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Soil sealing degree as factor influencing urban soil contamination with polycyclic aromatic hydrocarbons (PAHs)

Abstract: The objective of the study was to determine role of soil sealing degree as the factor influencing soil contamination with polycyclic aromatic hydrocarbons (PAHs). The study area included four sampling sites located within the administrative boundaries of the Toruń city, Poland. Sampling procedure involved preparing soil pits representing three examples of soil sealing at each site: non-sealed soil as a control one (I) and two degrees of soil sealing: semi-pervious surface (II) and totally impervious surface (III). Together with basic properties defined with standard procedures (particle size distribution, pH, LOI, content of carbonates) content of selected PAHs was determined by dichloromethane extraction using gas chromatography with mass spectrometric detection (GC-MS). Obtained results show that urban soils in the city of Toruń are contaminated with polycyclic aromatic hydrocarbons. Soil sealing degree has a strong influence on the soil contamination with polycyclic aromatic hydrocarbons. Totally sealed soils are better preserved from atmospheric pollution including PAHs. Combustion of grass/wood/coal was the main source of determined PAHs content in examined soils.

Keywords: technogenic soils, urban soils, Ekranic Technosols, soil sealing, soil contamination, PAHs

INTRODUCTION

Polycyclic aromatic hydrocarbons (PAHs) are organic compounds composed of two or more fused aromatic benzene rings as part of their structure. They can arrive into the environment components, like soils, from both natural, anthropogenic, and technogenic sources (Tsibart and Gennadiev 2012). PAHs belong to a group of persistent organic pollutants (Vacha et al. 2010) due to their chemically stable structure and low bioavailability (Lamichhane et al. 2016). Despite the fact that there is a large amount of research on the contamination of different landscape components (including soils), many problems related to the origin and behavior of PAHs in the environment are still unclear (Tsibart and Gennadiev 2012).

Soil sealing has a significant impact on the functioning of soil. This problem often affects fertile agricultural lands, increases the flood risk and water deficiency, and contributes to global warming (Couch et al. 2007, Scalenghe and Ajmone Marsan 2009, EU Technical Report 2011). Sealing of soils is listed as one of the main causes of soil degradation in the European Union together with soil erosion, decrease of soil organic matter content, soil compaction, etc. (European Commission 2006). There are two examples of the degree of soil sealing (Nestroy 2006).

First of them is the total imperviousness (caused by solid concrete or asphalt), the second is sealing with semi-pervious surface (e.g. cobblestones, concrete paving slabs or openwork concrete structures) which allow partial penetration of water and air.

The objective of the study was to determine the influence of soil sealing degree on soil contamination with polycyclic aromatic hydrocarbons (PAHs).

MATERIAL AND METHODS

The study area included four (A, B, C, D) sampling sites located within the administrative boundaries of the Toruń city, Poland (Fig. 1). Detailed characterization of each sampling site is presented in Table 1.

Sampling procedure involved preparation of soil pits representing three examples of soil sealing at each site. These examples are: non-sealed soil as a control one (I) and two degrees of soil sealing: semi-pervious surface (II) and totally impervious surface (III). At sampling site “A” only the control profile (I) and the profile sealed with the semi-pervious surface (II) have been studied. Field works were conducted between April of 2012 and September of 2013.

Samples from sealed soil were collected from topmost horizon (top 10 cm of it, if thicker) that survived the process of pavement construction and

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<http://www.degruyter.com/view/j/ssa> (Read content)

