



RESEARCH PAPER

Determination of Organochlorine Pesticides' Content in Dried Medicinal Plants and Their Infusions—Risk to Human Health

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Abstract

The study investigates the organochlorinated pesticides (OPCs) content in eight medicinal plant species (*Achillea millefolium*, *Artemisia absinthum*, *Hypericum perforatum*, *Tilia sp.*, *Matricaria chamomilla*, *Taraxacum officinale*, *Calendula officinalis*, *Crataegus monogyna*), purchased from pharmacies, as well as in infusions prepared from these plants. A total number of 24 samples were analyzed by gas chromatography—electron capture detector (GC-ECD). The transfer rate of OPCs from dried herbs to infusions, as well as the health risk of OPCs residues in infusions (estimated daily intake (EDI) and Hazard Index (HI) were calculated. All medicinal plants (MPs) contained pesticides in different concentrations. For the compounds Heptachlor, p,p' DDT, p,p' DDD, Aldrin, Dieldrin, concentrations well above the maximum residue limits (MRLs) were obtained. The transfer rate of organochlorine pesticides from dried plant to infusion ranged from 3.95% for Heptachlor (*Artemisia absinthum*) to 57.71% for Dieldrin (*Achillea millefolium*). The correlation between the OCPs content in the MPs and their infusion are statistically analyzed using the t-test for independence in high dimension, based on the correlation distance. Significant result was found for all OCP detected in the two media, with the content in the MPs significantly correlated with the content in the infusion. The EDI of the detected pesticides was lower than the Acceptable Daily Intake (ADI), and the Hazard Index values were less than 1 ($HI < 1$) in all infusions. Also, the cumulative hazard index (CHI), showed values less than 1 ($CHI < 1$) for all types of tea. These results indicate that residues of OCPs in the analysed infusions pose minimal risk to human health. However, taking into account the large transfer variations obtained, the results of the study recommend the development of control protocols for herbal infusions intended for human therapy that include the measurement of OCPs concentrations.

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Highlights

- Organochlorine pesticides (OPCs) in eight medicinal plants and their infusions were detected, and the transfer rate were calculated.
- The correlation between the OCPs content in the medicinal plants and their infusion are statistically analyzed.
- Residual pesticides in infusions would not pose dietary risk to consumers.

Keywords Organochlorine pesticides contamination · Medicinal plants · Risk analysis

Introduction

Organochlorine pesticides are synthetic molecules used in agriculture to prevent and kill plant pests (insects, mites, nematodes, fungi, bacteria). These substances are also used for public health practices, such as the control of mosquitoes and other vector organisms of certain bacteria, viruses or parasites. Even some products used as household disinfectants contain organochlorine pesticides.

Contemporary agricultural practices require the use of organochlorine pesticides for extensive cultures. The negative effects of these practices are that: pests have adapted to these substances and increasing amounts of pesticides are needed for effective treatments, and on the other hand, these substances also contaminate surrounding soils and river water or surface water tables. The extensive use of these substances has led to persistent pollution of soil and water, with a major negative impact on human health, taking into account the fact that organochlorine pesticides pass in unmodified form or as residues in products intended for human consumption (tea, spices, medicinal plants) (Ozbey and Uygun 2007a, b; Jiang et al. 2020; Siraj et al. 2021; de Andrade et al. 2023).

Since pesticides pose health risks to consumers and are considered to have potentially harmful effects on the environment, international agencies such as the European Union (EU), World Health Organization (WHO) have proposed maximum residue limits (MRLs) in food and feed of plant and animal origin (EU 2005; WHO 2007).

Although all these aspects are known and attempts are being made to regulate them, the studies carried out on those who use them show that the risks to which they expose themselves and, at the same time, the consumers are being ignored. Studies show that education programs are needed to raise awareness among agricultural producers about the indiscriminate use of pesticides, which can lead to contamination of cultivated products and serious health problems (Sharafi et al. 2018).

The present study is part of the approach to raise awareness of the urgent need to change the quantities and types of organochlorine pesticides used in agriculture. Starting from the finding that OCPs exist in MPs on the pharmaceutical market (Bratu et al. 2023), the work continues the

investigation, determining the transfer of OCPs from dried MPs into infusions prepared from them, which are intended for medical therapies. Eight medicinal species, well known and used in Europe for the treatment of mild ailments, were studied.

The plants included in the study were selected both on the basis of their popularity among consumers and on the basis of the different areas from which they can be harvested. Some of them are exclusively cultivated (*Calendula officinalis*), some can be harvested from wild areas (*Hypericum perforatum*).

There are relatively few studies on the transfer of organochlorine pesticides and their residues from dried medicinal herbs to infusions for therapy (Ozbey and Uygun 2007a, b; Ozbey and Uygun 2007a, b; Lino and Silveira 1997; Fang et al. 2022).

A comparative study conducted by Florea et al. (2020), highlights the differential transfer of some pesticides from dried MPs in several types of preparations for therapeutic use (infusion, decoction, cold macerate). The conclusions of the study indicate infusions as the preparation in which the highest transfer occurs.

However, studies on the amount and way of transfer of pesticides from MPs to infusions (which are the most common therapeutic preparations) have not fully clarified these issues, especially with regard to the plant species-transfer relationship of OPC's in infusions.

There are several studies related to the transfer of some pesticides used in tea (*Camelia sinensis*) crops from the dried plant to infusion (Heshmati et al. 2021; Lu et al. 2020).

Likewise, Wang et al. (2019), studied the transfer mode of 42 pesticide molecules from dried tea leaves into infusion in correlation with their physicochemical properties.

