

THE POLYCYCLIC AROMATIC HYDROCARBONS CONTENT IN SELECTED SILTY SOILS

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ZAWARTOŚĆ WIELOPIERŚCIENIOWYCH WĘGLOWODORÓW AROMATYCZNYCH (WWA) W WYBRANYCH GLEBACH PYŁOWYCH

Celem pracy było określenie zawartości wielopierścieniowych węglowodorów aromatycznych w zależności od intensywności i sposobu użytkowania gleby. Do badań wytypowano dwa typy gleb, tj. czarnoziem nie zdegradowany wytworzony z lessu oraz madę rzeczna właściwą wytworzoną z utworów pyłowych. W każdej objętej badaniami glebie wybrano po pięć obiektów: sady jabłoniowe, chmielniki, pola uprawne, trwałe użytki zielone i naturalne ekosystemy leśne. Do badań pobierano próbki z głębokości 0–10, 10–20 oraz poniżej 35 cm. W uzyskanym materiale oznaczono zawartość 16 WWA metodą HPLC-UV oraz zawartość węgla organicznego. Zawartość wielopierścieniowych węglowodorów aromatycznych wyraźnie zależała zarówno od typu gleby, jak również od sposobu jej użytkowania. Jedyne w przypadku dwóch obiektów (chmielnika i trwałego użytku zielonego) zarówno czarnoziem jak też mada rzeczna charakteryzowały się najwyższą zawartością WWA. Wpływ sposobu użytkowania na zawartość zarówno sumy WWA jak też poszczególnych związków wyraźniej zaznaczył się w czarnoziemie aniżeli w madzie rzecznej. Migracja WWA w głąb profilu glebowego podobnie jak zawartość tych związków zależała od typu gleby i sposobu użytkowania. Zarówno jednak w madzie rzecznej jak też czarnoziemie obserwowano wzrost udziału naftalenu wraz z obniżaniem się głębokości. Efekt wypłukiwania zanieczyszczeń w głąb profilu zaznaczył się jednak wyraźniej w przypadku mady rzecznej.

Summary

The aim of the present study was to determine the content of polycyclic aromatic hydrocarbons in relation to the intensity and type of soil management. Two types of soil were selected for the present studies, i.e.: Haplic Phaeozem developed from loess and Eutric Fluvisol originating from silty formations. Five objects were chosen in each of the soils included in the present study, i.e.: apple orchards, hop gardens, arable fields, grassland and natural forest ecosystems. Samples were collected from the depth of 0–10, 10–20 and below 35 cm. In the study material obtained, the content of 16 PAHs was determined by means of the HPLC-UV method. The content of polycyclic aromatic hydrocarbons clearly depended both on the soil type and soil management method. Only in the case of two objects (hop garden and grassland plot) both the Haplic Phaeozem and the Eutric Fluvisol were characterized by the highest PAH content. The influence of soil management method on both the PAH sum and the content of individual compounds was more clearly marked in the Haplic Phaeozem than in the Eutric Fluvisol. PAH migration deeper into the soil profile similarly to their content depended on the soil type and soil management method. However, both in the Eutric Fluvisol and in the Haplic Phaeozem, an increase in naphthalene share was observed with a decrease in depth. Nevertheless, the effect of pollutant leaching deeper into the soil profile was more clearly marked in the case of Eutric Fluvisol.

INTRODUCTION

Polycyclic aromatic hydrocarbons (PAH) belong to a group of persistent organic pollutants [12]. As they originate from diversified sources, they are present in numerous elements in the environment [12, 18]. However, eventually 90% of them end up in the soil [19] where they can be accumulated, volatilized and leached deeper into the soil profile, or/and biodegraded [12, 24]. Accumulation of xenobiotics in the soils can lead to their uptake by plants [17] and, in consequence, penetration into the human food chain. The intensity of the processes described above is determined by soil properties and environmental conditions. There is relatively little information in the literature [5, 11, 21, 23] on the influence of soil management method on the content of polycyclic aromatic hydrocarbons. The studies carried out so far have been limited to comparisons of PAH content levels in arable soils and grassland soils, only. As a result of differentiated soil management methods, specific biochemical conditions relating to the activity of microorganisms (e.g. the effect of rhizosphere) [10] arise alongside changes in soil properties which can favor or limit the range of PAH losses in the soil.

