ±0.16	29.66 ±0.16	71.30 ±0.29
Total carc. PAHs	10.46 ±0.02	4.50 ±0.01	14.92 ±0.11

- not detected

in vegetable samples, especially in aboveground parts, shows that compounds capable of adsorption on surfaces of leaves contaminated the air, e.g. in samples of parsley the concentration of anthracene in leaves was 11.82 ng/kg, in roots 7.61 ng/kg, the concentration of chrysene – 1.42 ng/kg in leaves (total PAHs = 147.48 ng/kg) and 1.81 ng/kg in roots (total PAHs = 29.66 ng/kg), respectively in carrot samples the concentration of benzo(a)anthracene in leaves was 0.25 ng/kg (total PAHs = 84.97 ng/kg), whereas in roots 0.11 ng/kg (total PAHs = 56.61 ng/kg, sea Fig. 3, 4). An analysis of plant samples shows that similarly to other

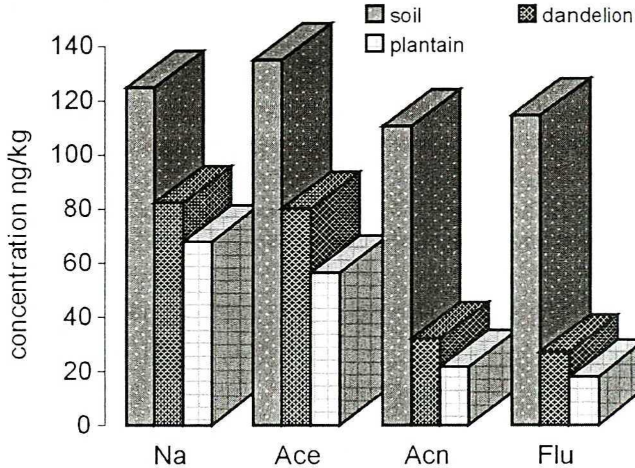

Fig. 3. Comparison of sorption of selected PAHs contents in soil and plants samples

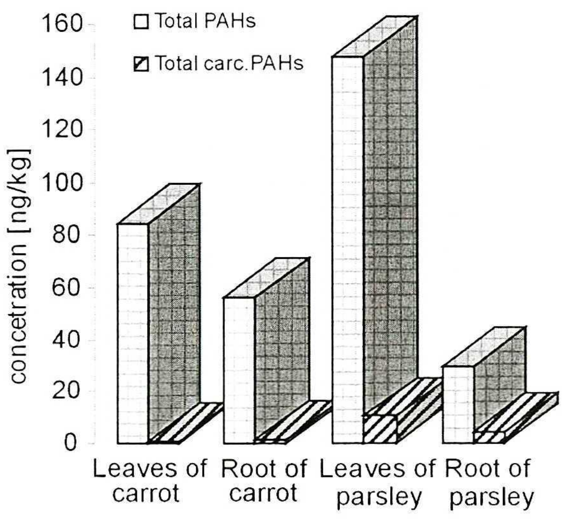

Fig. 4. Concentration of total 16 PAHs and total carcinogenic PAHs in root and leaves in carrot and parsley

samples, they contain a wide range of PAHs, practically only few compounds were not detected (B(a)P, B(ah)A, B(ghi)P). The concentrations of PAHs are not high (range 82.43 – 1.70 ng/kg, Tab. 5). Strange us enough that cucumber contained 13 of 16 PAHs in the amount bet tan 1.22 ng/kg for B(b)F to 8.89 ng/kg for Fl. Coltsfoot contains the lowest number of PAHs and in the lowest concentrations (total PAHs = 28.43 ng/kg, Tab. 5).