The aim of the study is to highlight and quantify the transfer of organochlorinated pesticides from dried MPs marketed in pharmacies into infusions prepared for therapy and to estimate the risk they pose to health. Also, the correlation between the OCPs content in the medicinal plants and their infusion are statistically analyzed using a test based on the correlation distance. The results of the study could help develop control protocols for herbal

infusions intended for human therapy that include measurement of OCP concentrations.

The study provides new data on the amount of OCPs that are transferred by infusion from therapeutic plant products, as well as on the species-differentiated transfer of organochlorine pesticide molecules. Statistical processing of the obtained results provides data on the species-dependent differential transfer of OCPs in infusions prepared for therapeutic use. The study also provides important information on the health risk that herbal infusions may pose under these conditions.

Materials and Methods

Collection and Preparation of Tea Samples for Analysis

Samples from eight different types of dried MPs were selected from the pharmaceutical market from Romania: *Hypericum perforatum* (St. John's Wort); *Artemisia absinthium* (Wormwood, Absinthe); *Tilia* sp. (Linden); *Matricaria chamomilla* (Chamomile); *Achillea millefolium* (Yarrow); *Taraxacum officinale* (Dandelion); *Calendula officinalis* (Pot Marigold); *Crataegus monogyna* (Oneseed hawthorn). For each type of plant, three samples were purchased. All samples were grounded to homogenise before extraction and clean-up. OCPs were extracted from dried plant by microwave method with acetone/hexane (1:1) (v:v), from infusion by liquid–liquid using hexane/ dichloromethane,(3:1) (v:v) mixture.

2.5 g of dried material were added into a receptacle with 30 mL solvent. The samples were submitted to microwave extraction at $E(w) = 1000$ and 120°C for 50 min, in a MILESTONE START E (Italy) equipment and filtered through filter paper.

The infusions were prepared according to the indications mentioned by the manufacturer. For this purpose, 5 g of dried plant were submerged in 0.600 L of boiling-hot tap water. The infusion was brewed under cover for 10 min, and OCPs were extracted with 25 mL hexane/ dichloromethane, (3:1) (v:v) mixture. The obtained extracts were purified on a florisil column to remove any interference with other compounds that are not of interest and concentrated up to 25 mL on water bath using a Kuderna-Danish concentrator and under gentle nitrogen flow and analysed by GC-ECD technique. The internal standards 2,4,5-Trichlorobiphenyl for OCPs were added to the samples for quantifying the overall recovery on the analytical procedures and to remove potential matrix effects (IAEA-MEL 1995).

GC-ECD Analysis of OCPs

Organochlorine pesticide, were analysed using a gas chromatograph Perkin Elmer Clarus 500 equipped with an electron capture detector (ECD). The working conditions for the gas-chromatographic analysis were: Column: $30\text{ m} \times 0.25\text{ mm} \times 0.50\text{ }\mu\text{m}$ Elite-5MS; Carrier gas: helium, 1 mL/min; Split/Spitless Injector (S/S), injection made in split mode; split flow 25 mL/min; injector temperature: 330°C ; detector gas: nitrogen; detector temperature, 330°C . Oven temperature program: 180°C (0 min); 7°C/min up to 230°C (10 min); 15°C/min up to 250°C (2 min). The quantification of the specific OCPs was achieved by comparing the sample peaks areas to those of known standards. Individual standards for 9 OCPs (Hexachlorobenzene, Lindane, Heptachlor, Aldrin, Dieldrin, Endrin, p,p'DDE, p,p'DDD, and p,p'DDT), were used for the calibration curves for GC-ECD. The values of the retention times, linearity, and limits of detection (LOD) of OCPs are presented in Tables 1.

The correlation coefficients derived from the linear regressions were higher than 0.9800, which show a strong correlation between the peak height ratios and the concentrations. The detection limits ranged between $0.0002\text{--}0.0005\text{ mg}\cdot\text{Kg}^{-1}$. The average recoveries of OCPs in herb samples were from 90 to 98%, which indicates that the reproducibility of the method was satisfactory. Blank analyses were also performed in order to assess the interference from the sample.

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

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

where: C_i -concentration (ng/L) of OCPs in the infusion; V_i -volume (L) of the infusion; C_t -concentration (ng/g) of OCPs in dry tea; m_t - weight (g) of dry plant prepared for making infusion.

Risk Analysis

In order to fully understand the potential risks of these OCPs to human health, the levels of exposure to OCPs found in this study compared with the acceptable daily intake as estimated by the Food and Agriculture Organization of the United Nations/World Health Organizations (FAO/WHO 2017). For each type of organochlorine pesticide exposure, the estimated daily intake (EDI, $\mu\text{g/kg}$ body weight/day) was calculated using formula (2):

$$\text{EDI} = \frac{C \times \text{DR}}{\text{BW}} \quad (2)$$

where EDI is the estimated daily intake, C—the concentration of pesticide residues in the infusion ($\mu\text{g/L}$), DR—the recommended dietary allowance (200 mL/zi) and BW—the average body weight (70 kg).