The aim of the present study was to determine the influence of varied, long term soil management methods on the content of polycyclic aromatic hydrocarbons. A possibility of PAH migration deeper into soil profiles was also evaluated with respect to soil management method.

MATERIALS AND METHODS

The study objects consisted of 10 soil profiles located in the area of the following macro-regions: the Wołyńsko-Podolska Upland and the Lublin-Lwów Upland, and in the area of the following mezzo-regions: Grzęda Sokalska (Haplic Phaeozem developed from loess) and Kotlina Chodelska (Eutric Fluvisol developed from silt). Five objects with differentiated long-term soil management methods were chosen in each of the mezzo-regions studied: apple orchards older than 20 years, old hop gardens older than 20 years, at least one set of hundred year old arable fields with any crop sequence, and about 50 year old extensively used grasslands and natural forest ecosystems with trees older than 150 years.

Soil samples for the present studies were collected during the summer season of 2004. The soil was collected from the following sites: orchard – herbicidal rows between trees, hop garden – in the row between karpas; field – from the inter-row of winter wheat; grassland – at random; forest – from places with limited reach of tree roots. Soil samples were collected from two horizons of 0–10 cm and 10–20 cm selected from the arable-humus (Ap) or humus (Ah – in forests) levels and additionally from one horizon outside the influence of the working mechanisms of agricultural machines and tools below 35 cm from levels Ah or AhC. Some physico-chemical properties of soil used in the experiment are presented in tables 1 (soil texture) and 2 (pH).

The soil was dried at room temperature and sieved through a 1 mm sieve. In the samples obtained, the content of polycyclic aromatic hydrocarbons was determined by means of methods presented in other studies [14, 16]. The samples were extracted in an ultrasonic bath (Sonic-3, Polsonic, Poland) with two batches of dichloromethane (2 x 40 cm³). The extract was centrifuged and evaporated to dryness. The residue was then dissolved in a mixture 4 cm³ of acetonitrile : water (1:1 v/v) and purified by solid phase extraction (SPE)

Table 1. The soil texture in investigated soils

Objects	Soil layer	Haplic Phaeozem			Eutric Fluvisol		
		1.0–0.1	0.1–0.02	<0.02	1.0–0.1	0.1–0.02	<0.02
Apple orchards	0–10	3	58	39	31	51	18
	10–20	1	58	41	30.5	42.5	27
	> 35	0.5	57.5	42	27	45	28
Hop garden	0–10	1.5	54.5	44	24.5	43.5	32
	10–20	1	55	44	17	42	41
	> 35	0.5	53.5	46	21	57	22
Arable fields	0–10	0.5	53.5	46	21.5	49.5	29
	10–20	0.5	53.5	46	22	50	28
	> 35	0.5	53.5	46	21.5	48.5	30
Grassland	0–10	2	57	41	22	46	32
	10–20	0.5	56.5	43	16.5	43.5	40
	> 35	0.5	54.5	45	18	42	40
Forest	0–10	3	59	38	6	50	44
	10–20	1	56	43	4	49	47
	> 35	0.5	57.5	42	3	48	49

Table 2. The pH [in KCl] of the investigated soils

Objects	Soil layer	Haplic Phaeozem	Eutric Fluvisol
Apple orchards	0–10	6,1	5,1
	10–20	5,7	4,9
	> 35	6,0	4,2
Hop garden	0–10	6,0	5,1
	10–20	6,7	4,7
	> 35	7,5	5,5
Arable fields	0–10	5,7	5,0
	10–20	5,8	4,5
	> 35	6,9	5,4
Grassland	0–10	5,2	5,7
	10–20	5,6	5,8
	> 35	5,7	6,5
Forest	0–10	5,2	5,0
	10–20	5,4	5,0
	> 35	5,8	5,2

using C₁₈ Octadecyl columns (JT Baker-Mallinckrodt, Germany). For the separation of 16 PAHs, an analytical Spherisorb S5 PAH (250 x 4.6 mm I.D., 5 µg by Schambeck SFD GmbH, Germany) column with chemically bound C18 phase was used. The mobile phase (acetonitrile: water, 82:18, v/v) flow was set to 1 cm³ min⁻¹. Detection was carried out at 254 nm. The

column was installed in a thermostated oven at 31°C (LCO 101, ECOM, Czech Republic). Data acquisition and analysis was performed using the Clarity Lite Chromatographic Station (DataApex, Czech Republic).

