

RADIONUCLIDES CONTENT IN SOME MEDICINAL PLANTS COMMONLY USED IN ROMANIA

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Abstract

The aim of this work was the evaluation of the radioactive contamination level in some medicinal plants commonly used in Romania, available as herbal teas. The selected plant species were: *Tilia cordata*, *Matricaria chamomilla*, *Calendula officinalis*, *Ocimum basilicum*, *Achillea millefolium* and *Hypericum perforatum*. The investigation of radioactive contamination was achieved by assessing global alpha and beta activity, and by determining the activity of natural ²¹⁰Po, ²¹⁰Pb, ²³²Th, ²³⁸U radionuclides, and artificial ¹³⁷Cs, ⁹⁰Sr radionuclides. The values of artificial ¹³⁷Cs and ⁹⁰Sr radionuclides were below the detection limit (60 mBq/kg). The maximum levels of natural radionuclides were recorded as follows: ²¹⁰Po and ²³⁸U in *Ocimum basilicum* and *Achillea millefolium* (8 mBq/kg and 40 mBq/kg, respectively), ²¹⁰Pb in *Matricaria chamomilla*, *Achillea millefolium* and *Hypericum perforatum* (30 mBq/kg) and ²³²Th in *Achillea millefolium* and *Hypericum perforatum* (60 mBq/kg). The values detected for the investigated radionuclides were lower than EU maximum limits or those reported in literature. *Achillea millefolium* showed the strongest tendency to accumulate natural ²¹⁰Pb, ²¹⁰Po, ²³²Th and ²³⁸U radionuclides.

Rezumat

Tema acestei lucrări o constituie evaluarea nivelului de contaminare radioactivă a unor plante medicinale folosite frecvent în România și care sunt condiționate sub formă de ceaiuri. Speciile vegetale analizate au fost: *Tilia cordata*, *Matricaria chamomilla*, *Calendula officinalis*, *Ocimum basilicum*, *Achillea millefolium* și *Hypericum perforatum*. Investigarea contaminării radioactive s-a realizat prin evaluarea activității alfa și beta globale, precum și prin determinarea activității radionuclizilor naturali ²¹⁰Po, ²¹⁰Pb, ²³²Th, ²³⁸U și artificiali ¹³⁷Cs, ⁹⁰Sr. Valorile radionuclizilor artificiali ¹³⁷Cs și ⁹⁰Sr au fost sub limita de detecție (60 mBq/kg). Nivelele maxime ale radionuclizilor naturali au fost înregistrate după cum urmează: ²¹⁰Po și ²³⁸U în *Ocimum basilicum* și *Achillea millefolium* (8 mBq/kg și respectiv, 40 mBq/kg), ²¹⁰Pb în *Matricaria chamomilla*, *Achillea millefolium* și *Hypericum perforatum* (30 mBq/kg), iar ²³²Th în *Achillea millefolium* și *Hypericum perforatum* (60 mBq/kg). Valorile concentrațiilor detectate pentru radionuclizii investigați au fost inferioare limitelor maxime stabilite pe

plan european sau celor raportate în literatură. *Achillea millefolium* a prezentat cea mai pronunțată tendință de acumulare a radionuclizilor naturali ^{210}Pb , ^{210}Po , ^{232}Th și ^{238}U .

Keywords: medicinal plants, radioactivity contamination, radionuclides.

Introduction

Medicinal plants were and they continue to be widely used in the world to prevent or treat various diseases. It is estimated that about 25% of all modern medicines are derived directly or indirectly from medicinal plants. Also, about 80% of the world population (especially in developing countries) uses herbal medicine as the primary source of health care [1]. The current global increase in the consumption of herbal products, in the conditions of a scientifically validated use, raises the issue of compliance with certain criteria of quality, safety and efficacy. Exogenous contaminants, such as pesticides, microorganisms, heavy metals and radionuclides, can affect both the therapeutic properties of herbal products and the health of patients. Radioactive contamination became a major concern for the pharmaceutical industry as a result of the Chernobyl incident and it is a determining factor in the quality control of medicinal plants [6]. Medicinal plants' capacity to absorb radionuclides, both from the atmosphere and from the soil, favours the transfer to human body of these contaminants, thereby contributing to the increase of the level of internal irradiation of the consuming population [7]. WHO emphasizes that the risks on consumers' health depend both, on the specific of the radionuclides and on the level of contamination, as well as on the duration of their use and the administered dose [17]. Lately, the interest in the determination of the transfer of radionuclides from the environment to humans increased, and implicitly the contribution of herbal products' consumption to the level of internal irradiation [2, 3]. So far, in Romania, little study was made on the determination of the radioactive content of plant species [5, 9]. The assessment of radioactive contamination of medicinal plants not only contributes to determining the quality of the plant material, but it also provides useful information on the safety level of their consumption by humans. The aim of this work was the evaluation of the radioactive contamination level in some medicinal plants commonly used in Romania, available as herbal teas.

