Activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs radionuclides in Turkish medicinal herbs, their ingestion doses and cancer risks

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Abstract

Twenty-two medicinal herb samples, each representing a distinct species, were collected from Turkish markets and measured by gamma spectrometric method. The activity concentration of ^{226}Ra in medicinal herbs was found in the range of minimum detectable activity (MDA) and 15.1±2.2 Bqkg^{-1}. The activity concentration of ^{232}Th was ranged from MDA values to 3.5±0.8 Bqkg^{-1}. The activity concentration of ^{40}K was varied between 50.0±16.8 and 1311.5±57.3 Bqkg^{-1}. All ^{137}Cs activity concentrations of medicinal herbs were found to have lower than MDA values. The bone surface dose, lower large intestine and colon doses were found to be 182.9 μSvy^{-1}, 18.8 μSvy^{-1} and 18.7 μSvy^{-1} respectively. The highest committed effective dose originated from annually ingestion 1kg medicinal herb was calculated notably low as 9.0 μSv. The cancer risk of ingestion of medicinal herbs was found to be small enough to be neglected. The selected Turkish medicinal herbs are considered safe for human consumption.

Keywords: activity concentration, radionuclide, gamma spectrometry, medicinal herb, ingestion dose, cancer risk

1. Introduction

The naturally occurring radionuclides, which are ^{238}U and ^{232}Th decay series and ^{40}K, are the main sources of radiation in the earth's crust. The activity concentration of these primordial radionuclides in soil and many rocks are generally at low levels. However, there are encountered some soils and rocks with high activity concentrations in the earth’s crust because of their mineral contents. In some cases, radionuclides are accumulated themselves in the soil due to the natural causes. Sometimes, activity concentrations of natural radionuclides in soil may reach to elevated levels subsequent mining or similar industrial activities (1). Moreover, artificial radionuclides (^{131}I, ^{137}Cs etc.) arising from radioactive fallout subsequent to nuclear accidents or nuclear tests may cause pollution of soil (2). Both natural and artificial radionuclides with high activity concentrations may pose a danger for individuals because of external exposure and especially internal exposure resulting from the ingestion of radionuclide through the food chain (3).

Root uptake is the initial and significant step of radionuclide transfer from soil to the plant in the food chain. The potential harm of the radionuclide taken into the body through plant to the individual is closely related to the behaviour of radionuclide in the body. Approximately 80% of radium content taken into body by ingestion leaves the body by faeces and only 20% of them enter the bloodstream. Radium accumulates in bones and teeth, because the chemical structure is very similar to calcium. Release of radium from bones is very slow; therefore, a little amount of ingested radium would not removed from the bones lifelong. Similarly, thorium is taken into body through the intestinal tract and it accumulates on the bone surface (4). Therefore, critical organs where radionuclides most accumulate and their organ doses also need to be taken into account while the radiological evaluation is performed through the effective doses.

Turkey has influenced from artificial radionuclides originated from Chernobyl nuclear accident (5-8). There are still more than forty landfill disposal sites where were stored agricultural products affected by the nuclear accident in the country. Furthermore, there is also known many industrial waste disposal sites containing naturally occurring radionuclides or high background radiation areas such as mines containing uranium and thorium decay series radionuclides (9). A significant portion of these fields is still being used as farmland.

Many studies have been performed about radionuclide content of soil and agricultural products in the country for years (10&11). Most of these studies were related to farmlands and cultivated plants. Limited number of studies was performed about the radionuclide content of the plants that are grows spontaneously in nature and collected by individuals. Medicinal plants, which are used for numerous health benefits, are the most common member of the natural plants group. They were also commonly used as spices and flavourings in traditional meals in the country.

Medicinal plants have been used for different purposes by Turkish people for centuries. Turkish people generally prefer to use unaffected medicinal plants collected from nature - not cultivated or not factory production. Medicinal plants may collect from high background radiation areas and radionuclides may cause undesirable radiation dose to individuals through the food chain. Therefore, determination of the radionuclide content of the herbs is essential in the view of radiation protection measures.