Table 5. Content of PAHs [ng/kg] in samples of the leaves of wild plant species in a distance of 5 m from the international route (samples collected September 8, 2001, n = 7)

Compound	Dandelion	Plantain	Coltsfoot
Na	82.43 ±0.05	67.92 ±0.03	2.36 ±0.01
Ace	80.28 ±0.04	56.54 ±0.02	5.12 ±0.02
Acn	31.97 ±0.04	21.63 ±0.02	-
Flu	27.22 ±0.05	17.96 ±0.03	-
Fen	16.76 ±0.03	23.84 ±0.01	1.70 ±0.02
An	11.15 ±0.01	17.92 ±0.04	7.22 ±0.03
Fl	10.13 ±0.02	12.85 ±0.02	-
Pir	-	15.40 ±0.01	-
B(a)A	19.92 ±0.02	-	-
Ch	11.88 ±0.01	15.09 ±0.03	6.70 ±0.02
B(b)F	17.64 ±0.01	10.01 ±0.02	-
B(k)F	11.38 ±0.02	13.18 ±0.04	5.33 ±0.02
B(a)P	-	-	-
D(ah)A	12.23 ±0.02	-	-
B(ghi)P	-	13.56 ±0.01	-
IP	11.21 ±0.03	-	-
Total PAHs	344.20 ±0.35	285.90 ±0.28	28.43 ±0.12
Total carc. PAHs	84.26 ±0.09	51.84 ±0.10	12.03 ±0.04

- not detected

Analysis of diagrams of PAH sorption by plants confirms high ration of adsorption of these compounds by soil, especially by humic layer. Monitoring of impurity level is important in the case of these plants because they are consumed or used in therapeutics.

It is remarkable that in majority of samples, acenaphthylene and acenaphthene were the main components. These compounds are not carcinogens [12, 21]. It is necessary to emphasize that the most carcinogenic benzo(a)pyrene was detected only in few samples. PAHs can be synthesised by soil organisms or can be formed during transformation of organic substances into peat and lignite. Therefore, these compounds should be treated as impurities of autochtonic as well as allochtonic origin [22, 27].

The obtained results show, in comparison with similar investigations by other authors, low pollution of the analysed soil and plants [4, 6, 22]. In his investigation Blumer [6] found between 40 and 130 micrograms of benzo(a)pyrene in one kg of dry substance. Lower contents of PAHs determined Borneff and Kunte in soil, southward of Darmstadt, also remote from factories and settlements: concentration of benzo(a)pyrene were under few μg/kg, concentrations of benzo(g,h,i)perylene were between 10 and 70 μg/kg and benzo(b)fluoranthene – up to 110 μg/kg [22]. In Norway, in area where concentration of PAHs was on the level of natural background, Aamot and co-workers found up to 128 μg/kg of fluoranthene and up to 58 μg/kg of benzo(a)pyrene. According to Dutch directives, prepared for a program of recultivation of contaminated areas, standard soil regarded as uncontaminated, may contain up to 100 μg/kg of fluoranthene and up to 100 μg/kg of benzo(a)pyrene. A total content of all PAHs can not exceed 1000 μg/kg [1, 3, 23, 25]. It is worth adding, that investigations carried out in seven allotments in Cracow showed total content of six hydrocarbons (including benzo(a)pyrene, fluoranthene and benzo(g,h,i)perylene) in range the between 382 and 3411 μg/kg [2, 13, 29]. A total concentration of cancerogenous PAHs in investigated soil was 556.03 ng/kg, and total concentration of all PAHs 1.51 μg/kg, whereas according to Polish law permitted concentration in soil of group A is 1000 μg of dry weight. In the investigated soil neither this value was not exceeded nor values permitted for individual PAHs Benzo(a)pyrene, the most cancerogenous compound existed in concentration 112.45 ng/kg, only. Its permitted concentration for unpolluted soil is 20 μg/kg of dry weight. These data show that medium results of PAH concentration in investigated samples do not exceed permitted values and according to the literature are within limits of natural concentration.

CONCLUSIONS

A comparison of the concentration of PAH in soil and plants samples collected in different distances from the road, showed unambiguously that concentration of these compounds decreases with the distance in soil an in whole ecosystem. Although the differences in PAH concentrations, between species are relatively small this study clearly shows that plant architecture and leaf hairs influence the dry deposition of PAHs.

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