*ORIGINAL PAPER*

# TRANSFER OF POLYCHLORINATED BIPHENYLS (PCBS) FROM MEDICINAL PLANTS INTO INFUSIONS FOR THERAPEUTIC PURPOSES

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***Abstract.*** *This study aimed to investigate the content of polychlorinated biphenyls (PCBs) in various medicinal plants, commercially available in Romania, as well as in herbal infusions. Microwave extraction was used to separate PCB compounds from dried plants, and liquid-liquid extraction was used for infusions. The PCB content in the samples was determined using gas chromatography equipped with an electron capture detector (GC-ECD). The results show that, in medicinal plants, hexa- and heptachlorinated compounds (PCB138 and PCB180) predominate, with higher concentrations found in yarrow (Achillea millefolium). In infusions, PCB compounds ranged from 0.7 ng/L (dandelion) to 10.79 ng/L (yarrow), with concentrations varying by plant species and organ (leaf, flower, or fruit). The percentage transfer of polychlorinated biphenyls from plants to infusions was also calculated, and the results ranged from 2.70 to 27.01%. It can be concluded that the transfer percentage of polychlorinated biphenyls from medicinal plants, as analysed in infusions, is relatively small and depends on the initial toxic content and the plant organ used for infusion.*

***Keywords****: PCBs; medicinal plants; infusions; transfer rate.*

## 1. INTRODUCTION

At the current stage of human society's evolution on the planet, it faces the widespread pollution of the natural environment, largely due to anthropogenic activities. Among them, organic pollutants, whether polycyclic aromatic hydrocarbons (PAHs), organochlorine pesticides (OCPs), or polychlorinated biphenyls (PCBs), are the most dangerous for human health, exhibiting teratogenic, neurotoxic, and immunotoxic properties, or adversely affecting reproductive processes [1,2].

Most of these compounds are lipophilic, are deposited in the lipid-rich regions of living cells and can thus be stored for long periods in plant or animal tissues, from which they can be transferred to the human body through food. A major problem is the frequent use of medicinal plants for the treatment of minor ailments, as part of traditional therapy [3,4]. Organic pollutants that accumulate in medicinal plants can be transferred to therapeutic

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preparations, which interfere with the effects of the treatment. Recent studies have demonstrated the transfer of potentially carcinogenic polycyclic aromatic hydrocarbons, as well as organochlorine pesticides or their derivatives, resulting from biotransformation, from medicinal plants to preparations intended for human therapy [5-9].

Polychlorinated biphenyls are highly persistent organic pollutants in the natural environment [10]. Structurally, they belong to a class of organic compounds with 1-10 chlorine atoms attached to two coupled biphenyl nuclei at different positions, having the general chemical formula $C1_2H_{10-x}Cl_x$. These substances began to be produced by the synthesis industry starting in 1929 and were used as ingredients in paints, as plasticizers, insulators, heat-transfer fluids, and additives in hydraulic installations, etc. In the 1970s, their production was discontinued due to their persistence and toxicity. PCBs have been detected in almost all environmental samples analyzed (including aquatic systems, soils, plants, bird eggs, and mammalian milk). To date, two-thirds of the total amount of PCBs produced globally remains in the environment [11,12].

Polychlorinated biphenyls can also be generated as by-products in human activities involving combustion. Because of their high toxicity and practically unlimited lifespan, their levels in products intended for human and animal consumption must be carefully and continuously monitored. The largest quantities of this family of compounds are found in paints, plastics, and rubber. To monitor the presence and quantities of these compounds, modern extraction and quantification methods have been developed [13-15]. These methods have revealed the presence of PCBs in many food categories, such as dairy products and milk, vegetables, and non-alcoholic beverages. Contamination can occur both through the natural environment (water, air, soil) and through plastic packaging.

Studies on aromatic plants used in food have shown quantifiable levels of PCBs in rosemary and oregano, likely due to their partitioning into the lipophilic volatile oils of these plants [16]. Tea (*Camellia sinensis*) is a plant subject to PCB contamination (which, given its worldwide consumption, is a worrying concern) [16-18]. Some studies report the presence of PCBs in 16 species of medicinal plants harvested from relatively clean wild areas, as well as in infusions and decoctions prepared from these plants [19].