The hazard index (HI) for each detected OCP was calculated as the ratio of the estimated daily intake (EDI, $\mu\text{g/kg}$ body weight/day) to the acceptable daily intake (ADI, $\mu\text{g/kg}$ body weight/day), which are considered to be safe levels of exposure. If the HI is > 1 , it may indicate an intolerable health threat, so the sources, pathways and routes of pesticide introduction need to be further evaluated (Eissa et al. 2014). The HI was calculated with relation (3):

$$HI = \frac{EDI}{ADI} \quad (3)$$

The cumulative hazard index (CHI) was evaluated using the Reeffstrup et al., (2010) Eq. (4):

$$CHI = \frac{EDI_1}{ADI_1} + \frac{EDI_2}{ADI_2} + \dots + \frac{EDI_n}{ADI_n} = \sum_i^n \frac{EDI_i}{ADI_i} \quad (4)$$

where EDI_1 , EDI_2 , EDI_n and EDI_i are the estimated average daily intake of each individual pesticide. ADI_1 , ADI_2 , ADI_n and ADI_i are the acceptable daily intake for each pesticide.

All experiments were carried out triplicate and the data were presented as mean $\pm$ standard deviation.

Statistical Analysis

The correlation between the OCPs content in the medicinal plants and their infusion are statistically analyzed. For each OCP detected, a nonparametric test was used to detect the independence between the OCP content in the plant and the corresponding tea infusion. The motivation behind is that, if the OCP content is transferred on from the plant to infusion, the two samples would be highly correlated. The distance correlation, introduced in Székely and Rizzo (2013), measures the dependence between random vectors in arbitrary dimensions. It takes values between 0 and 1 and it is equal

to 0 if and only if the random vectors are independent. This represents an advantage over the traditional Pearson correlation coefficient, for which an estimated value of 0 does not necessarily indicate the independence of the samples. However, the sample distance correlation, i.e., the empirical counterpart used in the computations, approaches 1 as the dimension increases, even though the random vectors are independent (see Table 1 in Székely and Rizzo 2013). To correct for this issue, Székely and Rizzo (2013) introduced the modified distance correlation, constructed using an unbiased estimator of the distance covariance. In contrast with the distance correlation, the modified distance correlation can lead to negative squared distance covariance values, a situation which prevents the computation of distance correlation. Moreover, the modified distance correlation is less efficient if the higher moments of the distributions do not exist. However, besides its asymptotic unbiasedness property, the modified distance correlation does not require normality of the random vectors, making it applicable even for smaller samples.

For a random sample $(X_i, Y_i) \in R^{2m}$, with $X_i = (X_{i,1}, \dots, X_{i,m})$ and $Y_i = (Y_{i,1}, \dots, Y_{i,m})$, $i = 1, \dots, n$, the modified distance correlation statistic was calculated with formula (5):

$$\mathcal{R}_n^* = \frac{Cov_n(X, Y)}{\sqrt{Cov_n(X, X)Cov_n(Y, Y)}}, \quad (5)$$

where $Cov_n(X, Y)$ is the modified distance covariance statistic.

The t-test for independence in high dimension investigates if X and Y are independent. More precisely, for each OCP, the test considers the following null hypothesis (H_0) and the alternative hypothesis (H_1):

H_0 : X and Y are independent versus H_1 : X and Y are not independent,

where X is the random vector of pesticides detected in the medicinal plant and Y is the random vector of pesticides

Table 1 Retention time, linearity, and limits of detection (LOD) of OCPs

OCPs	Retention Time	Linearity (R^2)	LOD ($\mu\text{g/g}$) dw*
Hexachlorobenzene (HCB)	3.176	0.9925	0.0005
Lindane (γ -HCH)	5.118	0.9909	0.0004
Heptachlor	7.060	0.9909	0.0003
Aldrin	7.886	0.9587	0.0003
Dieldrin	11.220	0.9860	0.0003
Endrin	12.200	0.9945	0.0004
p,p' dichlorodiphenyldichloroethylene (p,p'DDE)	10.800	0.9751	0.0002
p,p' dichlorodiphenyldichloroethane (p,p'DDD)	12.790	0.9917	0.0002
p,p'dichloro-diphenyl-trichloroethane (p,p'DDT)	15.100	0.9909	0.0002

LOD-detection of limit; R^2 -correlation coefficient; *dw: dry weight

detected in the infusion. Under the null hypothesis, the test statistic (6),

$$T_{\lambda} = \sqrt{\nu - 1} \frac{R_{\lambda}^*}{\sqrt{1 - (R_{\lambda}^*)^2}}, \quad (6)$$

is Student t distributed with $\nu - 1$ degrees of freedom, where $\nu = \frac{n(n-3)}{2}$. The test, implemented as “dcor.test” in the R package “energy” (R version 4.3.2 2023), computes the distance correlation between two samples X and Y with the Euclidean distance with index between 0 and 2, and then compares the value with the distance correlation obtained by the permutation of the two samples. Under independence, the p-value of the test is a multiple of $1/(k+1)$, where k is the number of permutations.

Results and Discussions

OPCs in Dried Medicinal Plants and Infusions

Table 2 shows the content of organochlorine pesticides in the medicinal plants (MPs) selected for the study. The obtained values show that the highest mean concentration of $6.27 \text{ mg} \cdot \text{kg}^{-1}$ was obtained for p,p'-DDD (*Taraxacum officinale*), and the lowest value $0.046 \text{ mg} \cdot \text{kg}^{-1}$ for p,p'DDE (*Crataegus monogyna*).

Also, for Heptachlor, p,p' DDT, p,p' DDD, Aldrin, Dieldrin, concentrations well above the maximum residue limits (MRLs) were obtained (WHO 2007). In the case of MPs, the following order of OPCs compounds presence is observed: *Taraxacum officinale* > *Achillea millefolium* > *Tilia sp.* > *Matricaria chamomilla* > *Hypericum perforatum* > *Calendula officinalis* > *Artemisia absinthium* > *Crataegus monogyna*. The concentrations in decreasing order of the individual pesticides were: p,p,DDD > p,p,DDT > Dieldrin > Endrin > Aldrin > Heptachlor > γ -HCH > p,p,DDE.

