

## HEALTH RISK ASSESSMENT OF NATURAL AND ARTIFICIAL RADIONUCLIDES IN MEDICINAL PLANTS

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**ABSTRACT.** In the present work, we determined activity concentrations of ten Medicinal herbs using gamma spectrometry. The radioactivity concentration of <sup>226</sup>Ra, <sup>232</sup>Th, <sup>40</sup>K and <sup>137</sup>Cs were 2.82, 0.63, 984.32, 0.28 Bq/kg, respectively. These doses are not hazardous to the general public's health. The mean values of radium equivalent activity (Raeq), absorbed dose rates (D), annual effective dose (De) and external hazard index (Hex) are 79.51 Bq/kg, 42.73 nGy<sup>-1</sup>, 52.40 μSv and 0.21, respectively. Medicinal plants used to make tea do not contain a concentration of tested radionuclides that would cause negative consequences to one's health.

**Keywords:** medicinal herbs, gamma spectrometry, radiological hazard, radioactivity concentrations.

### INTRODUCTION

The ability to determine the effects of radiation exposure from both terrestrial and cosmogenic sources includes knowledge of radionuclide distribution and radiation levels in the surrounding environment. The radionuclides of the uranium and thorium series, as well as <sup>40</sup>K, are the major sources of natural radioactivity in the terrestrial world (JEVREMOVIC *et al.*, 2011). It has been pointed out that cancer in living tissues has resulted from radiation emission caused by radioactive nuclides. As a result, the amount of radionuclides in human food must be determined since it presents a high risk for cancer. Global organizations have put emphasis on recognizing the impact of radiation on the population in order to take effective intervention steps to minimize the effects of radiation (UNSCEAR 2000; DA SILVA *et al.*, 2018; GARCÉZ *et al.*, 2018b). Plants have been recognized to be contributors to the radioactive fallout effect as they provide a large surface that increases the effect of the radioactive fallout. In addition, plants could absorb the radionuclide from the ground through roots thus increasing the risk of radiation. Plants that have been used in traditional medicine have had a rich history all over the world (KANDIĆ *et al.*, 2020). The therapeutic and pharmacological properties of most of them are well-known and a growing number of people

have been choosing herbal medicine to boost their health conditions in recent decades (PETROVSKA, 2012; DAR *et al.*, 2017). Notably, Medicinal herbs are a good example of plants that provide a wide surface to the radioactive fallout since they grow approximately to a height of five meters and have broader leaves (JIBIRI *et al.*, 2016). As a result, the plants used in traditional medicine are outstanding plants for tracking and researching radioactive fallout. Nuclear dust and ashes emitted in the atmosphere reach the earth's ground and plants via air or raindrops on the earth's surface, later settling on plant leaves or the soil. Rainfall and air are considered agents for radioactive fallout on the earth's surface (HIRONO and NONAKA, 2016; ZEHRINGER *et al.*, 2018). Research studies have been conducted on soil and plant radioactivity, as well as on environment effect assessment by use of gamma-ray spectrometry. Detection efficiency is an important variable when measuring sample activity and is usually determined using standard sources with characteristics that include chemical composition, density, and physical attributes. The difference that could be registered when determining detection efficiency could be minimized if Marinelli beakers are utilized in the procedure (ABDI *et al.*, 2006). Dose estimation is one of the most significant radiological risks. The annual intake of radionuclides is being used to measure the dose. The cancer risk and the committed effective dose are critical in determining the effect it will have, thus appropriate intervention mechanisms are deployed to mitigate radioactive impacts (GARCÊZ, 2018a; LOPES *et al.*, 2020).

The aim of this study is to determine the activity concentrations of natural and artificial radionuclides and radiological hazard parameters in ten Medicinal herbs that could be used as tea. Medicinal herbs used as spices could be more harmful than tea, so additional tests are needed before we could say with certainty that the use of medicinal herbs is safe.

## MATERIALS AND METHODS

### *Sample collection and preparation*

Samples were purchased at health food stores in Kragujevac, Serbia. The names of tested medicinal herbs are given in Table 1.