All values of PAH were expressed on a dry-weight basis of sample dried for 24 hours at a temperature of 105°C. PAH determination was carried out twice for each sample and the result was calculated as an arithmetical mean of these determinations. The content of total organic carbon (TOC) was calculated by Turin's method as modified by Simakov. A statistical analysis was carried out using ARStat and Statistica 5.0 software.

RESULTS AND DISCUSSION

Total PAHs content – influence of soil type and type of soil management

Figure 1 presents content pattern of polycyclic aromatic hydrocarbons in the soils studied. The total content of the PAH depended on the study object in the case of experiment carried out on soil A (Haplic Phaeozem) and ranged from 168.4 to 764.0; 104.3 to 208.7 and 62.1 to 95.4 µg/kg in the depth 0–10, 10–20 and below 35 cm, respectively. Analyzing the content of the compounds studied in the surface soil horizon, only statistically insignificant differences between them were found in the case of two objects only (S, P). Moreover the lowest content of the PAH sum was noted as well in these latter soils. The highest content of PAHs was observed in the soils of grasslands and hop gardens. With an increase in the depth, a differentiation in the content of the PAH sum related to specific objects studied was observed. At a depth of 10–20 cm, significant differences in the content of the PAH sum as compared to other objects studied were noted in the case of the soils of an apple orchard and hop garden, whereas in the depth of below 35 cm, similarly as at a depth of 0–10 cm, significant differences were related to the soils of grasslands and hop garden.

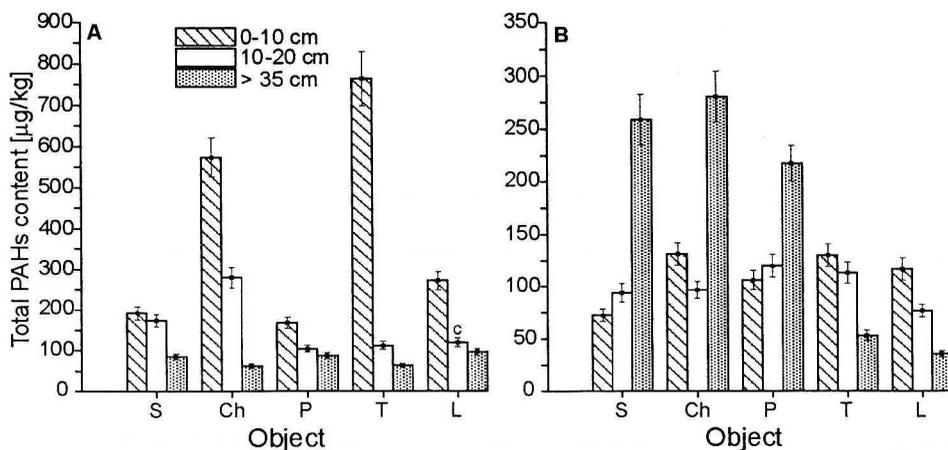


Fig. 1. The total PAHs content in investigated soils

A – Haplic Phaeozem, B – Eutric Fluvisol, S – apple orchards, Ch – hop garden, P – arable fields, T – grassland, L – forest

In the Eutric Fluvisol (soil B), the PAH levels were significantly varied in individual objects, however significant differences were not as numerous as the ones described previously. The content of the PAH sum depended on the depth and took on the following values: 72.5–131.5; 77.0–119.8 and 35.6–280.5 $\mu\text{g kg}^{-1}$, respectively, in the following soil horizons: 0–10, 10–20 and below 35 cm. Similarly to the case described above, the highest values of the PAH sum in the surface soil horizon were noted, similarly as in the case described above, in the soil of the grassland (T) and hop garden (Ch). Slightly lower values (statistically non-significant) were noted in the case of the soil of the arable field (P) and the forest (L). The lowest PAH content was found under the apple orchard (S).

It can be concluded from the data presented in Figure 1 that the influence of soil management on the PAH content clearly depended on the soil type. Each type of soil, subject to its origin and conditions of development, was characterized by specific physical, chemical and biological properties [25]. A significant factor determining the fate of polycyclic aromatic hydrocarbons in the soil is organic matter [22]. Its amount [3], composition and properties are all important [2, 20]. Hence, it can be assumed that transformations and migration of xenobiotics in the soils of differentiated properties, despite the same type of plant breeding, would differ depending on the soil type. The above regularity is confirmed when the content of the PAH sum is compared among the soil types studied.

