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ACCUMULATION RATE OF POLYCHLORINATED BIPHENYLS (PCBs) IN DANDELION (*TARAXACUM OFFICINALE*) IN THE CONDITIONS OF SOIL CONTAMINATION WITH OIL DERIVATIVES

STOPIEŃ KUMULACJI POLICHLOROWANYCH BIFENYLI (PCB) W MNISZKU LEKARSKIM (*TARAXACUM OFFICINALE*) W WARUNKACH ZANIECZYSZCZENIA GLEBY SUBSTANCJAMI ROPOPOCHODNYMI

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The present study concerns PCBs accumulation in soil at two depths as well as in Taraxacum officinale plants. An analysis of the distribution of different groups of PCBs congeners in plant and soil material revealed the highest content of hexa and hepta-CB fractions. Statistical test of pair comparison has revealed significant differences in accumulation rates for the following pairs: penta-chlorinated congeners are accumulated at a higher rate than tetra-chlorinated ones and hexa- chlorinated congeners are accumulated at a higher rate than penta-chlorinated ones. This phenomenon occurs in the area of railway junctions (areas heavily polluted with oil derivatives).

INTRODUCTION

Polychlorinated biphenyls (PCBs) are organic pollutants frequently found in the environment. Due to their persistence and lipophilic properties, which lead to their bioaccumulation in living organisms, they became a serious threat for natural environments [3–5, 12–14] The fact that it is impossible to utilise them fully and efficiently, as well as extremely high costs associated with this process, have led to a situation when PCBs are illegally disposed of in Western Europe through adding them to used oils and sale to Eastern European countries, including Poland [1]

Despite the fact that there are abiotic and biotic ways of transforming polychlorinated biphenyls in the natural environment, their disappearance is a very slow process and decomposition of the total amount of PCB in the environment does not exceed 1% [15].

Specific, synanthropic plant species can be found near railway lines and junctions. Some researchers claim that there is a group of species associated with railway areas [11]. Dandelion (*Taraxacum officinale*) is one the most common of those species. Being present at all railway areas and having big surface of leaf blades, it fully meets the demands for a potential bioindicator.

The aim of this study was to:

- establish the degree of accumulation of various groups of PCB congeners in parts of dandelion (*Taraxacum officinale*) plants emerged above the ground surface,
- assess changes in the content of different groups of PCB congeners (depending on the chlorination level of the biphenyl system) in soil at two depths (level 1: 0–20 cm, level 2: 20–40 cm)
- assess the suitability of dandelion plants as bioindicators of pollution in railway areas.

EXPERIMENTAL

The research was carried out near railway junctions Iława Glówna and Tarnowskie Góry, as well as in the vicinity of two strategic railway lines: Warsaw – Gdańsk (near the town of Iłowo) and Katowice – Gdynia (two study sites near Warlubie and Laskowice). Control sites were established at a distance of 2 km from study sites.

Analyses concerned soil and parts of dandelion (*Taraxacum officinale*) emerged above the ground which were separated from the roots in a laboratory [10].

Soil and plant samples in railway junction areas were collected in specific sites (washing plant, side track, re-loading ramp, platforms), within a 200 m – wide belt alongside railway tracks. In the vicinity of strategic railway lines, soil and plant samples were collected at a gradient of distances from the track: 0 m, 10 m, 15 m, 30 m, 50 m and 100 m. Altogether, during the two consecutive years of the study, 176 soil samples and 70 plant samples were collected.

Laboratory procedures

50 g of the 2-mm-sieved soil samples or 10 g dried and grounded plants were extracted with hexane : acetone (2:1). 10 ml of each extract was washed 3 times with concentrated H_2SO_4 and afterwards with distilled water. The organic layer was collected and concentrated to 1 ml volume. Hexane extracts were used for further analysis. The analytical method for hexane extracts was high resolution gas chromatography with HP 980 gas chromatograph equipped with ECD detector. Quantitations were performed in capillary column P1: 30 m x 0.53 mm, 2.65 μm Crosslinked Methylsilicone EUM; carrier gas N_2, temperature programme from 80^oC at 30^oC/min up to 190^oC, at 6^oC/min to 280^oC, 280^oC during 1 min., 6^oC/min up to 310^oC, 310^oC during 10 min.

For total PCBs content estimation, the mixture of 5,5 mg/l Arochlor 1242, 11,8 mg/l Arochlor 1254, 4,5 mg/l Aroclor 1260, congeners: 1, 30, 209 was used.