The aim of this work is to determine the natural and artificial radioactivity of selected medicinal herbs for calculation of their ingestion dose. For realize this study, twenty-two kinds of medicinal herbs including some spices and flavourings were collected from traditional Turkish markets. After they were measured by gamma spectrometers, ingestion dose were calculated. It was assumed that 1 kg medicinal herb was consumed per year by individuals for ingestion dose calculations. Cancer risks were calculated and finally results were discussed.

2. Materials and Methods

2.1 *Sampling and sample preparation*

Twenty-two kinds of Turkish origin medicinal herb samples were purchased from markets and traditional spice- sellers of Ankara. Medicinal herb samples were separately labelled and brought to the Laboratory. In order to remove moisture medicinal herb samples were heated in a temperature controlled drying oven at 80ºC until constant mass was obtained. Following, samples were grinded and homogenized. Adequate samples were put into cylindrical plastic analysis containers. Plastic analysis containers have a 6cm diameter and 5cm high. Then, samples were weighed and hermetically sealed with parafilm to prevent escape of radon gas. ^{226}Ra is usually determined using the most intensive gamma transitions of its progenies (^{214}Pb and ^{214}Bi). Thus, sealed samples should be kept at least for 30 days to avoid disequilibrium problems between ^{226}Ra and its progenies before the measurements.

2.2 Radioactivity Measurement

Radioactivity measurements were carried out by using gamma spectrometers in Sarayköy Nuclear Research and Training Centre's Laboratories. Gamma spectrometry laboratory was accredited by TURKAK (Turkish Accreditation Agency) accepted by ILAC (International Laboratory Accreditation Cooperation) in 2009. Gamma spectrometry laboratory has joined comparison tests since then and are audited by TURKAK experts annually. The laboratory has various types of gamma spectrometry systems loaded GAMMAVISION-32 or GENIE-2000 software equipped with n-type or p-type germanium detectors (well type, vertical and horizontal) and DSA-1000 or DSPEC multichannel analyser. Detector relative efficiencies of gamma spectrometry systems are varied between 12% and 150%.

Radioactivity measurements were performed by using a gamma spectrometer, which has a p-type high-purity germanium detector with 451 cm^3 active volume. The detector has 110% relative efficiency and 85:1 peak-to-Compton ratio. The energy resolutions of the detector are 2.1 keV for ^{60}Co at 1332.5 keV and 1.3 keV for ^{57}Co at 122 keV. The detector was installed in front opening split-top shielding. The shielding is composed of 10-cm-thick lead protection, 9.5-mm steel outer housing, 1-mm-thick tin layer, 1.5-mm-thick copper layer. The system was equipped with a DSA-1000 digital spectrum analyser.

Before the measurements, energy calibration was done by using peaks of ^{241}Am, ^{137}Cs and ^{60}Co radionuclides which were obtained from a spectrum of aforementioned radioactive standard point sources. An analytical function was fitted to energy curve plotted versus the channel number of peak centroid.

The efficiency calibration of gamma spectrometry system was performed with a spectrum taken from counted certified radioactive standard volume source, which has the same size with the measurement container of sample. The volume source is 79829-839 coded commercially available source and it has a vegetation matrix similar with the samples. The efficiency calibration source contains 13 radionuclides that have energy range of 59.5-1836.1 keV.

The activity concentrations of the samples were determined from desired radionuclide's own energies or gamma ray photopic of their decay products. The activity concentration of ^{226}Ra was calculated from 295.2, 351.9 keV gamma-ray energies of ^{214}Pb and 609.3 keV of ^{214}Bi. Due to overlapped peaks of ^{235}U (185.7keV) and ^{226}Ra (186keV) radionuclides in the spectrum, 186 keV photopic of ^{226}Ra was not preferred for calculations. The activity concentration of ^{232}Th was calculated from 338.4 and 911.2 keV of ^{228}Ac and 583.2 keV gamma-ray energies of ^{208}Tl. The activity concentration of ^{40}K was determined by using its own energy of 1460.8 keV. The contribution of ^{232}Th via its decay product nuclide ^{228}Ac (1459.2 keV peak) near to the 1460.8 keV peak was neglected because of the small contribution while activity concentration of ^{40}K was calculated (12). Activity concentration of ^{137}Cs was calculated from 661.7 keV photopic energy of ^{137m}Ba radionuclide, which is the daughter product of ^{137}Cs. The activity concentrations of the samples are calculated by the following formula (13):