Some of these plants grow in natural habitats and contamination may be due to plant-specific bioaccumulation processes (Cajka et al. 2012).

It can be seen that some compounds (pp'-DDD, p,p,DDT, Dieldrin and Endrin) were detected in high concentrations in some plants (*Taraxacum officinale*, *Tilia sp.*, *Matricaria chamomilla*, *Achillea millefolium*), while in others they were present in very low concentrations or not detected at all (*Hypericum perforatum*, *Artemisia absinthium*, *Crataegus monogyna*). The later ones may originate from areas with lower levels of contamination, or their concentrations may decrease as a result of natural factors (such as, precipitation, volatilisation, photolysis, biodegradation), (Kaushik et al. 2009). It is also worth mentioning

that the three plants are usually harvested from wild areas, further away from the land where intensive agriculture is practiced, and thus more protected from organochlorine pesticide contamination.

In our study, the compound hexachlorobenzene (HCB) was not detected in any of the analysed MPs. It is considered a very persistent compound due to its chemical stability and resistance to biodegradation. In soil, HCB binds strongly to organic matter, accumulating in plants and crops (Barber et al. 2005). Although, its commercial production has been banned in most parts of the world, however currently, HCB contamination can mainly originate from chemical production processes (intermediate or additive compound) and incineration processes (Liu et al. 2013).

Among the HCHs compounds, the γ -HCH isomer (Lindane) was detected in 37.5% of the plants, being below the limit of OPCs in Herbal Medicines (WHO 2007), except for chamomile (Table 2). This plant is one of the species proposed for phytoremediation of petroleum contaminates oils, being known for its ability to accumulate lipophilic organic substances from contaminated soils (Raygan et al. 2014).

The results obtained by us are lower than those reported by Witczak et al. (2018) in herbal teas from Poland and those reported by Murtaç et al. (2018), in MPs collected from Albania.

In a previous paper we presented the results of the determination of the pesticide content in 14 MPs on the pharmaceutical market, in correlation with five toxic elements (Bratu et al. 2023). The results of the study led to the conclusion that there are statistically significant correlations between the levels of certain pesticides and toxic elements. The levels of OPC's determined were comparable with those in the present study.

From the group of cyclodrynes, the compounds aldrin, dieldrin and endrin were present in 75% of the samples, in high concentrations in the plants *Taraxacum officinale* ($0.97 \text{ mg} \cdot \text{kg}^{-1}$), *Matricaria chamomilla* ($2.32 \text{ mg} \cdot \text{kg}^{-1}$), and *Achillea millefolium* ($2.31 \text{ mg} \cdot \text{kg}^{-1}$), being above the MRLs for the pesticides (WHO 2007). This group of compounds (Drins) are chemicals that have been applied in agriculture to soil insects and in public health to control pests. (Chandra et al. 2021).

Aldrin exhibits low phytotoxicity, plants are only affected at high concentration levels; it is readily metabolised to Dieldrin, its presence being closely related to that of Dieldrin (Siraj et al. 2021). In our study, the presence of both compounds in MPs, indicates the presence of more recent source of contamination, and a less longer contact of plant with Aldrin, leading to the degradation of Aldrin to Dieldrin.

Heptachlor, was also a compound detected in the analysed MPs, in concentrations ranging from $0.15 \text{ mg} \cdot \text{kg}^{-1}$ (*Achillea millefolium*) to $0.88 \text{ mg} \cdot \text{kg}^{-1}$ (*Artemisia absinthium*), being above the MRL values. In nature, Heptachlor is metabolised

by soil bacteria and fungi to its more persistent derivative, Heptachlorpoxide (Sosan et al. 2020).

The values obtained by us were comparable to those obtained by Witczak et al., (2018), in herbal teas, but higher than those reported by Siraj et al. (2021), where the metabolite Heptachlorpoxide predominates.

DDTs and its metabolites were detected in about 50% of the herb samples, in concentrations ranging from 1.163 mg·kg⁻¹ to 12.633 mg·kg⁻¹. The lowest concentration of 0.046 mg·kg⁻¹ was recorded for p,p'-DDE (*Crataegus monogyna*), while the highest value of 6.27 mg·kg⁻¹ was obtained for p,p'-DDD (*Taraxacum officinale*). The higher values of p,p'-DDD may be a cause of metabolic conversion and isomerisation of p,p'-DDT compound by solar radiation to p,p'-DDD (Barriada-Pereira et al. 2005).

Some studies indicate that DDT, a commonly used insecticide, generates p,p' DDE and p,p'-DDD as primary products by dechlorination reaction (Qiu and Zhu 2010). The ratio of p,p'-DDT to (p,p'-DDE + p,p'-DDD) is often used to identify the source of DDT. A low ratio of p,p'-DDT/(p,p'-DDE + p,p'-DDD) suggests DDT contamination from a more distant time period, while a value > 1.0 indicates its recent application (Unyimadu et al. 2019).

In this study, the ratio p,p'-DDT/(p,p'-DDE + p,p'-DDD) was zero in 5 MPs, as no p,p'-DDT, or its metabolites (DDD + DDE) were detected. In the plants *Achillea millefolium*, *Taraxacum officinale* and *Calendula officinalis*, the ratio exceeded 1, indicating more recent exposure to DDT. In addition, the DDD/DDE ratio greater than one observed in *Calendula officinalis*, suggests that DDT underwent aerobic biodegradation (Doong et al. 2002).