For quantitation of individual PCB homologues the mixture of specific isomers the following congeners were used: 1, 30, 28, 52, 101, 118, 138, 153, 180, 209.

RESULTS AND DISCUSSION

Stain pollution with oil derivatives occurs at study sites only in the soil between tracks and it practically does not exist on the slopes of the railway embankment. Therefore, the statistical analysis concerning relationships between soil and plant pollution with PCB congeners was performed for junction (high organic pollution) and railway line (low pollution) areas independently.

Dandelion is a plant that seems to be a very good bioindicator of organic pollution. It has numerous features which are considered suitable for bioindication. It is an ubiquitous species, it does not have specific soil requirements and it also seems to be resistant to even high soil contamination with different substances. It can be found in most places in railway areas, since it is able to grow back quickly after herbicides are applied. It also has leaves with large blade surface areas where, amongst others, toxic substances can be accumulated.

This study has revealed varying levels of PCB congener pollution of the emerged parts of dandelion. In most of the sites these levels were higher than those in soil. Recorded PCB pollution in plants seems to be the basis for comparative analyses of railway area contamination with these substances.

Railway junctions

While analysing soil and plant contamination with PCB congeners in railway junction areas it was found that PCB content in soil at the depth of 20–40 cm in most samples (75%) ranged from 50 to150 ng/g, while the level of surface layer (0–20 cm) pollution for most samples varied from 100 to 200 ng/g. For emerged plant parts it ranged from 100 to 300 ng/g. It is worth noting that PCB content in plants varied strongly (fig. 1).

Analyses of the content of different groups of PCB congeners in soil and plant material from railway junction areas have revealed that, of all PCB fractions, hepta-CB was the most abundant in soil at two depths, as well as in plants and its mean content was 80–100 ng/g in soil and 50–200 ng/g in plant material. Large differences were found for contamination of deeper soil layers with mono-CB and surface layers with penta-CB. The content of other groups of PCB congeners in soil and plants was rather even, around 50 ng/g. Large variation was found for plant contamination levels with all groups of PCB congeners.

On the basis of professional literature it can be said that some PCB components will persist in soil due to their physical and chemical properties [6, 8].

The period of PCB persistence in soil depends directly on the degree of biphenyl chlorination (less chlorinated PCB congeners have a higher rate of evaporation), the amount of the organic substance in soil and the type of the substrate [2, 7].

Analyses of the percentage content of different groups of PCB congeners for railway junctions (in total) allow us to say that (fig. 2) hepta-CBs had the highest percentage share in most of the material and their respective values were:

1. for soil at 20–40 cm – from less than 10% up to 90% (around 50% in most cases);
2. for soil at 0–20 cm – from less than 10% up to 70% (20–60% in most samples);
3. for plants – from less than 10% up to 70% (10–50% in most samples).

Hexa-CBs also had a significant percentage share in the contamination of plant material (from less than 20% up to 40%, on average) and in their case the situation is reversed – there is less of them in deeper soil layers than near the surface and in plants.

In the case of other groups of PCB congeners no clear trends were found in the percentage share of their content in analysed samples.

High levels of PCBs found in soil from railway junction areas would, therefore, appear to be the result of not only the amount of the pollutant, but also of their dilution in oil derivatives persisting on the surface. Oil and grease form a specific, impermeable membrane on the soil surface and they seem to form a barrier for PCBs, which stops them from penetrating deeper into the ground for a while. After grease, oil and other oil derivatives are decomposed, some of PCB congeners are released.

The observed levels of PCB pollution at two depths in railway junction areas are in accordance with these predictions. For most soil samples, contamination levels in deeper layers were lower than those from surface layers and the variation in the results was smaller. The situation looked rather different in areas adjacent to railway lines. The distribution of soil contamination levels for surface layers did not differ from the distribution of deep layer contamination levels significantly in most of those samples. In those areas, oil derivative spills do not occur at such levels as they do in railway junction areas, so that the distribution of PCB compounds in soil is more or less even.

One should also note the fact that there are rather significant differences in the level of PCB contamination in soil and plants from relative small areas.

Railway lines

The variation in results concerning soil and plant contamination with PCBs near railway lines is much larger in plant material than in soil (from around 50 up to about 800 ng/g for plants to 100–250 ng/g for soil at both depths) (fig. 3).

The distribution of the content of different PCB congener groups in soil and plants from the vicinity of both railway lines formed a characteristic pattern. It was found that the highest concentration levels concerned hepta-CBs in soil and plants (here the variation in the results was the highest). Moreover, mono-CB concentration levels in plant material are also varied.