$$A = \frac{N}{\varepsilon(E_\gamma).P_\gamma.t.M} \qquad (1)$$

where N corresponds the net peak area of gamma-ray energy, $\varepsilon(E_\gamma)$ denotes the absolute efficiency, P_γ is the gamma-ray yield per decay, t and M denotes the counting time and sample mass respectively. Minimum Detectable Activity (MDA) calculations were performed by the following formula (14):

$$MDA = (1.64)\sigma_n/\varepsilon.P.t.w \qquad (2)$$

where σ_n stands for standard deviation of the background in the region of interest and equal square root of the number of count for the background spectrum; ε is absolute efficiency; P is emission probability of gamma decay; t and w denote measurement time and weight of the dried sample respectively.

3. Results and discussion

3.1 *Activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs radionuclides of the samples*

The activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs radionuclides in medicinal herbs are presented with their statistical uncertainties (2σ) in Table 1. A less than sign (<) was used to indicate the below Minimum Detectable Activity (MDA) values of the detector. Only activity concentrations of the radionuclides, which were above the MDA values, were used in the calculations and assessments in this work. The results given in Table 1 indicated that activity concentrations of ^{226}Ra in samples are ranged from Minimum Detectable Activity (MDA) value to 15.1 Bqkg^{-1} with a mean value of 5.9 Bqkg^{-1}. The activity concentrations of ^{232}Th in medicinal herbs were found to be between 1.6 Bqkg^{-1} and 3.5 Bqkg^{-1} with a mean value of 2.8 Bqkg^{-1}. The activity concentrations of ^{40}K varied between 50.0 Bqkg^{-1} and 1311.5 Bqkg^{-1} with a mean value of 500.2 Bqkg^{-1}. It is obvious that the activity concentrations of ^{40}K have more higher than the activity concentrations of ^{226}Ra and ^{232}Th. The activity concentrations of ^{137}Cs radionuclide in all samples were found to have below the MDA values.

The comparison of activity concentrations with the similar studies (15-18) encountered in literature are presented in Table 2. Except from rosehip, lavender and sage, activity concentrations of ^{226}Ra in this study are clearly seen to be lower than the literature given in Table 2. The activity concentrations of ^{232}Th radionuclide in this work are lower than with the similar studies. Only for linden, an activity concentration of ^{40}K radionuclide in this study was found to be higher than the similar study performed by Solecki et al.(17)

3.2 *Radiation doses of samples*

^{226}Ra, ^{232}Th and ^{40}K are radiologically significant radionuclides occurs naturally in very low concentrations in earth's crust. ^{226}Ra and ^{232}Th can be taken into body by ingestion generally foods or drinks. Both ^{226}Ra and ^{232}Th radionuclides behave chemically as if calcium after they are taken into body. While 80% radium leaves the body in faces, 20% transfers to all organs of body also bones. Likewise, 70% of thorium enters the bloodstream and accumulates in bones. Thorium has a biologic half-life of 22 years and hence, one can easily express that remarkable portion of ^{226}Ra and ^{232}Th radionuclides remains in the bones all person's lifetime (4). Potassium is the essential element for the human body and radioactive ^{40}K comprises 0.012% of natural potassium. The ^{40}K distributes to all organ and soft tissue radiation doses especially intestinal and colon. Therefore, bone, intestinal and colon doses should be taken into consideration. Aforementioned organ doses and effective doses were calculated for adult person by using the following formula (19):

$$E_{ing} = h_{ing}.q.f_d.f_c \qquad (3)$$

Parameters and parameter specific values are given in Table 3 (20&21). Exponantial term of original formula given in reference 21 was neglected because no decay time during or before scenario was assumed due to radionuclides with a long half-life.

Bone surface doses, lower large intestine doses, colon doses and effective doses were calculated for each herb and the results were given in Figure 1, Figure 2, Figure 3 and Figure 4 respectively.