Our results showed that OPCs in the analysed MPs show higher values than the data reported by Siraj et al. (2021); Amirahmadiet al. (2013) and lower than those shown by Witczak et al. (2018).

It should be noted that of all the studied plants, *Calendula officinalis* are the only cultivated species and therefore have the potential to take up DDT from agricultural soils.

A number of environmental factors may influence the levels and distribution of these pollutants. It has been reported the application of these pesticides in the wet season leads to an easier plant uptake (Mumtaz et al. 2015). Also, in the dry season, due to the action of microorganisms, transformations take place at the soil level that cause photodecomposition and isomerisation of some compounds (e.g. HCH, parent compound) (Agbeve et al. 2013).

In addition, different physicochemical characteristics of soils (organic matter, activity of microorganisms) as well as pesticide properties (hydrophobicity) may influence degradation, adsorption and bioaccumulation of pesticides in plants (Sweetman et al. 2005). It has been mentioned that DDT is more hydrophobic than HCH. Hydrophobic compounds are strongly bound to soil organic colloidal particles, resulting in lower uptake and transfer to the plant (Calvelo Pereira et al. 2006).

A study published by Pirsahab et al. (2017), highlights the influence of sunlight on the concentration of pesticides in apples, concluding that sunlight significantly decreases the concentrations of diazinol and chlorpyrifos.

Besides environmental factors and methods of extraction and analysis of pesticides can influence their concentration in consumed products. (Sharafi et al. 2015; Beneta et al. 2018; Takim and Aydemir 2023).

The persistent nature of organochlorine pesticides in the environment from previous applications in agriculture and public health may be another reason for their presence in plants (Jayaraj et al. 2016). The concentration of these compounds in plant tissue due to the bioaccumulation is higher than their presence in environmental media such as air and water (Cajka et al. 2012).

Table 2 Organochlorine pesticides (OPC) concentrations in medicinal plants (mg·Kg⁻¹)

mg·Kg ⁻¹	<i>Hypericum perforatum</i>	<i>Artemisia absinthium</i>	<i>Tilia sp.</i>	<i>Matricaria chamomilla</i>	<i>Achillea millefolium</i>	<i>Taraxacum officinale</i>	<i>Calendula officinalis</i>	<i>Crataegus monogyna</i>	MRL* (mg·Kg ⁻¹)
HCB	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	0.1
Lindane	0.10 ± 0.01	BLD	BLD	1.61 ± 0.70	BLD	BLD	0.16 ± 0.01	BLD	0.6
Heptachlor	0.27 ± 0.01	0.88 ± 0.11	0.34 ± 0.02	0.79 ± 0.10	0.15 ± 0.01	0.41 ± 0.03	BLD	BLD	0.05
Aldrin	0.69 ± 0.03	0.43 ± 0.01	0.87 ± 0.03	0.34 ± 0.01	0.25 ± 0.01	0.97 ± 0.12	BLD	BLD	0.05
Dieldrin	0.93 ± 0.11	BLD	1.25 ± 0.12	2.32 ± 0.53	0.21 ± 0.01	0.24 ± 0.01	BLD	0.234 ± 0.01	0.05
Endrin	BLD	0.19 ± 0.01	0.157 ± 0.01	1.04 ± 0.21	2.318 ± 0.56	BLD	0.053 ± 0.00	0.200 ± 0.01	0.05
p,p'DDE	0.81 ± 0.03	BLD	BLD	BLD	BLD	BLD	0.305 ± 0.01	0.046 ± 0.00	0.2
p,p'DDD	BLD	BLD	BLD	0.97 ± 0.13	4.37 ± 0.50	6.27 ± 1.20	1.023 ± 0.41	BLD	0.2
p,p'DDT	BLD	BLD	4.77 ± 0.21	BLD	0.44 ± 0.01	1.06 ± 0.18	0.759 ± 0.04	BLD	0.2
ΣOCP	2.8 ± 0.40	1.50 ± 0.20	7.387 ± 1.45	7.07 ± 1.20	7.738 ± 2.01	8.97 ± 1.89	2.30 ± 0.50	0.48 ± 0.02	

BLD below detection limit

*MRL- Maximum residue levels (mg·Kg⁻¹) of organochlorine pesticide in Herbal Medicines (WHO 2007)

Organochlorine pesticides were also determined from the infusions prepared from the MPs studied in order to estimate the percent transfer rate of these compounds in herbal tea (Table 3). Therefore, the transfer rate is a critical measure used to determine compliance with MRL standards and to identify potential human health risks of medicinal tea consumption. This transfer rate indicates the rate at which pesticides move from dried plants into the aqueous solution during the infusion process for herbal tea preparation (Lu et al. 2020).

In plant infusions, OPCs ranged from $1.31 \mu\text{g}\cdot\text{L}^{-1}$ (*Achillea millefolium*) and $13.98 \mu\text{g}\cdot\text{L}^{-1}$ (*Taraxacum officinale*). In terms of individual compounds, p,p'DDD were found to be present in high concentrations in *Taraxacum officinale* and *Achillea millefolium*, and Endrin in low concentrations in *Tilia sp.*, *Artemisia absinthium*, and *Calendula officinalis*.

It can be concluded that, depending on the plant species, the individual OPC compounds, show variations in their share in infusions. Thus, there is a high abundance of DDTs and cyclodrynes, both in infusions from cultivated plants (*Calendula officinalis*) and those grown in natural areas (*Artemisia absinthium*, *Achillea millefolium*, *Crataegus monogyna*).