Based on the results reported in the literature, our work sheds light on the transfer of 7 PCBs (PCB 28, PCB 52, PCB 101, PCB 118, PCB 138, PCB 152, and PCB 180) from medicinal plants intended for human therapy, purchased from pharmacies, into infusions prepared for consumption. The results indicate that this group of organic pollutants must be carefully monitored in products intended for human consumption, in accordance with norms developed by international organizations.

## 2. MATERIALS AND METHODS

### *2.1. MATERIALS*

#### 2.1.1. Samples

Samples from different types of dried medicinal plants (MPs) were purchased from various pharmaceutical markets in Romania. These materials included leaves of wormwood (*Artemisia absinthium*) and dandelion (*Taraxacum officinale*), and flowers of yarrow (*Achillea millefolium*), St. John's wort (*Hypericum perforatum*), linden (*Tilia sp.*), chamomile (*Matricaria chamomilla*), as well as fruits of one-seed hawthorn (*Crataegus monogyna*) and rose hips (*Rosa canina*).

### 2.1.2. Sample preparation and clean-up

The sample extraction and analysis procedures were performed according to previously described analytical methods [20].

For the extraction of polychlorinated biphenyls (PCBs) from dried plant material, 2.5 g of homogenized sample was mixed with 30 mL of acetone/hexane (1:1, v/v). Microwave-assisted extraction was performed using a Milestone START E microwave solvent extraction system (MILESTONE Srl, Sorisole-Bergamo, Italy) operating at 1000 W, 120 °C, and 50 min, in closed extraction vessels according to the manufacturer's operating conditions. After extraction, the samples were cooled to room temperature and filtered through filter paper.

Infusions were prepared according to the manufacturer's instructions. Briefly, 5 g of dried plant material were immersed in 600 mL of boiling tap water and brewed under cover for 10 min. After cooling, PCBs were extracted using 25 mL of hexane/dichloromethane (3:1, v/v) in a separatory funnel.

The obtained extracts were purified using a Florisil column and concentrated to 1 mL using a Kuderna–Danish concentrator (Thermo Fisher Scientific Inc., Waltham, MA, USA) under a gentle nitrogen stream prior to GC-ECD analysis. The internal standard 2,4,5-trichlorobiphenyl was added to all samples to evaluate analytical recovery and minimize potential matrix effects [21].

### *2.2. METHODS*

Polychlorinated biphenyls were analyzed using a gas chromatograph, Perkin Elmer Clarus 500 (PerkinElmer U.S. LLC, Shelton, CT, USA) equipped with an electron capture detector (ECD). The working conditions for the gas-chromatographic analysis were: column: 30m×0.25mm×0.50 µm Elite-5MS; Carrier gas: helium, 1 mL/min; split flow: 25 mL/min; injector temperature: 330 °C; detector gas: nitrogen; detector temperature: 330 °C. The oven temperature program was: 180 °C (0 min); 7°C/min up to 230 °C (10 min); 15 °C (C/min) up to 250 °C (2 min). The quantification of specific PCBs was achieved by comparing the areas of the sample peaks to those of known standards. Calibration was performed using individual standards for 7 PCBs (PCB 28, PCB 52, PCB 101, PCB 118, PCB 138, PCB 152, and PCB 180), and quality control included blanks and spike recoveries to ensure analytical reliability. Table 1 presents the retention times and calibration parameters for each PCB analyte, including correlation coefficients ($R^2$) and corresponding LOD values. The correlation coefficients of the calibration curves ranged from 0.9952 to 0.9996, indicating good linearity, with all $R^2$ values exceeding 0.995. The detection limits ranged between 0.0003 and 0.0007 µg/g. The average recoveries of OPCs in herb samples ranged from 90% to 98%, indicating satisfactory reproducibility of the method. Each sample was analyzed in triplicate, and the results are expressed as the mean ± SD.

The transfer percentage of PCBs from plant to infusion was calculated using formula (1):

$$T(\%) = \frac{C_i \cdot V_i}{C_t \cdot m_t} \cdot 100 \quad (1)$$

where: *Ci*-concentration (ng/L) of PCBs in the infusion; *Vi*- volume (L) of the infusion; $C_t$ - concentration (ng/g) of PCBs in dry tea; $m_t$ - weight (g) of dry plant prepared for making infusion.