While comparing the percentage share of different PCB congener groups in the vicinity of railway lines, in a gradient of distances from the track, it was found, that hepta-CBs had the highest percentage share in soil and plants from all the analysed sites and that there is no clear relationship between the distance from the track and the share of different congener groups in soil (both depths) and in plants.

Analyses of the percentage share of different PCB congener groups in soil and plant material near railway lines have clearly revealed a similar trend as in the vicinity of railway junctions (fig. 4). The analysed material contained mainly hepta-CBs, but the distribution of results concerning their content in soil and plants is similar. In addition, high percentage share of hexa-CBs was found in plant material (i.e. similarly to railway junctions), while the share of other congener groups in the analysed samples was between 10 and 20 per cent.

The percentage share of all congener groups in the standard mixture (a mixture of Aroclors) was compared to that in soil (at two depths) and plants from railway junction and railway line areas (the average content in the study material) (table I). It was found that hepta-CBs had the highest percentage share in field samples (28- 51%), while their content in the master mixture was not so high (10%). On the contrary, those congener groups which have a rather high share in the master mixture (tetra-, penta-, hexa-CBs), were not so common in the field samples (with the exception of plants from railway junction areas).

Statistical regression analysis of soil and plant contamination with PCBs and their different congener groups in the vicinity of railway junctions and railway lines.

Railway junctions

The application of *Kendall's* independence test for the data concerning PCB content for combinations: soil (0–20 cm) – soil (20–40 cm) and soil (20–40 cm) – plants does not give any basis for rejecting the hypothesis that their content in soil (at two depths) is independent to that in plants. Therefore, there is no basis for establishing regression line equations. However, for the combination soil (0–20 %) – plants, *Kendall's* test allows for rejecting the data independence hypothesis at the significance level of p = 0.05. Therefore, the value of the correlation coefficient was calculated (r = 0.7) and the regression line equation was found. Fig. 5 shows the joint distribution of PCB content levels in soil and plants.

Data concerning the proportion of different congener groups in the analysed material was tested in the same way.

The test provided basis for establishing regression line equations (p = 0.05) and the values of correlation coefficients in the following combinations: for both soil (0–20 cm) and plants – penta-CB – hepta-CB, for soil (20–40 cm) and plants – tri-CB – hepta-CB, as well as for soil at two depths in the case of hepta-CBs. However, in other soil and plant contamination combinations analysed no such basis was found. Figure 6 illustrates the joint distribution of different PCB congener content in soil and plants.

Railway lines

In the case soil (both depths) and plant pollution with PCBs, in the area of railway junctions and railway lines, application of *Kendall's* independence test does not give the basis for rejecting the hypothesis of their content independence in soil and plants. Therefore, there is no basis for establishing regression line equations for the following combinations: soil (0–20 cm) – plants and soil (20–40 cm) – plants.

Figure 6 illustrates the joint distribution of PCB concentration levels in the following combinations: soil (0–20 cm) – soil (20–40 cm), soil (0–20 cm) – plants and soil (20–40 cm) – plants. In the case of soil contamination levels at two depths, application of Kendall's independence tests allows for rejecting the hypothesis of PCB content independence in the study material at the significance level of p = 0.05. Therefore, a regression line equation was calculated, as well as the value of the correlation coefficient (r = 0.5).

Figure 7 illustrates the joint distribution of concentrations of different PCB congener groups in the analysed dependency combinations for both soil layers and plant contamination. Of all these cases, it is only the combinations: mono, di-CB and hexa-CB for soil contamination at two depths (r = 0.6) that give the basis for rejecting the hypothesis of soil and plant pollution independence. Hence, regression line equations (p = 0.05) and correlation coefficients were calculated.

On the basis of regression analysis for soil and plant contamination it was found that there is no correlation between concentration levels of these compounds in most cases of areas adjacent to railway lines. A significant correlation exists for most plant and soil (both depths) pollution with mono- and di-CBs (there also is a weak correlation for tri-CBs). In railway junction areas correlation were found for surface soil layer and plant pollution with penta- and hepta-chlorine congeners. High correlation was also found for hepta-CBs at two depths of soil.

Therefore, a conclusion can be drawn that penta- and hepta-chlorine derivatives persist on leaf and soil surface in a larger proportion than other groups of congeners. Besides, the low content of less-chlorinated PCBs in soil and plants suggests that some of these substances evaporate and move at larger distances.