FIGURE 1. Location of the study area

TABLE 1. Sampling sites and soil profile descriptions

Profile No.	Sampling depth	Site description (administrative city section; street; site characterization)	
A I	12–21	Bielany city section;	roadside greenbelt covered with grass: control for profile A II
A II	12–21	at the Krzemieniecka Street – street being a part of the NCU Campus	the roadside sealed with concrete slabs sized 1x0.75 m with 5x10 cm holes
B I	14–24	Góry Zajęcze – forested area (sand dunes covered mainly with Scots pines) between the Bielany and the Chelmińskie Przedmieście city sections;	roadside greenbelt covered with grass: control for profiles B II and B III
B II	11–17	close to the Bema and the Gałczyński streets crossing	sidewalk sealed with small concrete sets (so called "polbruk")
B III	11–17		walkway sealed with bituminous pavement
C I	9–19	Na Skarpie city section;	roadside greenbelt covered with grass: control for profiles C II and C III
C II	7–17	at the Lubicka Street – eastern part of the major thoroughfare of the eastern part of the city	walkway sealed with solid concrete slabs sized 0.3x0.3
C III	8–18		walkway sealed with bituminous pavement
D I	27–37	Podgórz city section;	grass plot; control for profile D II and D III
D II	27–37	at the Stepowa Street – street close to the major thoroughfare of the southern part of the city	access street; sealed with concrete slabs 1x1m with holes 5x10 cm,
D III	27–37		access street; sealed with solid concrete slabs

from the corresponding horizons in case of non-sealed soils. Detailed sampling depths were described in Table 1. In total, 11 samples were collected for the laboratory analysis. The following soil properties were determined: particle size distribution by the sieve method and the hydrometer method (the Bouyoucos aerometric method modified by Cassagrande and Prószyński), the names of the texture classes were given in line with the classification of Soil Science Society of Poland – PTG 2008 (Polskie Towarzystwo Gleboznawcze 2009), pH potentiometrically for the soil-to-solution ratio of 1:2.5 using H_2O and 1 mol·dm³ KCl as the suspension medium, approximate content of organic matter was determined using loss on igni-

tion (LOI) analysis (each sample was ignited at temperatures of 550° in a muffle furnace), and content of carbonates by the Scheibler volumetric method. Content of selected PAHs was determined by dichloromethane extraction using gas chromatography with mass spectrometric detection (GC-MS) according to the US GEOLOGICAL SURVEY procedure (Furlong et al.1996) with following detection limits and mean method recovery (compound – detection limit/method recovery, both in $\mu g \cdot kg^{-1}$): acenaphthylene – 0.2/1.02, acenaphthene – 0.3/0.63, fluorene – 0.3/0.83, phenanthrene – 0.2/0.99, anthracene – 0.3/0.91, fluoranthene – 0.3/0.97, pyrene – 0.3/0.90, benzo[a]anthracene – 0.3/1.04, chrysene – 0.2/1.01,

FIGURE 2. The differences in some of the investigated soil properties depending on the soil sealing degree

benzo[b]fluoranthene – 0.5/1.04, benzo[k]fluoranthene – 0.3/1.10, benzo[e]pyrene – 0.3/0.97, benzo[a]pyrene – 0.4/0.97, perylene – 0.5/0.97, indeno[1,2,3-cd]pyrene – 0.3/1.20, dibenz[a,h]anthracene – 0.8/1.27, benzo[ghi]perylene – 1.0/1.20.

Box-and-whisker plots, (Fig. 2) were drawn using Statistica 9.0 software (Statsoft Inc.) to show the relationships between some soil parameters and soil sealing degree.

RESULTS AND DISCUSSION

Basic properties of the soils studied

In general all of the analysed samples were characterized by the sandy texture (sandy loam in case of sample B I) (Table 2). Therefore, the sealed soil

profiles (A II, B II, B III, C II, C III) meet the criteria of Ekranic Technosols (Arenic) according to WRB (IUSS Working Group WRB 2015). According to the Polish Soil Classification (PSC 2011) they were classified as Ekranosols (in Polish: ekranosole). Reference soil profiles (A I, B I, C I, D I) are characterized by morphology features of deformed (truncated or buried with relocated material) Brunic Arenosols (Rusty soils – gleby rdzawe; PSC 2011) or weakly developed Podzols (Podzolic soils – gleby bielcowe; PSC) which dominated in the area of river terraces in place when the city is located (Bednarek and Jankowski 2006, Charzyński et al. 2013a).