According to the calculation results given in Figure 1, ^{40}K contributes little radiation dose to bone surface in comparison with ^{226}Ra and ^{232}Th. Maximum ^{40}K dose for bone surface were calculated as 6.6 μSvy^{-1} for red pepper. ^{226}Ra and ^{232}Th doses for bone surface were found maximum values as 181.2 μSvy^{-1} and 42.0 μSvy^{-1} for sage and black pepper.

^{226}Ra and ^{232}Th make small contributions to lower large intestine dose in comparison with ^{40}K as it is seen in Figure 2. They are ranged between from zero to 2.3 μSvy^{-1}. Maximum ^{40}K dose were calculated as 18.4 μSvy^{-1} for red pepper. As in the case of lower large intestine, ^{40}K was found as a major dose contributor to colon dose in comparison with the ^{226}Ra and ^{232}Th (Figure 3). Dose contribution of ^{226}Ra and ^{232}Th are varied between zero to 1.5 μSvy^{-1}. Mean value of ^{40}K dose were found as 6.8 μSvy^{-1} for worked medicinal herbs. Maximum ^{40}K dose contribution value was found to be 18.4 μSvy^{-1} for red pepper.

Total bone surface dose was found a maximum value as 182.9 μSvy^{-1} for sage in all medicinal herbs (Figure 1). Maximum lower large intestine and colon doses were calculated as 18.8 μSvy^{-1} and 18.7 μSvy^{-1} for red pepper (Figure 2 and Figure 3). The committed effective dose for interested medicinal herbs were calculated between 0.3 μSvy^{-1} (for rosemary) and 9.0 μSvy^{-1} (for red pepper) as given in Figure 4. The mean committed effective doses of Turkish medicinal herbs were found to be 4.5 μSvy^{-1}. The radiological impact of 4.5 μSvy^{-1} is very small in comparison with the recommendation of International Commission on Radiological Protection (ICRP) for annual effective dose of 1 $mSvy^{-1}$ and the global average annual radiation dose of 2.4 $mSvy^{-1}$ received by individual from all natural radiation sources (22&23).

3.1 *Cancer risks*

According to the ICRP, fatal cancer risk factor is 0.05 Sv^{-1} (24). It means that probability of death originated from cancer increases by 5% for a person who received a total radiation dose of 1 Sv during lifetime (25). The cancer risk for an adult person originated from ingestion of Turkish medicinal herbs was estimated by using the following relationship:

$$Risk = Dose(Sv) \times Risk\ factor(Sv^{-1}) \quad (4)$$

Cancer risks were calculated to be between 7.8×10^{-7} and 2.2×10^{-5} with an average 1.1×10^{-5} for 50 years exposure. The probabilities of increase of cancer risk from ingestion of Turkish medicinal herbs for 50 years vary between 0.00008% and 0.002% in other words. The probability of increase of cancer risk arising from natural radiation sources for 50 years was estimated to be 6.0×10^{-3} (in other words 0.6%) in case exposure from natural radiation sources was regarded as 2.4 $mSvy^{-1}$. It is clearly seen that cancer risk of ingestion of medicinal herbs is small enough to be neglected in comparison with the cancer risk originated from natural radiation sources.

4. Conclusion

In order to assess the additional radiation doses arising from ingestion of the Turkish medicinal herbs, the samples purchased from local markets and traditional sellers. The activity concentrations of natural and artificial radionuclides were determined by gamma spectrometry technique for all samples. The activity concentration of ^{137}Cs radionuclide was found to have below the detection limits for all samples. By using activity concentration results, three organ or tissue, where the most natural radionuclides accumulation is expected, doses were calculated. The bone surface dose, lower large intestine and colon doses were found to be 182.9 μSvy^{-1}, 18.8 μSvy^{-1} and 18.7 μSvy^{-1} respectively. However, in the effective dose calculations, all tissue doses are multiplied by tissue weighting factors. Thus, contributions of selected doses to the effective dose were found to have very small values. The committed effective doses for interested medicinal herbs were calculated between 0.3 μSvy^{-1} and 9.0 μSvy^{-1}. The probabilities of increase of cancer risk from ingestion of Turkish medicinal herbs for 50 years were found to be so small and they can be neglected. In the light of the study results, one can easily express that selected Turkish medicinal herbs are consumed confidently by the adult members of public.