Materials and Methods

Chemicals and apparatus

All chemicals and reagents were of analytical grade and were purchased from Merck (Germany). All spectrophotometric measurements were performed on Spekol 11 spectrophotometer (Germany).

Plant material

The vegetal products investigated were: flowers of linden (*Tilia cordata*, Tiliaceae), chamomile (*Matricaria chamomilla*, Asteraceae) and marigold (*Calendula officinalis*, Asteraceae) as well as the flowering aerial parts of basil (*Ocimum basilicum*, Lamiaceae), yarrow (*Achillea millefolium*, Asteraceae) and St. John's Wort (*Hypericum perforatum*, Hypericaceae). Plant species were chosen because they are common plants, easily accessible and frequently used in Romania as medicinal tea. The samples were purchased in bulk packaged form SC Fares SA Orăștie, in March 2012.

Sample preparation

Plant samples were brought in the form of white ash through complete drying (2 kg of each sample, 105°C, 48 h) and calcination in the furnace (initially 105°C, 48 h, and then gradually increasing the temperature by 50°C to 450°C, 12 days, and then maintaining it at 450°C, 48 h) [10].

Measurement of the global alpha and beta activity

It was performed using the global low-background alpha and beta counting system (PROTEAN MPC 2000, USA). The standard alpha/beta sources were: ^{241}Am (alpha source) and $^{90}\text{Sr}+^{90}\text{Y}$ (beta source). The calculation of the global alpha activity (Λ), respectively the beta one, was performed using the formula:

$$\Lambda = G \times (R-F)/m \times M \times \epsilon \quad \text{Bq/kg,}$$

where: G=mass of the total residue (g); R=global count rate of the sample (impulses/sec); F=count rate corresponding to the radiation background (impulses/sec); m=mass of residue deposited on the tray (g); M = mass of fresh sample (kg); ϵ =the measuring efficiency of the device determined by a standard source (namely alpha, beta).

The measuring efficiency of the device (ϵ), determined through a standard alpha or beta source, was calculated using the formula:

$$\epsilon \text{ alpha/beta} = (R_s - F)/\Lambda_{\text{calc}},$$

where: R=the count rate of the standard source (impulses/sec); F=the count rate corresponding to the radiation background (impulses/sec); Λ_{calc} =the alpha/beta activity of the calibration source (Bq/kg).

Measuring the activity of ^{137}Cs , ^{210}Po , ^{210}Pb , ^{90}Sr , ^{232}Th , ^{238}U radionuclides

The determination of ^{137}Cs activity was carried out after the separation in an acid environment, by adsorption on ammonium phosphomolybdate [12].

After separation from ^{45}Ca , ^{140}Ba , ^{226}Ra , ^{210}Pb , and in the presence of the Sr pace, ^{90}Sr precipitated in the form of strontium carbonate in the presence of ammonium carbonate. The determination of ^{90}Sr activity was achieved by drying the precipitate and measuring the beta activity using the low background global alpha and beta system [13]. ^{232}Th and ^{238}U were

determined quantitatively according to the concentration through absorption on active charcoal. After the separation of ^{238}U from ^{232}Th and their purification by ion exchangers, the complex of arsenazo (III)- Th^{4+} , respectively arsenazo (III)- U^{4+} was measured spectrophotometrically [14]. The determination of ^{210}Po [15] and ^{210}Pb [16] concentrations was done after their separation. ^{210}Po from the sample was electrochemically deposited on a nickel disc, and after reaching ^{210}Po - ^{210}Pb equilibrium, ^{210}Pb was determined by repeating the electrochemical deposition process.

Results and Discussion

The analysed samples presented a global alpha activity ranging between 3.20 Bq/kg (*Ocimum basilicum*) and 10.75 Bq/kg (*Hypericum perforatum*), while the global beta activity ranged between 214.5 Bq/kg (*Calendula officinalis*) and 429.40 Bq/kg (*Hypericum perforatum*) (Table I).