Approximate content of soil organic matter measured based on LOI varied from 0.28 to 6.63% (Table 2) with no significant trend according to the soil sealing degree. This result is in opposite to other

TABLE 2. Selected physical and chemical properties of the investigated samples

Profile No.	Depth (cm)	LOI (%)	pH		CaCO ₃ (g·kg ⁻¹)	Percentage share of fraction (mm)				Text. class*
			H ₂ O	KCl		>2	2–0.05	0.05–0.002	<0.002	
A I	12–21	6.63	7.2	7.1	6.6	0.4	98	2	0	m. sand
A II	12–21	4.55	6.9	6.6	2.4	1.3	99	1	0	m. sand
B I	14–24	3.17	7.8	7.1	12.2	14.4	62	30	8	g. s. loam
B II	11–17	0.75	7.4	6.5	2.5	6.7	91	7	2	g. m. sand
B III	11–17	4.46	6.9	6.3	<2	3.1	99	1	0	m. sand
C I	9–19	1.40	7.9	7.8	6.8	4.4	99	1	0	m. sand
C II	7–17	0.50	7.8	7.7	4.5	7.3	99	1	0	g. m. sand
C III	8–18	2.08	7.6	7.5	6.2	5.7	100	0	0	g. m. sand
D I	27–37	0.28	8.6	8.4	8	4.5	99	1	0	c. sand
D II	27–37	0.58	8.4	8.3	9.4	4.0	100	0	0	c. sand
D III	27–37	0.46	8.4	8.3	11.9	5.7	99	1	0	g. m. sand

* Textural class: m. – medium, c. – coarse, g. – gravelly, s. – sandy.

reported findings that soil sealing has strong influence on the organic carbon concentration (e.g. Pouyat et al. 2006, Churkina et al. 2010), as well as example of different studies from the Toruń city concerning the impact of soil sealing degree on microbial biomass and enzymatic activity (Charzyński et al. 2013b, Piotrowska-Długosz and Charzyński 2015). The reaction of soils was alkaline in all analyzed samples. Differentiation of the pH values, as well as the CaCO_3 content was not connected with the soil sealing as in case of the organic matter content (LOI) (Table 2, Fig. 2).

PAHs content in studied soils, its variability and origin

PAHs content in analyzed soils was characterized by large variability. It ranged from 73 to 5907 $\mu\text{g}\cdot\text{kg}^{-1}$ for the sum of the determined PAHs (Table 3). Eight out of nine compounds (beside the naphthalene) from the list defined in the Regulation the Polish Minister of Environment (Regulation of the Minister of Environment 2002) were examined in this study. Defined limits were exceeded for 46 individual cases and for 5 out of 11 total samples in case of calculated sum of the listed PAHs for the depth of 0–0.3 m in the built-up and urban areas. Content of benzo[a]pyrene exceeded the allowable limits to the utmost, even more than 15 times more in case of sample B I (Table 3).

Sum of the determined PAHs and benzo[a]pyrene content shows a significant impact of the soil sealing degree on the soil contamination with polycyclic aromatic hydrocarbons (Fig. 2). Concentration of these harmful compounds clearly decrease with the soil sealing degree as the totally sealed soils were the least, and non-sealed control ones were the most contaminated. Therefore, despite the fact that soil sealing is without any doubt negative phenomenon some positive impact could be also present. In these case it is the suppressing of the atmosphere originated pollution. The positive aspect of some soil degrading factors e.g. soil erosion leading to the preservation of the organic soils with the colluvium was reported already (e.g. Smólczyński 2006, Mendyk et al. 2015).