5. Funding

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Captions for the figures

Figure 1. Bone surface doses with contributions of radionuclides

Figure 2. Lower large intestine doses with contributions of radionuclides

Figure 3. Colon doses with contributions of radionuclides

Figure 4. Committed effective doses with contributions of radionuclides

Figure 1

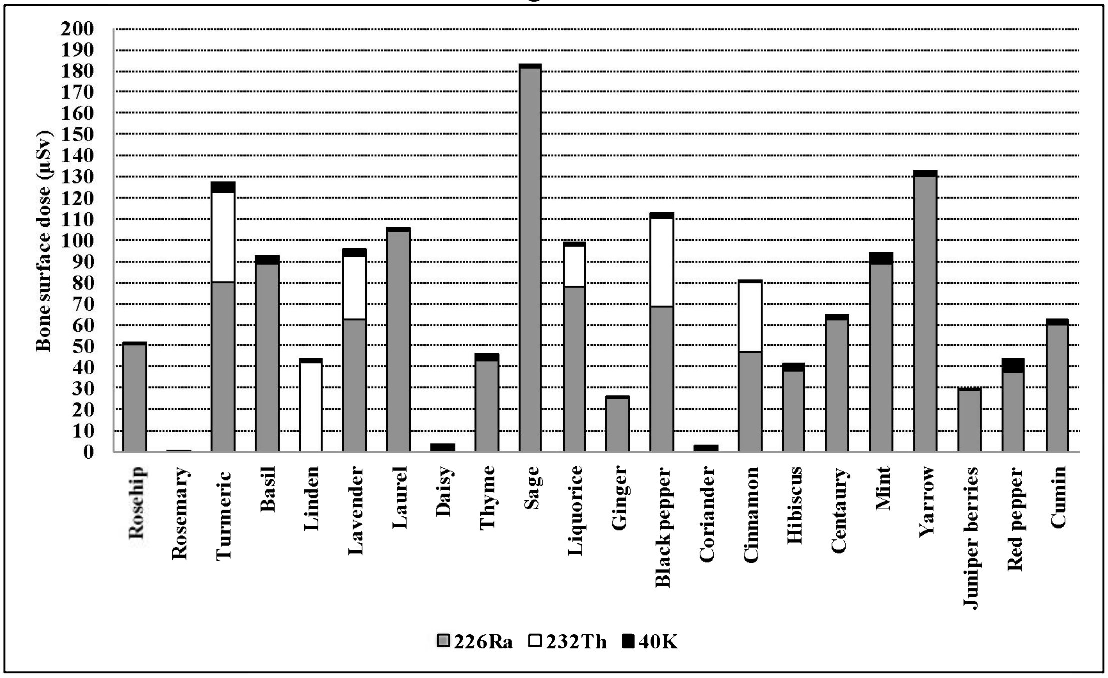


Figure 2

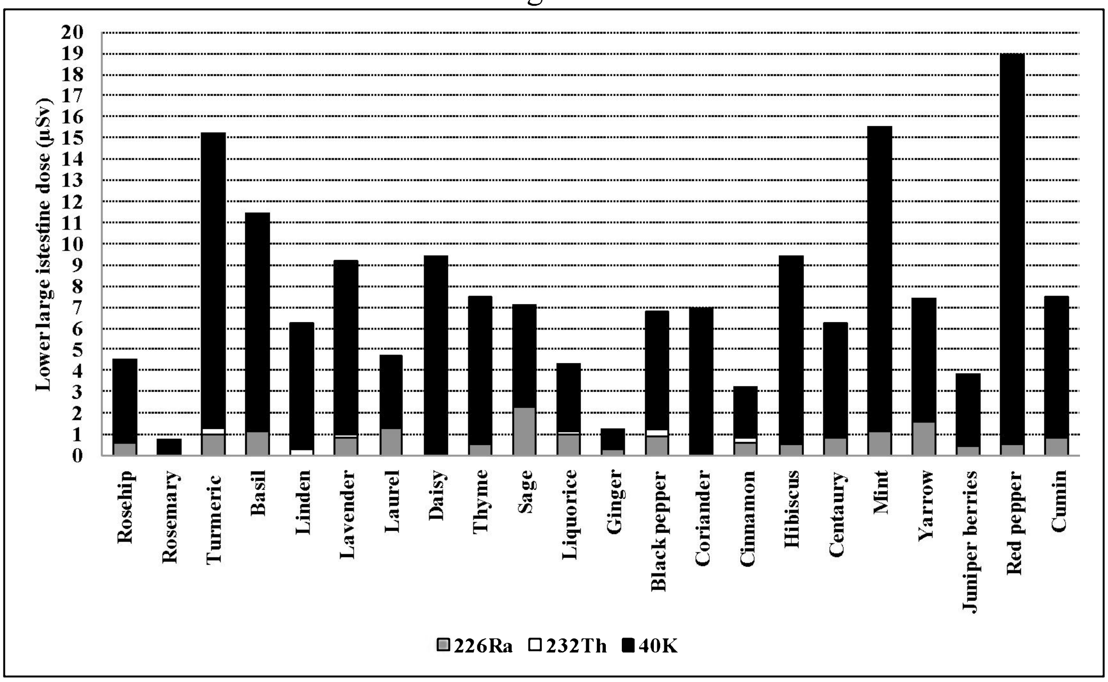


Figure 3.

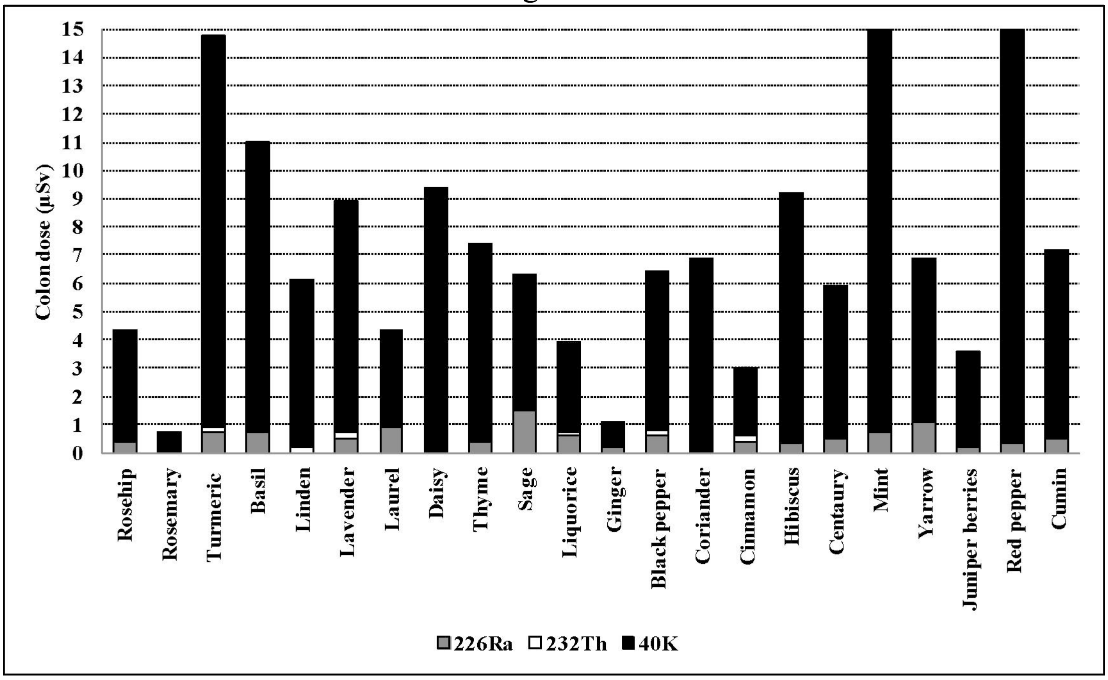
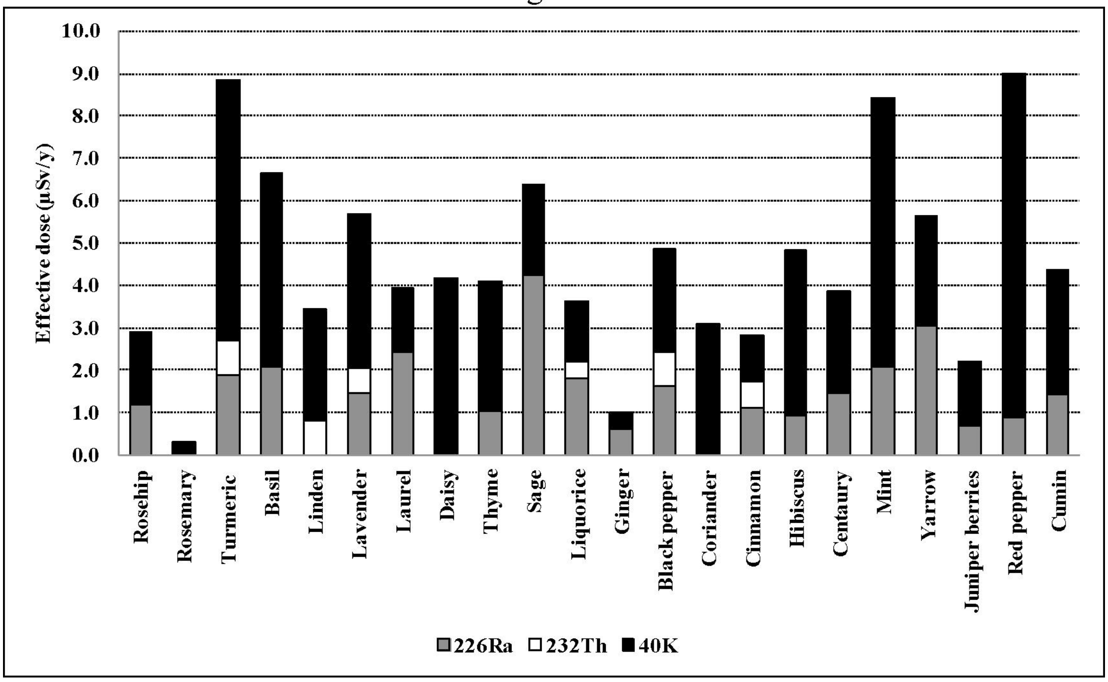