**Table 1. Retention time, monitored ion, linearity, and limits of detection (LOD) of PCBs in tea.**

| PCBs | Retention Time [min] | Linearity $R^2$ | LOD [µg/g] d.w. |
|---|---|---|---|
| 2,4,4′-trichlorobiphenyl (PCB 28) | 18.827 | 0.9996 | 0.0004 |
| 2,2′,5,5′-tetrachlorobiphenyl (PCB 52) | 20.801 | 0.9979 | 0.0003 |
| 2,2′,4,5,5′-pentachlorobiphenyl (PCB 101) | 25.880 | 0.9988 | 0.0006 |
| 2,3′,4,4′,5-pentachlorobiphenyl (PCB 118) | 29.591 | 0.9992 | 0.0004 |
| 2,2′,3,4,4′,5′-hexachlorobiphenyl (PCB 138) | 30.819 | 0.9994 | 0.0006 |
| 2,2′,4,4′,5,5′-hexachlorobiphenyl (PCB 152) | 32.362 | 0.9952 | 0.0007 |
| 2,2′,3,4,4′,5,5′-heptachlorobiphenyl (PCB 180) | 36.257 | 0.9992 | 0.0003 |

## 3. RESULTS AND DISCUSSION

### *3.1. RESULTS*

Regarding PCB concentrations, the European Union has established maximum limits for food products, but no safe limits have been set for vegetables or plant products, limiting the ability to draw general conclusions about safety. Table 2 shows the PCB content in the medicinal plants selected for the study.

**Table 2. Polychlorinated biphenyls in medicinal plants.**

| | Leaves | | | Flowers | | | Fruits | |
|---|---|---|---|---|---|---|---|---|
| **PCBs, [ng/g]** | ***Artemisia absinthium*** | ***Taraxacum officinale*** | ***Achillea millefolium*** | ***Hypericum perforatum*** | ***Tilia* sp** | ***Matricaria chamomilla*** | ***Crataegus monogyna*** | ***Rosa canina*** |
| PCB28 | 1.155±0.21 | 2.217±0.15 | <LOD | <LOD | <LOD | 7.263±1.21 | <LOD | <LOD |
| PCB52 | 2.104±0.16 | <LOD | <LOD | 3.3297±0.45 | BLD | <LOD | 2.599±0.14 | <LOD |
| PCB101 | <LOD | <LOD | <LOD | 2.4619±0.13 | 1.038±0.09 | <LOD | 3.886±0.42 | 3.262±0.42 |
| PCB118 | <LOD | 0.0244±0.01 | 8.1183±1.21 | <LOD | <LOD | <LOD | 2.256±0.18 | <LOD |
| PCB153 | <LOD | <LOD | 7.0240±0,54 | <LOD | <LOD | 1.932±0.14 | <LOD | <LOD |
| PCB138 | <LOD | 0.0075±0.01 | 6.8664±0.43 | 5.5084±0.34 | 3.002±0.13 | 4.163±0.22 | 5.057±0.61 | 4.029±0.27 |
| PCB180 | <LOD | 0.0064±0.01 | 4.7937±0.21 | 4.2001±0.17 | 2.107±0.11 | 5.633±0.46 | <LOD | <LOD |
| ∑PCB | 3.26±0.28 | 2.255±0.11 | 26.8024±2.75 | 15.500±1.09 | 6.147±0.58 | 18.992±2.1 | 13.799±1.24 | 7.291±0.89 |

*Note: LOD is the limit of detection.*

The concentrations of PCBs detected in the plants were below the European Union's maximum level of 40 µg/kg for food [22]. Therefore, the examined samples had values below the reference level.

Polychlorinated biphenyl compounds were also analyzed in infusions prepared from the studied plants to evaluate the extent of their transfer into tea; the results are presented in Table 3.

**Table 3. Polychlorinated biphenyls in tea infusion.**

| | Leaves | | | Flowers | | | Fruits | |
|---|---|---|---|---|---|---|---|---|
| **PCBs, [ng/L]** | ***Artemisia absinthium*** | ***Taraxacum officinale*** | ***Achillea millefolium*** | ***Hypericum perforatum*** | ***Tilia* sp** | ***Matricaria chamomilla*** | ***Crataegus monogyna*** | ***Rosa canina*** |
| PCB28 | 0.911±0.11 | 0.700±0.08 | <LOD | <LOD | <LOD | 10.304±1.92 | <LOD | <LOD |
| PCB52 | 2.627±0.23 | <LOD | <LOD | 5.661±1.04 | <LOD | <LOD | 4.466±1.03 | <LOD |
| PCB101 | <LOD | <LOD | <LOD | 4.558±0.28 | 1.618±0.14 | <LOD | 6.191±0.79 | 3.539±0.31 |
| PCB118 | <LOD | <LOD | 6.024±1.23 | <LOD | <LOD | <LOD | 3.441±0.24 | <LOD |
| PCB153 | <LOD | <LOD | 8.019±1.87 | <LOD | <LOD | 3.319±0.26 | <LOD | <LOD |
| PCB138 | <LOD | <LOD | 8.917±0.69 | 4.334±0.45 | 3.541±0.27 | 5.627±0.51 | 2.007±0.42 | 1.007±0.09 |
| PCB180 | <LOD | <LOD | 10.790±1.55 | 6.043±1.03 | 1.954±0.15 | 2.093±0.23 | <LOD | <LOD |