Fluoranthene and pyrene are dominating compounds when taking into accounts the means of the results obtained for all of the analyzed samples (Table 3). Similar results were obtained for the urban soils of London (Vane et al. 2014). In case of some particular instances this situation could be different. These examples were samples characterized by domination of benzo[b]fluoranthene and benzo[ghi]perylene in case of A II sample and indeno[1,2,3-cd]pyrene and benzo[ghi]perylene for B I, respectively. All of these five compounds are listed as the pyrolytic compounds

TABLE 3. Concentration ($\mu\text{g}\cdot\text{kg}^{-1}$) of selected polycyclic aromatic hydrocarbons in the investigated samples

Profile No.	acenaphthylene	acenaphthylene	fluorene	phenanthrene	anthracene*	fluoranthene*	pyrene	benzo[a]anthracene*	chrysene*	benzo[b]fluoranthene	benzo[k]fluoranthene	benzo[e]pyrene	benzo[a]pyrene*	perylene	indeno[1,2,3-cd]pyrene	dibenz[a,h]anthracene	benzo[ghi]perylene*	17 PAHs	8 PAHs*
A I	20	2	3	45	20	333*	313	226**	210*	380	183*	281	346*	99	353	76	371*	3261	1734*
A II	13	<1	1	18	6	215*	176	114*	98	120	65	82	101*	14	88	19	82	1213	699
B I	6	47	41	568*	140*	1024*	874	463*	427*	416	260*	317	465*	119	333	52	355*	5907	3702*
B II	4	1	3	63	13	205	185	107*	108*	145	78	110	139*	33	126	20	135*	1475	848
B III	2	<1	<1	6	2	39	41	26	34	54	31	46	43*	7	61	6	62	462	243
C I	7	5	6	71	12	153*	131	69	64	69	36	57	64*	17	52	10	55	878	524
C II	6	2	2	27	6	98	91	51	51	69	35	58	58*	15	58	10	61	698	387
C III	<1	<1	<1	5	<1	10	9	4	6	6	3	5	<3	<3	<5	<5	<5	73	37
D I	11	9	13	231*	19	553*	418	184*	219*	231	119*	169	200*	31	172	41	175*	2795	1700*
D II	13	7	10	168*	17	484*	379	177*	200*	242	111*	165	193*	30	169	40	177*	2582	1527*
D III	18	4	8	130*	28	412*	351	195*	210*	295	131*	229	229*	54	239	57	261*	2795	1596*
Mean	9	7	8	121	24	321	270	147	148	184	96	138	167	38	151	31	158	2018	1182

* PAHs listed in the Regulation of the Polish Minister of Environment (2002) and values exceeding the limits defined in these document.

coming from incomplete combustion of the oil and its products (Tsibart and Gennadiev 2012) or the combustion of fossil fuels in general (McCready et al. 2000, Vane et al. 2007, Vane et al. 2014).

PAH isometric ratios were also used in order to characterize the PAH main source in soil and sediments (Vane et al. 2013, 2014). Following ratios were used in these paper: phenanthrene/anthracene compared with fluoranthene/pyrene (Budziński et al. 1997, Vane et al. 2014) and benzo[a]anthracene/benzo[a]anthra-

FIGURE 3. Isometric ratio plot of Phenanthrene/Anthracene and Fluoranthene/Pyrene

PC* - petroleum combustion; MS* - mixed sources

FIGURE 4. Isometric ratio plot of Benzo[a]anthracene/Benzo[a]anthracene+Chrysene and Fluoranthene/Fluoranthene+Pyrene