Figure 4



Captions for the table

Table 1. The activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in medicinal herb

Table 2. Comparison activity concentrations with the similar studies encountered in literature

Table 3. Parameters used for dose calculations (20&21)

Table 1

Botanical name	Traditional name	^{226}Ra (Bq kg^{-1})	^{232}Th (Bq kg^{-1})	^{40}K (Bq kg^{-1})	^{137}Cs (Bq kg^{-1})
Fructus cynosbati	Rosehip	4.2±1.2	<0.7	275.3±14.5	<0.1
Rosmarinus officinalis	Rosemary	<2.4	<1.4	50.0±16.8	<0.2
Rhizoma curcumae	Turmeric	6.7±1.2	3.5±1.8	996.3±45.3	<0.1
Ocimum basilicum	Basil	7.4±1.2	<1.4	736.4±38.6	<0.2
Tilia tomentosa	Linden	<3.5	<1.8	422.3±28.5	<0.3
Stoechas lavender	Lavender	5.2±1.3	2.5±0.7	588.0±99.5	<0.5
Laurus nobilis	Laurel	8.7±1.1	<1.0	243.1±16.6	<0.1
Flos chamomillae	Daisy	<0.6	<0.9	672.9± 13.0	<0.2
Thymus vulgaris	Thyme	3.6±1.1	<1.0	496.6±29.0	<0.9
Salvia tchihatcheffii	Sage	15.1±2.2	<1.5	343.1±21.8	<0.3
Osmorhiza longistylis	Liquorice	6.5±1.1	1.6±0.9	230.0±25.3	<1.0
Rhizoma zingiberis	Ginger	2.1±0.7	<0.6	61.8±9.4	<0.1
Piper nigrum	Black pepper	5.7±0.9	3.5±0.8	399.0±41.9	<1.8
Coriandrum sativum	Coriander	<0.5	<0.9	496.0±25.1	<0.1
Cortex cinnamomi	Cinnamon	3.9±0.9	2.8±0.6	172.3±10.5	<0.1
Malva sylvestris	Hibiscus	3.2±1.3	<1.0	634.2±13.4	<1.0
Hypericum perforatum	Centaury	5.2±1.5	<1.0	386.0±6.5	<1.0
Mentha longifolia	Mint	7.4±3.2	<2.2	1026.4±54.1	<0.2
Achillea millefolium	Yarrow	10.9±1.5	<1.3	417.3±23.6	<0.2
Juniperus communis	Juniper berries	2.4±0.9	<0.7	242.1±13.8	<0.1
Capsicum annuum grossum	Red pepper	3.1±0.9	<0.8	1311.5±57.3	<0.2
Fructus cumini	Cumin	5.0±0.9	<0.8	475.6±23.2	<0.1