Table I

The average global alpha and beta activity (Bq/kg) in the samples of medicinal plants

Samples	Sample mass (kg)	Ash mass (g)	Global alpha activity (Bq/Kg)	Global beta activity (Bq/Kg)
<i>Flowers</i>				
<i>Tilia cordata</i>	2	191.20	4.57	249.30
<i>Calendula officinalis</i>	2	171.40	5.41	214.50
<i>Matricaria chamomilla</i>	2	185.20	4.51	411.72
<i>Aerial parts</i>				
<i>Ocimum basilicum</i>	2	295.00	3.20	380.21
<i>Achillea millefolium</i>	2	95.80	6.54	333.45
<i>Hypericum perforatum</i>	2	95.00	10.75	429.40

In the samples of the investigated species, the concentrations of artificial radionuclides (^{137}Cs and ^{90}Sr) were below the specific detection limit (60 mBq/kg) (Table II).

Table II

Radionuclides concentrations (mBq/kg) in the samples of medicinal plants

Vegetal product	Species	Artificial radionuclides (mBq/Kg)		Natural radionuclides (mBq/Kg)			
		^{137}Cs	^{90}Sr	^{210}Pb	^{210}Po	^{232}Th	^{238}U
flowers	<i>Tilia cordata</i>	< 60	< 60	-	1	-	10
	<i>Calendula officinalis</i>			10	1	10	20
	<i>Matricaria chamomilla</i>			30	1	50	10
aerial parts	<i>Ocimum basilicum</i>			20	8	50	40
	<i>Achillea millefolium</i>			30	8	60	40
	<i>Hypericum perforatum</i>			30	6	60	30

In terms of natural radionuclides, ^{210}Pb and ^{232}Th levels were below the detection limit of the device in the case of *Tilia cordata* species. The maximum concentration of ^{210}Pb (30 mBq/kg) was recorded in the samples of *Matricaria chamomilla*, *Achillea millefolium* and *Hypericum perforatum*. Also, ^{232}Th was detected in higher concentrations in *Achillea millefolium* and *Hypericum perforatum* (60 mBq/kg). The maximum levels of ^{210}Po and ^{238}U were determined in samples of *Ocimum basilicum* and *Hypericum perforatum* (8 mBq/kg and 40 mBq/kg, respectively) (Table II).

The alpha and beta particles exert their harmful action on humans by ingestion or inhalation, which is why determining their activity in medicinal plants is an important indicator in the assessment of radioactive contamination of the human body. According to the determination of global alpha and beta activity, *Ocimum basilicum* showed a minimum global alpha value (3.20 Bq/kg), while for global beta, the lowest was determined in *Calendula officinalis* (214.50 Bq/kg). Unlike these two, *Hypericum perforatum* had maximum values both for the global alpha and beta activity (10.75 Bq/kg and 429.40 Bq/kg, respectively). A study carried out in our country in 2005, reported for the aerial part of the *Calendula officinalis* species, collected from Arad County, a global alpha activity below the detection limit of the device, and a global beta activity of 204 Bq/kg. The absence of radioactive contamination of the cultivated species is correlated with normal radioactivity determined for the cultivation area and it demonstrates the influence of soil in the transfer of radionuclides in plants [9]. The slightly higher values obtained in this study for the same species (5.41 Bq/kg and 214.5 Bq/kg, respectively) remain within the same range of the radioactive contamination absence.

The concentrations of ^{137}Cs and ^{90}Sr artificial radionuclides in the analysed medicinal plants were below the detection limit of the device, and under the EU maximum limits in the case of contamination by nuclear accidents or other radiological emergencies: isotopes of Sr, mainly ^{90}Sr - 7500 Bq/kg; nuclides of half-life greater than 10 days, mainly ^{137}Cs - 12500 Bq/kg [11]. Also, the values recorded for natural radionuclides were lower than those reported by other authors [4, 8]. Long half-life radionuclides (^{232}Th , ^{238}U , ^{210}Pb) were detected in higher concentrations in all samples analyzed. Compared to the vegetal products flowers that were analyzed, the aerial parts of *Hypericum perforatum*, *Ocimum basilicum* and *Achillea millefolium* showed a tendency to concentrate ^{210}Po and ^{238}U radionuclides. Also, *Achillea millefolium* showed the most pronounced tendency to accumulate all investigated ^{210}Pb , ^{210}Po , ^{232}Th and ^{238}U natural radionuclides.

Conclusions

Tilia cordata, *Matricaria chamomilla*, *Calendula officinalis*, *Ocimum basilicum*, *Achillea millefolium* and *Hypericum perforatum* samples contain undetectable levels of artificial radionuclides (^{137}Cs , ^{90}Sr) and low levels of natural radionuclides (^{210}Pb , ^{210}Po , ^{232}Th , ^{238}U) that do not pose a risk to the health of medicinal teas consumers.

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