cene+chrysene compared with fluoranthene/fluoranthene+pyrene (Yunker et al. 2002, Vane et al. 2014) (Fig. 3 and 4). Values of the first two ratios shows that almost all of the samples were contaminated with PAH coming from pyrogenic sources (Fig. 3). This is confirmed with results of another two ratios (Fig. 4) that suggest the combustion of grass/wood/coal as the main origin of PAHs in the soils studied. On the other hand we can not exclude an illegal combustion of synthetic materials wastes e.g. plastic bottles as the other source of PAHs. Relatively high concentration of benzo[ghi]perylene compared to benzo[a]pyrene (values exceeding 3.14) (Vane et al. 2014, after Creaser et al. 2007) indicates the traffic exhaust as the major source of pyrolytic source. The values of this ratio ranged from 0.81 to 1.67, therefore it stand together with previous analyzed plots. All of the results are similar to those coming from the research conducted for the city of London (Vane et al. 2014), Miami (Banger et al. 2010), and New York (Marquez-Bravo et al. 2016). However this method alone is not definitive and could be a source of uncertainty (Vane et al. 2014 after Katsoyiannis et al. 2007, 2011).

There are significant differences in PAHs concentration between individual study sites in Toruń city. The highest content of analyzed compounds was observed on the Stepowa street (site D), in the southern part of the city. This site is also characterized by the smallest variation in PAHs content according to the soil sealing degree. Relatively high concentrations were also determined in samples collected at the sites A and B. These three sites (A, B, and D) were distinguished by location within or in the close vicinity to the single-family residences, whereas the site C (Table 3) was located in the largest block of flat housing area in the city. This fact, together with results explaining the grass/wood/coal combustion as the main source of PAHs in soils examined suggest that the individual heating systems of single-family residences are the main origin of these compounds in studied sites. The atmospheric source of the PAHs is obvious when analyzing the content of benzo[a]pyrene in the air suspended particles at kujawsko-pomorska zone and for the city of Toruń that in the period of 2009–2013 (WIOŚ Bydgoszcz 2014).

CONCLUSIONS

1. Urban soils in the city of Toruń were contaminated with PAHs. Determined concentrations exceeded the defined limits for the surface soil horizons of the urban areas (Regulation of the Polish Minister of Environment 2002), especially for the highly harmful benzo[a]pyrene.

2. Soil sealing degree has a strong influence on the soil contamination with polycyclic aromatic hydrocarbons. Totally sealed soils are preserved from atmospheric pollution including PAHs.
3. The main source of determined PAHs content in examined soils was the combustion of grass/wood/coal, most probably coming from the individual heating systems of the single-family residences.

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Stopień przykrycia gleby jako czynnik wpływający na zanieczyszczenie gleb miejskich wielopierścieniowymi węglowodarami aromatycznymi (WWA)

Streszczenie: Celem badań było określenie roli stopnia zasklepienia gleb jako czynnika wpływającego na ich zanieczyszczenie wielopierścieniowymi węglowodarami aromatycznymi (WWA). Obszar badań obejmował cztery lokalizacje poboru próbek w obrębie granic administracyjnych miasta Torunia. Próbki zostały pobrane z profili glebowych reprezentujących trzy przykłady przykrycia gleb: glebę nieprzykrytą jako profil kontrolny (I) oraz dwa stopnie zasklepienia gleb: powierzchnią częściowo przepuszczalną (II) i nieprzepuszczalną (III). Oprócz podstawowych właściwości oznaczonych za pomocą standardowych procedur (uziarnienie, pH, straty prażenia, zawartość węglanów) została oznaczona zawartość wybranych WWA po ekstrakcji dichlorometanem za pomocą metody chromatografii gazowej z detekcją spektrometrią mas. Uzyskane wyniki pozwalają stwierdzić, że gleby miejskie Torunia są zanieczyszczone WWA. Stopień zasklepienia gleb ma silny wpływ na poziom tego zanieczyszczenia. Gleby całkowicie przykryte są lepiej chronione przed zanieczyszczeniami pochodzenia atmosferycznego, w tym wielopierścieniowymi węglowodarami aromatycznymi. Spalanie trawy/drewna/węgla zostało zdiagnozowane jako główne źródło oznaczonych zawartości WWA w badanych glebach.

Słowa kluczowe: gleby technogeniczne, gleby miejskie, Ekranic Technosols, zasklepienie gleb, zanieczyszczenie gleb, WWA