Figures 1 and 2 illustrate the boxplot of the content of pesticides in the plant and in the infusion, respectively. In the plant, Lindane, Endrin, ppDDE and ppDDT show some outliers, while in the infusion medium, only ppDDD indicates some outliers. Moreover, if the content of pesticides is not detected in a certain MPs, is it also not detected in infusion formed using that plant. For all OPCs except HCB, where no content was detected, the test for independence in high dimension was applied at significance level 5%. The test does not assume any probability distribution of the data and the asymptotic distribution under the null hypothesis holds for samples larger than 10. The alternative hypothesis states the pesticide contents in the plant and the infusion are correlated. All the reported p-values were below the significance level, adjusted for the number of permutations, indicating that the amount of OPC in the plant is significantly correlated to the amount of OPC in the infusion, for all OPC detected. Table 4 shows the computed distance correlation with Euclidean distance with index 1 and 500 permutations. All computed distance correlations are above 0.5, indicating a strong dependence between the pesticide contents in the plant and in the infusion. Table 2 illustrates the difference between the modified distance correlation and the Pearson correlation coefficient. For the majority of the OPC, the latter tends to underestimate the values of the test statistic. Moreover, as mentioned in the Statistical analysis section, the Pearson correlation can only capture the linear dependence between the samples, while the modified distance correlation estimates non-linear dependence as well.

Transfer Rate (%) of OPCs

Herbal infusions could be a possible source of OPCs contamination for humans and the exposure depends on the concentration of residues in the MPs (Kottiappan et al. 2013).

Some studies showed that the transfer rate of pesticide residues is higher in the first 5 min of the infusion process (Cho et al. 2014). Generally, this infusion time is recommended on the package by manufacturers.

Table 5 shows the transfer rate of organochlorine pesticides in herbal infusions. The results show that the percentages ranged from 3.95% for Heptachlor (*Artemisia absinthium*) at 57.71% for dieldrin (*Achillea millefolium*), these values are much lower than those presented by Witczak et al. (2018), in herbal teas. Compounds p,p'DDDDD and p,p'DDT showed lower transfer rate values for most MPs.

Some studies have mentioned that the method of tea processing plays an important role in the transfer rate of pesticides, being affected by a number of factors, such as water temperature, infusion time, tea to water ratio, preparation process (TWR) (Cho et al. 2014; Florea et al. 2020). In this study, the tea infusion was prepared according to the manufacturer's recommended instructions (similar conditions), so we can exclude variations due to the method of infusion preparation. However, other factors such as solubility properties of pesticides and plant species may determine the content of compounds in the infusion, i.e. the plant-infusion transfer rate.

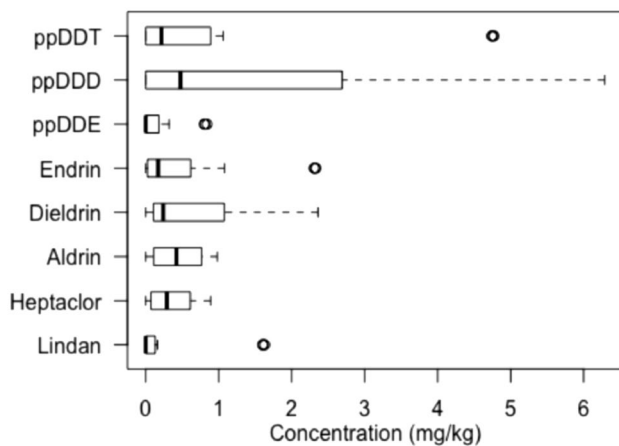
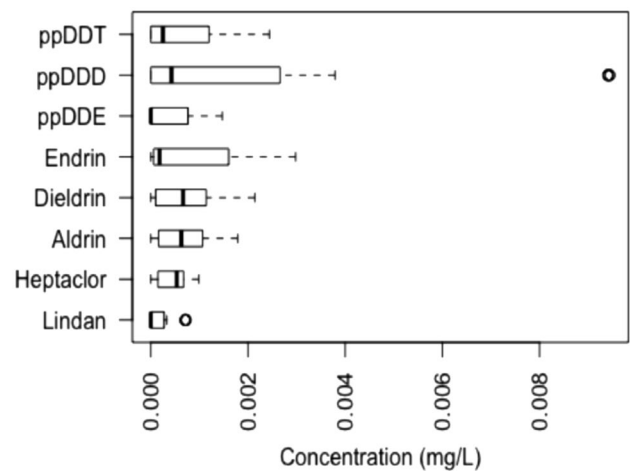
Previous studies have shown that in the traditional tea brewing method (infusion), the transfer rate of pesticide residues largely depends on the water solubility (Ws) and partition coefficient (log Kow) parameters (Chen et al. 2017; Paramasivam and Chandrasekaran 2014). This research indicates that relatively low water solubilities ($< 10 \text{ mg/L}$) and high partition coefficients result in low transfer rates of pesticides in infusions. Organochloride pesticides are hydrophobic substances with high log Kow (octanol water partition coefficient) values, and these pesticides might bind strongly to plant tissues, thus resulting in lower transfer into infusions (Abd El-Aty et al. 2014; Rissato et al. 2015).

On the other hand, each medicinal plant species absorbs contaminants differently; depending on its structure and chemical composition or on the species metabolic particularities, certain naturally occurring components may either reduce the release of contaminants from plant tissue, (Xia et al. 2017), or facilitate their transfer into aqueous infusions (e.g. essential oils with lipophilic molecules) (Hayward et al. 2015).