Table 1. List of the medicinal herbs grown on plantations in Serbia, used for radioactivity analysis.

Herbs	
Binomial nomenclature	English name
<i>Mentha piperita</i>	Peppermint
<i>Matricaria chamomilla</i>	Chamomile
<i>Equisetum arvense</i>	Horsetail
<i>Hibiscus sabdariffa</i>	Roselle
<i>Punica granatum</i>	Pomegranate
<i>Cassia sena</i>	Cassia
<i>Arctostaphylos uva-ursi</i>	Bearberry
<i>Urtica dioica</i>	Nettle
<i>Hypericum perforatum</i>	St. John's wort
<i>Achillea millefolium</i>	Yarrow

The samples were homogenized and sieved through a 170 mesh for radiometric analysis. The radionuclide activity concentration was determined using the dry weight value. The samples were put in a 450 mL polypropylene container with low background radiation. Plant samples were kept in the laboratory for 35 days to attain a secular equilibrium balance

(DA SILVA, 2018). A digital scale with a sensitivity of  $\pm 0.01$  g was used to weigh the samples.

### ***Radiactivity measurement***

The specific activities of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ ,  $^{40}\text{K}$  and  $^{137}\text{Cs}$  were measured using a coaxial HPGe detector (GEM30-70, ORTEC) with a relative efficiency of 30 % and energy resolution (FWHM) of 1.85 keV at 1.33 MeV ( $^{60}\text{Co}$ ). Gamma-activity of each sample was measured for 48 hours and it was determined through the intensity of emission lines in the spectrum after background subtraction. The specific activity of  $^{226}\text{Ra}$  was obtained as the weighted average activity of three separate gamma-ray lines of its decay products,  $^{214}\text{Pb}$  (351.9 keV) and  $^{214}\text{Bi}$  (609.3 and 1764.5 keV). The specific activity of  $^{232}\text{Th}$  was determined by the gamma-ray lines of  $^{228}\text{Ac}$  (at the energies of 338.3, 911.1 and 968.9 keV) and  $^{208}\text{Tl}$  (at the energies of 583.0 and 860.6 keV). The gamma ray lines at 1460.7 and 661.6 keV were used for estimating specific activities of  $^{40}\text{K}$  and  $^{137}\text{Cs}$ , respectively (MILENKOVIC *et al.*, 2015). For activity and background, a timer of 252,000 seconds was set. The background spectra were used to correct the isotopes' gamma-ray net peak areas. MAESTRO, a computer program, was used to perform the spectrum analysis.

Activity concentrations were obtained using Equation (1):

$$AC = \frac{N_L}{e \times m \times t \times P_g} \quad (1)$$

where:  $AC$  is the activity concentration (Bq/kg),

$N_L$  the net area of photopic interest,

$m$  is the sample mass (kg),

$e$  counting efficiency by a specific energy,

$P_g$  is the emission probability of the measured gamma-ray and

$t$  is the counting time (s).

### ***Calculation of radiological hazard***

The absorbed dose rate is a direct relationship between the radioactivity concentrations of radionuclides and their exposure. The formula (2) was used to measure the mean activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  (Bq/kg) in the given samples at 1 m above the ground surface (UNSCEAR, 2008):

$$D(\text{nGy} \cdot \text{h}^{-1}) = 0.462C_{\text{Ra}} + 0.604C_{\text{Th}} + 0.0417C_{\text{K}} \quad (2)$$

where,  $D$  is the absorbed dose rate in  $\text{nGy} \cdot \text{h}^{-1}$ , while  $C_{\text{Ra}}$ ,  $C_{\text{Th}}$  and  $C_{\text{K}}$  are the activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$ , respectively.

The annual effective dose was calculated by employing the conversion coefficient of  $0.7 \text{ Sv} \cdot \text{Gy}^{-1}$ :

$$D_E(\mu\text{Sv}) = 0.7 \cdot D \cdot t \cdot p, \quad (3)$$

where  $t$  represents the annual exposure time (8760 h) while  $p$  is the outdoor occupancy factor of 0.2 for time spent outdoors, implying that 20% of time is spent outdoors.