In the Haplic Phaeozem, which is characterized by a higher content of organic carbon, than the Eutric Fluvisol in most of the objects studied (Fig. 2), a higher PAH content was also determined. In this latter soil, a clearer differentiation of the content of PAH sum in individual objects was also observed in the surface horizon (0–10 cm) whereas in the Eutric Fluvisol the content of the PAH sum in the above mentioned horizon was at a similar level in all objects. The content of TOC in the profiles of Eutric Fluvisol was related to river flooding which covered the layers of “old” turf (sod) and carried alluvial deposits containing high quantities of the humified organic matter [25]. Additionally, inflowing waters can cause stronger leaching of pollutants deeper into the soil profile than in the case of other soils. In literature, the influence of dissolved organic carbon (DOC) on the transportation of hydrophobic organic pollutants including PAHs has been demonstrated [13]. The above phenomenon was also reflected in the results obtained by the present authors who clearly showed a markedly higher PAH content in the depth > 35 cm in the Eutric Fluvisol than in the higher depth of the soil profile in the case of most objects studied. The soils of grassland and forest for which the highest content of total organic carbon was also noted in the surface horizon, were an exception (Fig. 2). Some earlier studies on the migration of PAHs in the soils fertilized with sewage sludge [15] showed that a relatively high content of organic matter in the surface soil horizon (> 3%) can significantly limit transfer of these compounds deeper into the soil profile. The phenomenon of PAH migration into the soil profile shall be described with more detail in the further parts of this study in relation to individual PAHs.

Studies carried out by numerous other authors [4, 6] showed that organic matter can limit bioavailability, and hence biodegradation of polycyclic aromatic hydrocarbons. It can then be assumed that a higher PAH content in the Haplic Phaeozem than in the Eutric Fluvisol resulted from the limitation of their bioavailability which follows from a strong influence of organic matter (beside of elution pollutants from the Eutric Fluvisol as was discussed earlier). In literature, this type of influence is called contaminant sequestration [1].

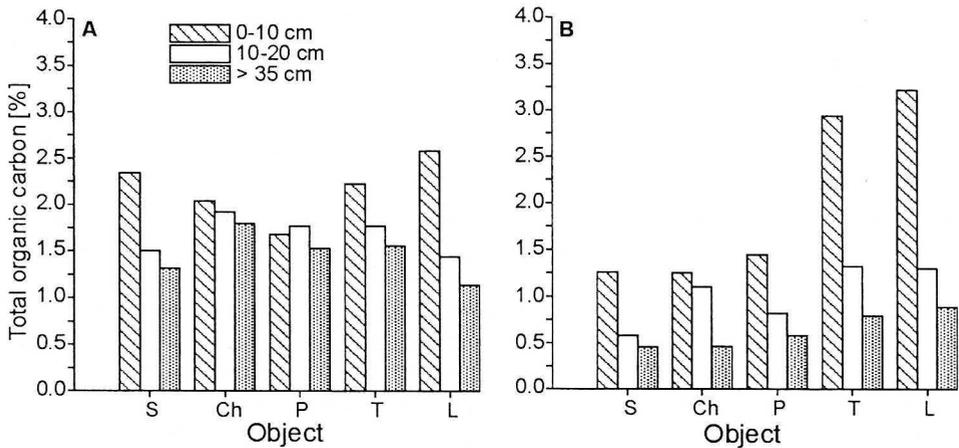


Fig. 2. Total organic carbon content in investigated soils

A – Haplic Phaeozem, B – Eutric Fluvisol, S – apple orchards, Ch – hop garden, P – arable fields, T – grassland, L – forest

Despite various origins and conditions of development of the soils studied in the present experiment, which could influence a varied formation of the PAH content depending on the management method in a given object as suggested earlier, it was found that both in the Haplic Phaeozem and Eutric Fluvisol, the highest PAH content was determined for the soils of hop garden and grassland. The content of organic matter which could explain differences in the PAH content between the soil types studied, cannot be considered as an explanation in this latter case. A low content of organic carbon in relation to other objects excludes its inhibitory influence on the degradation range of the xenobiotics considered. Hence, it can be concluded that different PAH content in individual objects resulted more from the management character and hence formation of specific biochemical conditions favoring degradation of the compounds studied than other factors.