*LOD- limit of detection*

Herbal infusions could be a source of PCB contamination for consumers, and exposure may depend on the concentration of compounds in the raw material. Some studies have shown that pollutant transfer is higher during the first 5 minutes of infusion [23,24]. Table 4 shows the percentage of transfer of polychlorinated biphenyl compounds in infusions.

**Table 4. Percentage transfer (%) of PCBs in infusions**

| % trans | Leaves | | | Flowers | | | Fruits | |
|---|---|---|---|---|---|---|---|---|
| **PCBs** | ***Artemisia absinthium*** | ***Taraxacum officinale*** | ***Achillea millefolium*** | ***Hypericum perforatum*** | ***Tilia* sp** | ***Matricaria chamomilla*** | ***Crataegus monogyna*** | ***Rosa canina*** |
| PCB28 | 9.46 | 2.70 | - | - | - | 17.02 | - | - |
| PCB52 | 14.97 | - | - | 20.42 | - | - | 20.58 | - |
| PCB101 | - | - | - | 22.21 | 18.69 | - | 19.11 | 13.05 |
| PCB118 | - | - | 8.90 | - | - | - | 18.35 | - |
| PCB153 | - | - | 13.69 | - | - | 20.61 | - | - |
| PCB138 | - | - | 15.58 | 9.44 | 14.15 | 16.23 | 8.16 | 2.99 |
| PCB180 | - | - | 27.01 | 17.26 | 11.68 | 12.20 | - | - |

„-„ -no transfer to infusion was taken into account when the pesticide residues in tea leaves are <LOD

## *3.2. DISCUSSIONS*

In medicinal plants, the following order of PCB presence was observed: *Achillea millefolium* > *Hypericum perforatum.*> *Crataegus monogyna* > *Rosa canina* > *Tilia sp* > *Artemisia absinthium* > *Taraxacum officinale*; while the concentration of individual compounds shows the order: PCB138> PCB180> PCB101> PCB28> PCB118> PCB153> PCB52.

The results show that among the analysed medicinal plants, polychlorinated compounds predominate, especially hexa- and heptachlorinated species (PCB138 and PCB180), whereas less chlorinated compounds (PCB28 and PCB52) are less abundant. On the other hand, PCB contamination levels are not constant, and each compound may be present at different concentrations in the analyzed products. In addition, higher levels of PCB118 and PCB153 were found in *Achillea millefolium*, and PCB28 in *Matricaria chamomilla*. This distribution may indicate that PCBs are absorbed mainly from soil and accumulate less in air. Given that the origin of the plants is unknown, some being harvested from the wild and others cultivated, we cannot estimate the sources of contamination for the medicinal plants analyzed. Previous studies have reported that high-molecular-weight polychlorinated compounds are more widespread in soils and bioaccumulate in plants [18].

Concerning the concentration of PCBs in medicinal plants, the results obtained are difficult to compare with other studies, as there is little data on these contaminants in herbal teas. Nevertheless, Nakata et al. (2002) reported values below 0.01 ng/g for ∑PCBs in tea leaves purchased from Shanghai markets, while Polder et al. (2010) reported average values of 1.51 ng/g for 16 PCB compounds in black tea leaves [25,26]. At the same time, Amakura et al. (2009) showed the presence of very low concentrations of 9.86 pg/g of dioxin-like PCBs in herbal teas, and Mosleh et al. (2014) mentioned that all tea samples tested did not contain PCB residues [27,28].

The differences observed between the results of this study and the data available in the literature may be related to plant type and contaminant composition, plant processing methods, and environmental factors [16]. It can be said that the accumulation of PCBs in plants depends on the degree of chlorination of the compounds; tri- and tert-chlorinated compounds, being more volatile, were detected at low concentrations in most plants, except

*Matricaria chamomilla*. This plant is one of the species proposed for phytoremediation, known for its ability to accumulate lipophilic organic substances from contaminated soils [29].