Table 3 OPCs in herbal tea infusions ($\mu\text{g}\cdot\text{L}^{-1}$)

$\mu\text{g}\cdot\text{L}^{-1}$	<i>Hypericum perforatum</i>	<i>Artemisia absinthium</i>	<i>Tilia sp.</i>	<i>Matricaria chamomilla</i>	<i>Achillea millefolium</i>	<i>Taraxacum officinale</i>	<i>Calendula officinalis</i>	<i>Crataegus monogyna</i>
HCB	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD
Lindane	0.32 ± 0.01	BLD	BLD	0.71 ± 0.10	BLD	BLD	0.234 ± 0.01	BLD
Heptaclo	0.41 ± 0.02	0.29 ± 0.01	0.989 ± 0.04	0.68 ± 0.23	0.677 ± 0.01	0.68 ± 0.01	BLD	BLD
Aldrin	0.4 ± 0.01	0.87 ± 0.03	1.12 ± 0.10	0.32 ± 0.01	1.01 ± 0.50	1.79 ± 0.18	BLD	BLD
Dieldrin	1.21 ± 0.20	BLD	0.29 ± 0.01	2.104 ± 0.12	1.08 ± 0.09	1.041 ± 0.27	BLD	0.21 ± 0.01
Endrin	BLD	0.15 ± 0.01	0.12 ± 0.01	2.982 ± 0.50	2.274 ± 0.20	BLD	0.21 ± 0.00	0.93 ± 0.20
p,p'DDE	1.31 ± 0.65	BLD	BLD	BLD	BLD	BLD	1.474 ± 0.27	0.22 ± 0.01
p,p'DDD	BLD	BLD	BLD	0.87 ± 0.05	3.77 ± 0.62	9.452 ± 1.78	1.533 ± 0.18	BLD
p,p'DDT	BLD	BLD	2.43 ± 0.32	BLD	0.51 ± 0.02	1.02 ± 0.15	1.35 ± 0.51	BLD

BLD below detection limit

**Fig. 1** Boxplot of OPCs content in medicinal plants**Fig. 2** Boxplot of OPCs content in infusions

Risk Assessment of OPCs Pesticide

Unlike green or black tea, herbal teas do not require special processing, such as fermentation (Heshmati et al. 2021), and may thus contain higher levels of contaminants, with risks to consumer health. Therefore, in addition to determinate total plant concentrations, studies on the risks associated with pesticides in herbal tea should also focus on the percentages of pesticides transferred during infusion processes. The results on the health risk of residues of OPCs in the herbal infusions analysed are presented in Table 6. The obtained values show that for the detected compounds, the estimated daily intake (EDI) ranged from $0.33\text{--}10\text{--}3$ and $26\text{--}10\text{--}3$ $\mu\text{g}/\text{kg}/\text{d}$. These values are lower than those for Acceptable Daily Intakes (ADI) proven by FAO/WHO (2017). Also, the values obtained for Hazard Index (HI) were < 1 in all infusions. These results may indicate that the residues of OPCs in the analysed infusions present a minimal risk to human health

(Shoiful et al. 2013). Average values of OPCs concentrations were used to determine the health risk indices.

In the present study, dieldrin had the highest HI value (0.059) for *Matricaria chamomilla*, the lowest value (0.0002) was obtained for ppDDE in *Hypericum perforatum*.

The cumulative hazard index (CHI), showed values less than 1 for all types of tea, suggesting that the population is not exposed to risk after consuming tea prepared from the MPs studied.

In other studies, for each type of pesticide, different values for HI have been reported; these differences may be related to different levels of pesticides in the dried plants, the parameters of the infusion process, as well as the daily consumption of different amounts of tea (Siraj et al. 2021; Jianga et al. 2020).

Table 4 The modified distance correlation between the plant and the infusion for each OPC, the corresponding p-value and the Pearson correlation coefficient

OPC	Lindan	Heptachlor	Aldrin	Dieldrin	Endrin	p,p'DDE	p,p'DDD	p,p'DDT
Modified distance correlation (Value of the test)	0.90073	0.60179	0.76772	1	0.89327	0.91957	0.96841	0.91671
p-value	0.001996	0.003992	0.001996	0.001996	0.001996	0.001996	0.001996	0.001996
Pearson correlation	0.9139	0.3726	0.7808	1	0.8054	0.8606	0.9642	0.9144

Limitations of the Study

The results of the study revealed both different OPC contents in the herbs studied and different transfers from the plant to the infusion. Firstly, a small number of MPs were analyzed, chosen mainly from the most commonly used ones. For a good understanding of how plant species influence the differential transfer of pesticide molecules, further research is needed, working with groups of several plants belonging to the same family.

Conclusions

All samples of MPs contained organochlorine pesticides in different concentrations. For the compounds Heptachlor, p,p' DDT, p,p' DDD, Aldrin, Dieldrin, there were determined high concentrations well above the maximum limit established by the EU Regulation.

The concentration of OPCs in the studied dry MPs varied decreasingly as follows: *Taraxacum officinale* > *Achillea millefolium* > *Tilia sp.* > *Matricaria chamomilla* > *Hypericum perforatum* > *Calendula officinalis* > *Artemisia absinthium*.

In herbal infusions, OPCs vary between 0.36 $\mu\text{g}\cdot\text{L}^{-1}$ (*Achillea millefolium*) and 13.98 $\mu\text{g}\cdot\text{L}^{-1}$ (*Taraxacum officinale*). Among the individual compounds, p,p'DDD presented the highest concentrations (*Taraxacum officinale* and *Achillea millefolium*), and Endrin the lowest concentrations (*Tilia sp.*, *Artemisia absinthium* and *Calendula officinalis*).