In the literature information on the influence of the soil management method on the PAH degradation range is scarce [9, 11, 23], and the existing studies present divergent data. Data from the LABO report [9] in which information on the determinations of PAH content in the soils of Germany were collected, pointed also to a higher content of the compounds determined in the soils of grasslands than in the arable soils. Maliszewska-Kordybach [11] observed twice higher PAH content in the soils of grasslands than in the arable lands. Similarly, Saison et al. [23] noted a significant influence of agro-technological treatments on the PAH degradation range. The authors mentioned above suggested that conditions created in the arable land relating to a higher availability of oxygen as a result of agro-technical treatments to the arable soil layer can favor the process of biodegradation of the compounds described. The plant species is also important in the description of the soil management method. It is known from the literature [10] that rhizospheres of various plants are characterized by various abilities to stimulate PAH degradation. It can be explained by the lack of influence of the type of plant cultures on the changes in the PAH content observed by other authors [5]. In the present study, in the Eutric Fluvisol there were no significant differences in the content of xenobiotics determined in the surface soil depth of the arable field and grassland either. On the other hand, a significant difference was observed in the Haplic Phaeozem (Fig. 1).

Individual PAHs content in relation to management method and soil depth

Figure 3 presents the percentage contribution of PAH in relation to the number of rings in the Haplic Phaeozem studied. The share of individual compounds clearly depended on the depth and also soil management method. Most often the predominant share was observed in the case of 2-ring naphthalene and, to a lesser degree, 3-ring compounds. Only in the case of two objects (S, P) the highest contribution of naphthalene was noted in all horizon of the soil profile studied. In the remaining objects, an increase in the share of xenobiotics both quantitatively and as related to the number of objects, was observed with an increasing depth. It should be stressed here, that in the case of the humus layer (i.e. 0–10 and 10–20 cm) the PAH composition in most of the objects studied was very similar. The clearest influence of an increasing depth on the content and share of individual xenobiotics was observed in the forest soils. It was related to a significant role of organic matter, as has already been mentioned earlier in this paper, which acts as a strong hydrophobic adsorbent of hydrophobic organic pollutants. A contribution of the most carcinogenic and mutagenic compounds, i.e. 5- and 6-ring, was the highest in the surface soil horizon and was decreasing