Lipophilic properties of PCB compounds increase with chlorination of the biphenyle ring. Therefore, it should be expected that more-chlorinated congener groups would constitute the majority of PCBs analysed. This is confirmed by the obtained results. High content of hepta-CBs was found in railway junction areas, in plants which grew in places where the soil was also highly contaminated with these substances. Such relationship does not occur in the vicinity of railway lines. Despite the fact that the highest percentage share in plant and soil pollution was found for hepta-CBs, no significant relationship between their content in soil and plants was found. It seems that, while in railway junction areas oil derivatives remain permanently on the surface of soil as stain pollutants, in the vicinity of railway lines grease, oil and similar spills are dispersed during train movement. This may lead to uneven pollution of soil and plants from the same sites.

Statistical analysis of different PCB congener group accumulation in plants and their penetration into the soil near railway junctions and railway lines.

Additionally, an attempt was made to establish which groups of PCB congeners are accumulated at the highest rate in emerged parts of dandelion. In order to do so, the ratios of the content of each group in plants and their content in surface soil layers were calculated for each study site. Subsequently, sign test was performed for each pair of congener groups in order to test the hypothesis that the calculated content ratios had the same distribution. If this hypothesis can be rejected then, there are basis to claim that plants accumulate congeners from one group at a significantly higher rate than those from a different group.

Similar calculations were done in order to investigate which groups of congeners penetrate deeper into the soil. To this end, the ratios of the content of each group in surface soil layers to their content in deeper soil layers were calculated and the results were tested using the sign test.

The above calculations were made independently for study sites within railway junction areas and areas adjacent to railway lines.

No significant differences were found in the rate of penetration of different groups of PCB congeners into the soil, neither in railway junction areas, nor in the vicinity of railway lines.

Comparisons between pairs, performed for accumulation rates of different groups of congeners by plants, have revealed significant differences in accumulation rates for the following pairs:

1. tetra-CB and penta-CB near railway junctions
2. penta-CB and hexa-CB near railway junctions
3. tetra-CB and hexa-CB near railway lines

4. penta-CB and hexa-CB near railway lines.

Statistical analysis concerning various PCB congener groups and their accumulation in plants, as well as penetration into the soil, has revealed that there are significant differences in the rate of tetra-, penta-, and hepta-CB accumulation in plants. It was found that plants accumulate tetra-CBs and hexa-CBs at a higher rate than penta-CBs. Comparison of these pairs has demonstrated that the rates of these congeners' accumulation are as follows: hexa-CB > tetra-CB > penta-CB. In the area of railway lines these rates are different and their comparison is as follows: hexa-CB > penta-CB > tetra-CB. Among the three groups of congeners it is those with strongest lipophilic properties that are accumulated at the highest rate (hexa-CB). While in the vicinity of railway lines the rate of hexa-, penta- and tetra-CB accumulation is in accordance with their lipophilic properties, in the area of railway junctions tetra-CBs are accumulated stronger than penta-CBs. This is probably due to the fact that plants growing in railway junction areas are additionally contaminated with other oil derivatives, which leads to a different distribution of accumulation rates among congener groups. It is worth considering why it was not possible to establish accumulation rates for other congener groups, mainly hepta-CBs the proportion of which in field samples is the highest. However, it should be remembered that substances emitted into the environment do not contain much of them (about 10%), so the revealed 30–50% share of these congeners in field samples is probably the result of their build-up which may not be connected to accumulation processes in lipids only.

Comparisons of other congener pairs did not demonstrate significant differences in the rates of their accumulation in plants, neither in the area of railway junctions, nor near railway lines.

Unfortunately, it is impossible to unequivocally establish what percentage of PCB congeners emitted by railway transport remains in the area where this pollution takes place and how much PCBs move to adjacent areas. The obtained results suggest that dandelion is suitable for bioindication of environmental pollution with PCBs.

CONCLUSIONS

Waste motor and transformer oils are formerly considered as a main source of PCBs emission into the environment in Poland [9]. However, on the basis of our results it can be said that railway transport constitutes a serious source of soil contamination with polychlorinated biphenyls. Moreover, high concentrations of the analysed substances in *Taraxacum officinale* plants from railway areas (with varying degrees of soil contamination with these substances) prove that this species may be suitable for PCB pollution bioindication surveys. It was found that there is no correlation between the content of tri-, tetra-, penta- and hexa-CBs in soil (both depths) and their content in plant material. Strong correlation which exists between soil pollution (both depths) and plant contamination with mono- and di-CBs in areas adjacent to railway lines and junctions, as well as strong correlation between hepta- CB content in soil (both depths) and plants near railway junctions indicate that:

1. highly-chlorinated PCBs are adsorbed on the surface of polluted soil which has high contents of organic substances (oil derivatives) and, therefore, they do not penetrate into the soil at a high rate

2. highly-chlorinated PCBs migrate into unpolluted soil with low content of organic substances (areas adjacent to railway lines and junctions)

3. PCBs are accumulated in emerged parts of dandelion

4. the rate of PCB accumulation o the leaf surface depends on the chlorination of the biphenyl in, which is in accordance with the natural kinship of these substances to lipids (the value of the $K_{(octanol\text{-}water)}$ increases with the degree of biphenyl ring chlorination)

5. less-chlorinated PCBs do not persist on the surface of soil and leaves, probably due to their high rate of evaporation.

M. Malawska, B. Wiłkomirski

ACCUMULATION RATE OF POLYCHLORINATED BIPHENYLS (PCBS) IN DANDELION (*TARAXACUM OFFICINALE*) IN CONDITIONS OF SOIL CONTAMINATION WITH OIL DERIVATIVES

Summary

The study concerned PCB accumulation in soil at two depths (S1: 0–20 cm, S2: 20- 40 cm) and in *Taraxacum officinale* plants. It was carried out within railway junctions and near railway lines. Various degrees of PCB contamination were detected in soil and parts of plants above the ground level. PCB content in most soil samples was between 100 and 250 ng/g, while in plants it varied from 100 to 800 ng/g. An analysis of the distribution of different groups of PCB congeners in plant and soil material has revealed that the content of hepta-CB and hexa-CB fractions was the highest of all PCBs for both soil depths, as well as for plants. No significant differences were found for the degree of various PCB congener group penetration into the soil, neither in railway junction areas, nor in the vicinity of railway tracks.

Statistical test of pair comparison, performed in order to establish the rate at which plants accumulate different groups of PCB congeners has revealed significant differences in accumulation rates for the following pairs: penta-chlorinated congeners are accumulated at a higher rate than tetra-chlorinated ones and hexa-chlorinated congeners are accumulated at a higher rate than penta-chlorinated ones. This phenomenon occurs in the area of railway junctions (areas heavily polluted with oil derivatives). It was found that in the area of railway lines (areas with low levels of pollution) hexa-chlorinated congeners were accumulated at a higher rate than tetra- and penta-chlorinated ones. No significant differences were found for other pairs of PCB congeners. The use of dandelion as a bioindicator of environmental pollution with PCB congeners seems to be a good and reliable source of information about the emission of those substances into the natural environment.

M. Malawska, B. Wiłkomirski

STOPIEŃ KUMULACJI POLICHLOROWANYCH BIFENYLI (PCB) W MNISZKU LEKARSKIM (*TARAXACUM OFFICINALE*) W WARUNKACH ZANIECZYSZCZENIA GLEBY SUBSTANCJAMI ROPOPOCHODNYMI

Streszczenie

Przeprowadzone badania dotyczyły kumulacji PCB w glebie z dwóch głebokości (S1:0–20 cm, S2:20–40 cm) oraz w roślinach *Taraxacum officinale*. Prace prowadzono na węzłach kolejowych i w sąsiedztwie linii kolejowych. Stwierdzono różne stopnie zanieczyszczenia w glebie i roślinach, przekraczające poziom tła. Zawartość PCB w większości próbek glebowych była w granicach 100 do 250 ng/g, podczas gdy w roślinach zawierała się od 100 do 800 ng/g. Analiza rozmieszczenia

poszczególnych kongenerów w materiale roślinnym i glebowym potwierdziła, że zarówno w glebie jak i w roślinach najwyższy poziom osiągnęły frakcje hepta- i heksa-CB. Statystyczny test znaków wykonany w celu ustalenia stopnia, w jakim rośliny akumulują różne grupy kongenerów PCB wykazał znaczące różnice stopnia kumulacji dla następujących par: kongenery pentachlorowane kumulują się w większym stopniu niż tetrachlorowane, a heksa-chlorowane w większym stopniu niż pentachlorowane. Takie zjawisko występuje na terenie węzłów kolejowych (tereny silnie skażone substancjami ropopochodnymi). W sąsiedztwie linii kolejowych (tereny o niskim skażeniu) heksachlorowane kongenery kumulowały się w większym stopniu niż tetra- i pentachlorowane pochodne. Nie stwierdzono znaczących różnic dla innych par kongenerów.

Na podstawie uzyskanych wyników można uznać mniszka lekarskiego za dobry bioindykator skażenia polichlorowanymi bifenylami.

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