The transfer rate of organochlorine pesticides from dried plant in the infusion varied from 3.95% for Heptachlor (*Artemisia absinthium*) to 57.71% for dieldrin (*Achillea millefolium*). The compounds p,p'DDD and p,p'DDT indicated lower values of the transfer rate for most medicinal plants. The statistical analysis provided sufficient evidence that the OPC contents in the MPs and infusion are statistically correlated.

The health risk assessment indicated HI and CHI values < 1 in all types of infusions. These results indicate that the residues of OCPs in the analysed infusions present a minimal risk to human health. However, taking into account the large transfer variations obtained, determined either by the characteristics of the OCP molecules or by the plant species, the results of the study recommend the development of control protocols for medicinal plant infusions intended for human therapy that include the measurement of OCPs concentrations.

Table 5 Transfer rate (%)

	<i>Hypericum perforatum</i>	<i>Artemisia absinthium</i>	<i>Tilia sp.</i>	<i>Matricaria chamomilla</i>	<i>Achillea millefolium</i>	<i>Taraxacum officinale</i>	<i>Calendula officinalis</i>	<i>Crataegus monogyna</i>
HCB	0	0	0	0	0	0	0	0
Lindane	14.22	0	0	5.29	0	0	17.55	0
Heptachlor	18.20	3.95	34.58	10.38	54.16	19.90	0	0
Aldrin	8.53	24.27	15.44	11.13	48.40	22.14	0	0
Dieldrin	15.61	0	9.40	10.87	57.71	52.01	0	10.97
Endrin	0	9.27	9.17	35.61	11.77	0	47.54	55.8
p,p'DDE	19.35	0	0	0	0	0	46.03	57.39
p,p'DDD	0	0	0	10.76	10.35	18.08	17.94	0
p,p'DDT	0	0	6.11	0	13.90	11.77	21.34	0

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

Table 6 Estimated daily intake (EDI), hazard index (HI) and cumulative hazard index (CHI) of OPCs in tea infusions

	Hypericum perforatum		Artemisia absinthium		Tilia sp.		Matricaria chamomilla		Achillea millefolium		Taraxacum officinale		Calendula officinalis		Crataegus monogyna	
	ADI	EDI	HI	EDI	HI	EDI	HI	EDI	HI	EDI	HI	EDI	HI	EDI	HI	EDI
Lindane	3	0.89·10 ⁻³	2.9·10 ⁻³	—	—	—	—	1.98·10 ⁻³	0.66·10 ⁻³	—	—	—	0.6·10 ⁻³	0.2·10 ⁻³	—	—
Heptaclor	0.5	1.1·10 ⁻³	2.2·10 ⁻³	0.8·10 ⁻³	1.6·10 ⁻³	2.7·10 ⁻³	5.4·10 ⁻³	1.9·10 ⁻³	3.8·10 ⁻³	1.8·10 ⁻³	1.9·10 ⁻³	3.8·10 ⁻³	—	—	—	—
Aldrin	0.1	1.3·10 ⁻³	13·10 ⁻³	2.4·10 ⁻³	24·10 ⁻³	3.1·10 ⁻³	31·10 ⁻³	0.89·10 ⁻³	8.9·10 ⁻³	2.8·10 ⁻³	5.0·10 ⁻³	50·10 ⁻³	—	—	—	0
Dieldrin	0.1	3.3·10 ⁻³	33·10 ⁻³	—	—	0.81·10 ⁻³	8.1·10 ⁻³	5.9·10 ⁻³	59·10 ⁻³	3.0·10 ⁻³	2.9·10 ⁻³	29·10 ⁻³	—	—	0.59·10 ⁻³	5.9·10 ⁻³
Endrin	0.2	—	—	0.43·10 ⁻³	2.15·10 ⁻³	0.33·10 ⁻³	1.6·10 ⁻³	8.3·10 ⁻³	41.5·10 ⁻³	6.36·10 ⁻³	—	—	0.59·10 ⁻³	295·10 ⁻³	2.6·10 ⁻³	13·10 ⁻³
p,p'DDE	1	3.67·10 ⁻³	3.67·10 ⁻³	—	—	—	—	—	—	—	—	—	4.1·10 ⁻³	4.1·10 ⁻³	0.11·10 ⁻³	0.11·10 ⁻³
p,p'DDD	1	—	—	—	—	—	—	2.4·10 ⁻³	2.4·10 ⁻³	10.5·10 ⁻³	26·10 ⁻³	26·10 ⁻³	4.2·10 ⁻³	4.2·10 ⁻³	—	0
p,p'DDT	20	—	—	—	—	6.8·10 ⁻³	0.34·10 ⁻³	1.4·10 ⁻³	0.07·10 ⁻³	1.4·10 ⁻³	2.8·10 ⁻³	0.14·10 ⁻³	3.8·10 ⁻³	0.19·10 ⁻³	—	0
CHI		0.054		0.0277		0.0464		0.1162		0.1039		0.1089		0.3036		0.0190

ADI acceptable daily intake(μg/kg/d), EDI estimated daily intakes (μg/kg/d), HI hazard index, CHI cumulative hazard index

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Authors Contributions MMB, SB and CB conceived, designed, analysed data, prepared and led revision of the manuscript. NAD, and performed laboratory analyses. VC supervised laboratory work and performed data review. SB, MMB, CB, VC, and NAM contributed to the editing and revision of the manuscript.

Declarations

Conflict of Interest No potential conflict of interest was reported by the author(s).

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