Table 2.

Medicinal herb	This work				Similar studies				
	^{226}Ra (Bq kg^{-1})	^{232}Th (Bq kg^{-1})	^{40}K (Bq kg^{-1})	^{137}Cs (Bq kg^{-1})	^{226}Ra (Bq kg^{-1})	^{232}Th (Bq kg^{-1})	^{40}K (Bq kg^{-1})	^{137}Cs (Bq kg^{-1})	Ref.
Rosehip	4.2	<0.7	275.3	<0.1	0.6	-	703	<0.3	[15]
Rosemary	<2.4	<1.4	50.0	<0.2	34.2	15.2	881	2.2	[16]
Turmeric	6.7	3.5	996.3	<0.1	13.6	8	1162	0.5	[16]
Basil	7.4	<1.4	736.4	<0.2	-	-	-	-	
Linden	<3.5	<1.8	422.3	<0.3	<1.5	<1.9	186.4	0.8	[17]
Lavender	5.2	2.5	588.0	<0.5	4	18.3	768.5	0.9	[18]
Laurel	8.7	<1.0	243.1	<0.1	-	-	-	-	
Daisy	<0.6	<0.9	672.9	<0.2	1.1	10.9	905.7	0.6	[18]
Thyme	3.6	<1.0	496.6	<0.9	90.9	15.7	1003	2.1	[16]
Sage	15.1	<1.5	343.1	<0.3	1.9	8.2	404.6	1.3	[18]
Liquorice	6.5	1.6	230.0	<1.0	20.8	4.7	421	MDA	[16]
Ginger	2.1	<0.6	61.8	<0.1	17.9	13.7	785	MDA	[16]
Black pepper	5.7	3.5	399.0	<1.8	-	-	-	-	
Coriander	<0.5	<0.9	496.0	<0.1	-	-	-	-	
Cinnamon	3.9	2.8	172.3	<0.1	16.6	-	240	<1.0	[15]
Hibiscus	3.2	<1.0	634.2	<1.0	6.2	13.1	694.9	0.3	[18]
Centaury	5.2	<1.0	386.0	<1.0	-	-	-	-	
Mint	7.4	<2.2	1026.4	<0.2	-	-	-	-	
Yarrow	10.9	<1.3	417.3	<0.2	-	-	-	-	
Juniper berries	2.4	<0.7	242.1	<0.1	4.5	-	342	<0.3	[15]
Red pepper	3.1	<0.8	1311.5	<0.2	-	-	-	-	
Cumin	5.0	<0.8	475.6	<0.1	9.2	2.4	535	MDA	[16]

Table 3.

	Parameter	Unit	Parameter values
h_{ing}	Dose coefficient for ingestion	(µSv Bq^{-1})	0.28 for ^{226}Ra; 0.23 for ^{232}Th; 0.0062 for ^{40}K
	Dose coefficient for bone surface	(µSv Bq^{-1})	12 for ^{226}Ra; 12 for ^{232}Th; 0.005 for ^{40}K
	Dose coefficient for lower large intestine	(µSv Bq^{-1})	0.15 for ^{226}Ra; 0.079 for ^{232}Th; 0.019 for ^{40}K
	Dose coefficient for colon	(µSv Bq^{-1})	0.099 for ^{226}Ra; 0.063 for ^{232}Th; 0.014 for ^{40}K
f_d	Dilution factor	-	1.0
f_c	Concentration factor for the activity	-	1.0
q	Annually ingested quantity	g a^{-1}	1000