Due to a non-uniform distribution of natural radionuclides, the actual activity level of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in the samples could be evaluated by means of a common radiological index such as radium equivalent activity (Raeq). The specific activity of materials containing

different amounts of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  according to BERETKA and MATHEW (1985) was calculated.

$$Raeq(\text{Bq/kg}) = C_{Ra} + 1.43C_{Th} + 0.077C_K. \quad (4)$$

The activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  in Bq/kg are represented by  $C_{Ra}$ ,  $C_{Th}$ , and  $C_K$ , respectively. The maximum radium equivalent operation that could be tolerated is 370 Bq/kg. The external hazard index, based on a criterion, has been implemented to restrict the radiation exposure attributable to natural radionuclides to the acceptable does equivalent limit of  $1 \text{ mSv.y}^{-1}$ . A model proposed by KRIEGER (1981) and BELIVERMIS (2010) has been used in the samples.

$$Hex = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810}. \quad (5)$$

The value of the external hazard index must not exceed the limit of unity in order to keep the radiation hazard to a minimum.

## RESULTS AND DISCUSSION METHODS

The measurement results of activity concentrations of natural radionuclides and  $^{137}\text{Cs}$ , as well as radiological hazard parameters are presented in Table 2.

Table 2. Descriptive statistics of radionuclide concentrations, air-absorbed dose rates, annual effective doses, Ra equivalent activity and external hazard index in analyzed plant samples.

Sample	$^{226}\text{Ra}$ (Bq/kg)	$^{232}\text{Th}$ (Bq/kg)	$^{40}\text{K}$ (Bq/kg)	$^{137}\text{Cs}$ (Bq/kg)	D (nGy/y)	De ( $\mu\text{Sv}$ )	Raeq (Bq/kg)	Hex
<i>M. piperita</i>	3.21	0.29	1156.36	0.61	49.88	61.17	92.66	0.25
<i>M. chamomilla</i>	3.04	0.81	1225.89	0.17	53.01	65.02	98.59	0.27
<i>E. arvense</i>	3.25	0.88	1247.70	0.68	54.06	66.30	100.58	0.27
<i>H. sabdariffa</i>	1.97	0.98	852.24	0.12	37.04	45.43	68.99	0.19
<i>P. granatum</i>	2.64	0.41	698.45	0.23	30.59	37.52	57.01	0.15
<i>C. sena</i>	3.89	0.69	1147.25	0.36	50.05	61.39	93.21	0.25
<i>A. uva-ursi</i>	2.69	0.45	963.14	0.24	41.68	51.11	77.50	0.21
<i>U. dioica</i>	2.56	0.73	1098.78	0.35	47.44	58.18	88.21	0.24
<i>H. perforatum</i>	1.35	0.22	441.03	0.03	19.15	23.48	35.62	0.10
<i>A. millefolium</i>	3.62	0.81	1012.34	0.06	44.38	54.42	82.73	0.22
<b>Min</b>	1.35	0.22	441.03	0.03	19.15	23.48	35.62	0.10
<b>Max</b>	3.89	0.98	1247.73	0.68	54.06	66.30	100.58	0.27
<b>Mean</b>	2.82	0.63	984.32	0.28	42.73	52.40	79.51	0.21
<b>SD</b>	0.72	0.25	243.42	0.21	10.49	12.87	19.50	0.05

Specific activities of radionuclides in samples were ordered in the following way:  $^{40}\text{K} > ^{226}\text{Ra} > ^{232}\text{Th} > ^{137}\text{Cs}$ . The activity of  $^{226}\text{Ra}$  in ten plant samples is in the range from 1.35 to 3.89 Bq/kg with a mean value of 2.82 Bq/kg. *Cassia sena* and *H. perforatum* had the highest and lowest activity concentrations, respectively. The activity of  $^{232}\text{Th}$  is in the range from 0.22 to 0.98 Bq/kg with a mean value of 0.63 Bq/kg. The highest and lowest activity concentrations were found in *H. sabdariffa* and *H. perforatum*, respectively. Even though the levels of  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  are very high according to the test samples of the soil collected in