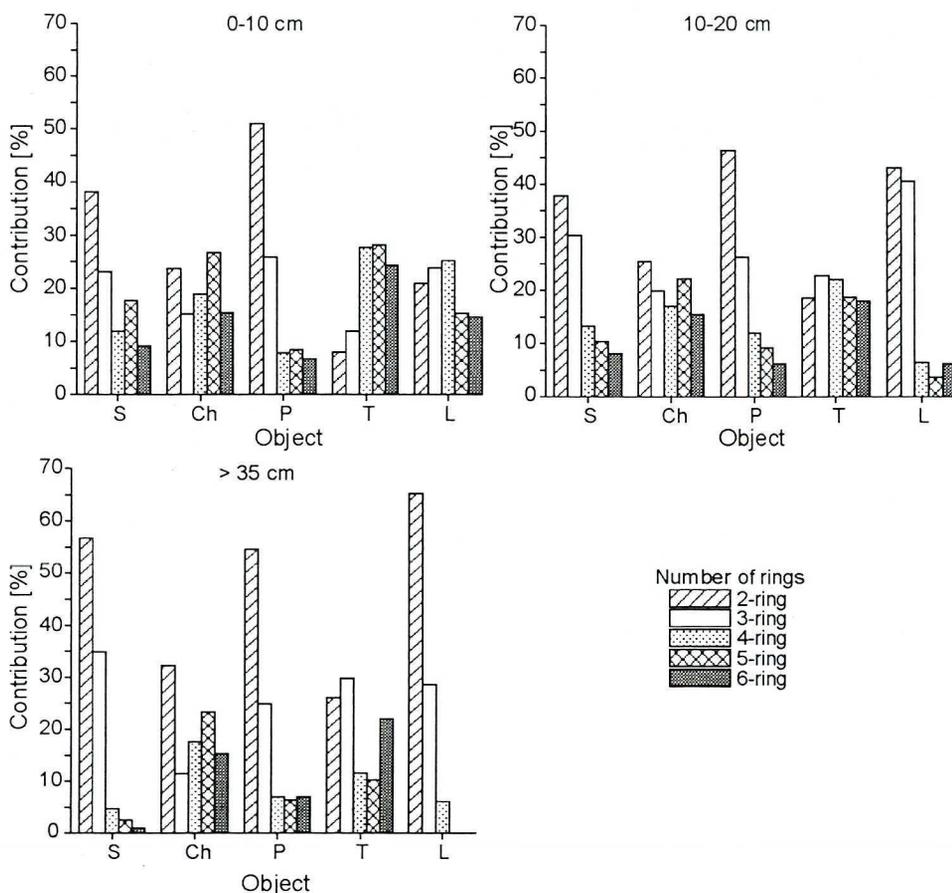


Fig. 3. Contribution of PAHs in Haplic Phaeozem in relation to number of rings (soil A)
S – apple orchards, Ch – hop garden, P – arable fields, T – grassland, L – forest

with an increasing soil depth. The presence of the above mentioned compounds was not noted in the case of forest soils at a depth of below 35 cm.

In the Eutric Fluvisol (Fig. 4), similarly to the soil described earlier, the composition of individual PAHs clearly depended on the utilization method and depth. However, in this soil, differences related to depth were more visible than in the previous case which was probably a result of pollutant leaching by the inflowing waters. It can be seen in a relatively low share of naphthalene in the surface soil horizon and especially high share of 4- and 6-ring PAHs. With an increasing depth, the above situation changes radically and a predominant share of naphthalene can be seen in most objects.

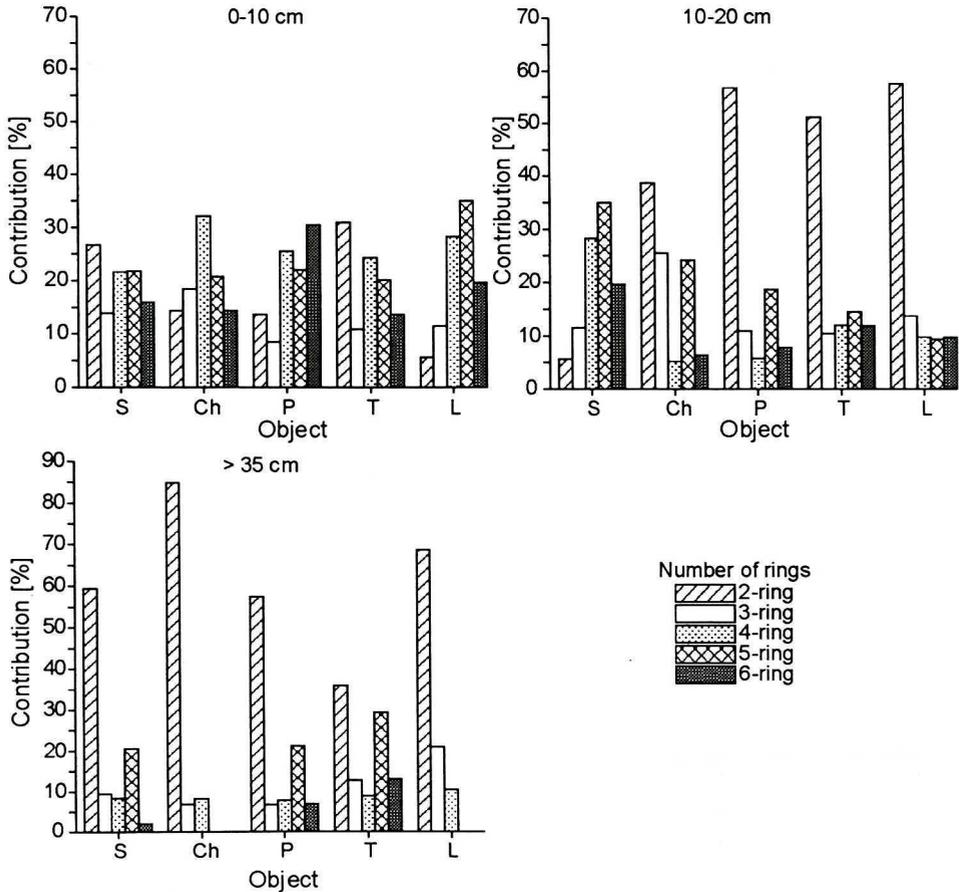


Fig. 4. Contribution of PAHs in Eutric Fluvisol in relation to number of rings (soil B)
S – apple orchards, Ch – hop garden, P – arable fields, T – grassland, L – forest