In contrast, the presence of PCB isomers (i.e., PCB138 and PCB153) may indicate the persistence of polychlorinated compounds in the soil resulting from prior contamination. Another study reported that PCB isomers, by the nature of multiple bonds (C-C, C-H, and C-Cl), present a series of properties (high molecular weight, low vapour pressure, low solubility in water), which can lead to their stability and persistence in the environment, with the potential to induce toxicity in various organisms and plants [30]. On the other hand, a series of physicochemical characteristics of soils (organic matter, microbial activity), as well as the properties of PCB compounds (hydrophobicity), can influence their degradation, adsorption, and bioaccumulation in plants [31].

The hydrophobicity of PCBs is given by the octanol-water partition coefficient (log K ow), which increases with the degree of chlorination [32]. The vapour pressure decreases with increasing degree of chlorination, indicating that less-chlorinated PCBs are more volatile [33]. The number of chlorine atoms in the ortho-position can also influence the vapour pressure, which increases with increasing number of substituents in the ortho-position (the ortho-position) [34]. In air, PCBs can oxidize to toxic derivatives, known as hydroxylated polychlorinated biphenyls (HO-PCBs) and methoxylated polychlorinated biphenyls (MeO-PCBs), which are more toxic than their parent compounds [35]. PCBs in the environment can also be oxidatively degraded by aerobic bacteria and other microorganisms, or reductively dehalogenated by anaerobic microorganisms. The biodegradability of PCBs depends largely on the substitution of chlorine, i.e., on the number and position of chlorine atoms [36].

The study of infusions prepared from plant raw materials (known to contain PCBs) showed the presence of PCBs in the infusions (Table 3), except for two preparations (*Taraxacum officinale* leaves and *Tilia sp.* flowers), where their content was below the detection level. In most infusion samples, PCB concentrations ranged from 0.7 ng/L (*Taraxacum officinale*) to 10.79 ng/L (*Achillea millefolium*). As for individual compounds, PCB28 and PCB180 were found to be present in high concentrations in *Matricaria chamomilla* and *Achillea millefolium*, while PCB28 showed low values in *Artemisia absinthium* and *Taraxacum officinale* leaves. It can be estimated that, depending on the plant species and plant organ (leaf, flower, fruit), individual PCB compounds show variations in terms of their share in the infusions.

The results of this study showed that the transfer rate of PCBs in infusions varies with the plant organ infused (leaves, flowers, fruits). Thus, the average transfer percentage of contaminants in infusions ranged from 2.70-27.01% (leaves), 9.44-22.21% (flowers), and 2.99-20.58% (fruits), indicating a relatively higher degree of transfer in infusions prepared from flowers. At the same time, it was observed that the PCB transfer percentage also varied depending on the initial pollutant content in the raw material, with a high transfer rate observed for both lower and higher initial concentrations (e.g., for samples of plant raw materials with a PCB content within the limit of 3 ng/g, the transfer percentage varied between 2.70 and 20.58%). However, the transfer percentage of polychlorinated biphenyls from medicinal plants in infusions is relatively low and depends on the initial toxic content and the plant organ used for infusion.

Some studies have reported that the infusion preparation method plays an important role in pollutant transfer rates. This is influenced by several factors, such as the temperature and time of infusion and the herb-to-water ratio (TWR) [23,37]. At the same time, it has been shown that in the traditionally recommended method for preparing tea (infusion), the transfer rate of organic pollutants depends on certain properties, such as water solubility (Ws) and partition coefficient (log Kow) [19,38]. PCBs are compounds with relatively low water

solubilities (<10 mg/L) and high partition coefficients, which leads to their low transfer rates in infusions.

## 4. CONCLUSIONS

This study investigated the presence of PCBs in medicinal plants and their infusions using GC-ECD. The results of the study showed that PCB concentrations in the analyzed samples varied depending on the type of plant and the composition and properties of the contaminant. All plants selected for analysis, purchased from the pharmaceutical market, had PCB concentrations below the European Union's maximum level of 40 μg/kg for food. As regards the transfer percentage of polychlorinated biphenyls into infusions, a relatively low value was observed, depending on the initial toxic content and the plant organ infused. The data obtained are useful for establishing acceptable levels of PCB contamination in both the initial plant materials and in the ready-to-use phytopreparations.

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