Serbia and other regions, it is interesting that medicinal herb products do not have such a high concentration of radionuclide activity (JEVREMOVIC *et al.*, 2011). The activity of  $^{40}\text{K}$  is in the range from 441.03 to 1247.73 Bq/kg with a mean value of 984.32 Bq/kg. The highest activity concentration of  $^{40}\text{K}$  was found in *E. arvensis*. This could be explained by the fact that plants have a high radionuclide transfer factor due to complex metabolic processes involving potassium. Additionally, the use of fertilizer may be another factor causing the  $^{40}\text{K}$  activity concentration increase (GARCÊZ, 2018a). The activity of  $^{137}\text{Cs}$  is in the range from 0.03 to 0.68 Bq/kg with a mean value of 0.28 Bq/kg.  $^{137}\text{Cs}$  appears to have negligible activity in most samples, especially in those two found to have 0.03 Bq/kg. In some Ukrainian and Middle Eastern countries, such as Turkey,  $^{137}\text{Cs}$  concentrations could be ten or more times higher than those measured in this analysis (HANDL *et al.*, 2003; DI GREGORIO *et al.*, 2004; DOWDALL *et al.*, 2005; KILIÇ *et al.*, 2009). According to JEVREMOVIC *et al.* (2011) and DJELIC *et al.* (2016) the results for the medicinal herbs *M. piperita* and *M. chamomilla* show some deviation. While the values for  $^{40}\text{K}$  are slightly higher, for  $^{232}\text{Th}$  smaller values were obtained.  $^{137}\text{Cs}$  is in agreement with the previously mentioned research. If we further compare natural radionuclides and  $^{137}\text{Cs}$  for *U. dioica*, *H. perforatum* and *A. millefolium*, according to DJELIC *et al.* (2016), we could conclude that only  $^{40}\text{K}$  shows significantly higher concentrations, while the values are significantly lower for  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{137}\text{Cs}$ .

As presented in Table 2, the radiological hazard parameters are assessed from the results of the activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  in the samples of medicinal herbs. The values for the absorbed dose rate range from 19.15 to 54.06 nGy.h<sup>-1</sup> with a mean value of 42.73 nGy.h<sup>-1</sup>. This value does not exceed the recommended international levels of 59 nGy.h<sup>-1</sup>. The value of the annual effective dose varies and may be from 23.48 to 66.30 µSv with a mean value 52.40 smaller than the annual average value of 66 µSv for external exposure to natural terrestrial sources of radiation (AL-ALAWY, 2020). The maximum value of calculated radium equivalent activity (100.58 Bq/kg) does not exceed the recommended value of 370 Bq/kg, which corresponds to a dose limit of 1 mSv for the general population (ICRP, 1990). In Table 2 the calculated values of the external hazard index with a mean value of 0.21 < 1 are also listed. The estimation of the average risk indicators is safe for radiation hazards. The findings of the study indicate that the obtained values are less than the average world limits. The current study and results may be an excellent overture for further research into herbal tea. In further studies, factors such as age and gender may be considered.

## CONCLUSION

Natural and artificial radioactivity levels of ten selected medicinal plants commonly used in Serbia were investigated using gamma spectrometry. The average values of specific activities of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ ,  $^{40}\text{K}$  and  $^{137}\text{Cs}$  were 2.82 Bq/kg, 0.63 Bq/kg, 984.32 Bq/kg, 0.28 Bq/kg, respectively. As a result, the study shows that the tested samples have no noticeable radioactivity except for  $^{40}\text{K}$ . The absorbed dose rates, the annual effective doses, radium equivalent activities, external hazard indexes were also estimated, and the mean values were 42.73 nGy.y<sup>-1</sup>, 52.40 µSv, 79.51 Bq/kg, and 0.21 respectively. All radiological hazard parameters are below the standard proposed by UNSCEAR (2000). Thus, quality monitoring must be carried out in the analysis of radionuclide concentrations in order to confidently discuss the safety of medicinal herb consumption.

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