A high share of naphthalene in the > 35 cm depth, observed in the present experiment together with a low content of 5- and 6-ring PAHs, was determined by the properties of these compounds, and predominantly by their solubility in water as well as values of $\log K_{ow}$ and $\log K_{oc}$ parameters. Naphthalene was characterized by the highest solubility in water among all PAHs analyzed and the lowest affinity to organic carbon expressed as \log

K_{oc} i K_{ow} . The above mentioned properties make migration of this compound deeper into the soil profile easy, whereas 5- and 6-ring PAHs with high molecular weight and considerable affinity to organic carbon are cumulated mainly in the surface layers of the soil profile. In the Eutric Fluvisol, in which the effect of elution is especially intense, the phenomenon of naphthalene elution was very clearly visible. The results obtained in the present study confirm studies by other authors who also observed an increase in the naphthalene share in the deeper soil layers and insignificant share of "heavier" compounds [7, 8, 15, 21].

A method of cluster analysis using Ward's algorithm and Euclidean measure of distance was applied to determine precisely the influence of the utilization method on the individual PAH content. The results presented in Figure 5 showed that the influence of the type of soil utilization on the composition of individual PAHs was more clearly marked in the case of Haplic Phaeozem than in the case of Eutric Fluvisol which was could be seen in the several times bigger ranges of bonding distances. Despite a clearly differentiated method of soil utilization, in the case of Haplic Phaeozem, the soil under apple orchard and arable land show most similarities in the content of individual PAHs. In the case of Eutric Fluvisol, similarities were observed in the soil of arable field and forest as well as in the soils of an orchard and hop garden.

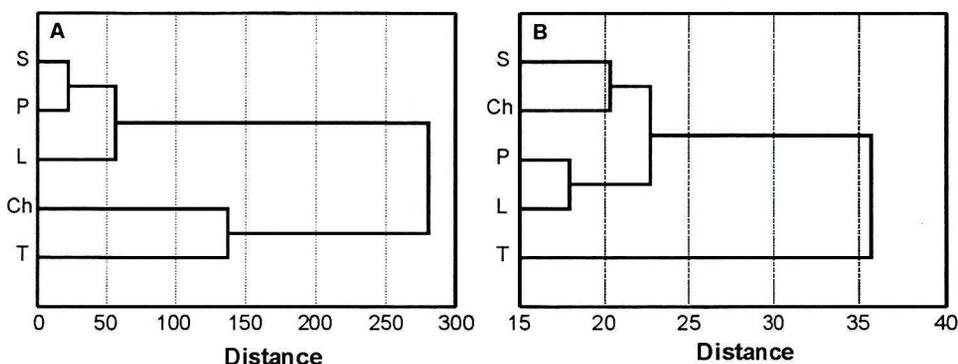


Fig. 5. Tree diagram of cluster analysis of PAH composition in investigated soils
A – Haplic Phaeozem, B – Eutric Fluvisol, S – apple orchards, Ch – hop garden, P – arable fields,
T – grassland, L – forest

CONCLUSION

1. The influence of long term soil management on the content, composition and leaching of PAHs clearly depended on the soil type.
2. In the case of two objects, i.e. hop garden and grassland the PAH content in both soils studied, showed the highest levels. However, both composition and differences in relation to the remaining objects were clearly varied and depended on the soil type.
3. In the surface horizon of the Haplic Phaeozem, 2-ring naphthalene was predominant in most objects, whereas in the Eutric Fluvisol, 4- and 6-ring compounds were predominant.
4. Both in the Eutric Fluvisol and Haplic Phaeozem, an increase in the naphthalene share was noted with an increasing depth. Stronger effects of pollutant leaching deeper into the soil profile were noted in the case of Eutric Fluvisol.

5. The higher levels of PAHs content at some sites were correlated with higher content of organic carbon.
6. The influence of the soil management method on the composition of individual compounds was more clearly marked in the case of Haplic Phaeozem than Eutric Fluvisol.

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