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Measurement of natural radioactivity in selected samples of medical plants in Iraq

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In present study, natural levels of radiation in some selected medical plants existing in the Iraqi stores were estimated to determine any activity concentration, radium equivalent and internal hazard index due to radionuclide, of 238U, 232Th as well as 40K, which occurs naturally. This activity concentration was identified by gamma-ray spectroscopy (NaI(Tl)). The findings indicate that, the rate of activity concentration for Uranum-238, Thorium-232 and Potasium-40 in the medical plants were (4.953$\pm$0.37) Bq.kg^{-1}, (2.916$\pm$0.12) Bq.kg^{-1} and (219.134$\pm$2.24) Bq.kg^{-1} respectively. The values of the radium equivalent ranged from (6.081$\pm$0.09) Bq.kg^{-1} to (44.608$\pm$0.46) Bq.kg^{-1} with the rate of (20.278$\pm$0.38) Bq.kg^{-1}, while the values of the internal hazard index ranged from (0.016) to (0.135) with the rate of (0.060). The natural radionuclides and activity of the radium equivalent in the medical plant samples were far below the world for the ingestion of naturally occurring radionuclide provided in UNSCEAR 2000 report. Also there was calculated the internal hazard index for all samples that were less than unity. The samples under study were analysed and discharged, therefore they can no be consumed anymore.

Key words: Natural radioactivity, medical plants, gamma-ray spectroscopy, radium equivalent and Iraqi markets.

INTRODUCTION

Natural radioactive decay like ^{238}U and ^{232}Th series in addition to radionuclide that occurs singly such as ^{40}K is found in the atmosphere and the earth in varied levels. The radioactivity in the agricultural land and in soil may transfer to the plants around. The radionuclide available in the environment is transferred to plants by two ways, first of which is the indirect method uptake from soil through roots. When food plants are developed in a polluted soil, the activity is switched to the roots from the soil and then in shoots. At the end, it transfers to the human diet Ramiza et al. (2010). This radionuclide can get into the plants during mineral uptake along with the nutrients and accrue in different areas and even it could reach the edible parts Brianna (2011). First, the direct method absorption; it occurs through aerial plant areas. A variety of workers have reviewed the presence of radioactivity in the plant organs Pooja and Rishi (2014). Herbs used as medicines for living beings may be called

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medical plant. Some of these plants are used as raw materials for the manufacture of drugs Kalač (2001). The level of natural radioactivity in some of these herbs is interesting taken from different viewpoints as, for instance, from the environmental point of view Kalač (2001). Viewed differently, medical plants are excluded from the edible plant group under study possibly due to the fact that the absorption of radioactive material through consumption has not been recognized or was considered insignificant (Ele Abiama et al., 2012). A medical plant with high levels of natural radioactivity can lead to health problems because they are usually consumed for long time.

There are many studies about the concentration of radioactivity in medical plants from different countries Desideri et a. (2010), Oni et al. (2011), Tettey et al. (2013) and Fahad et al. (2014). There was clear data base on natural radioactivity levels in medical plant samples despite the wide intake of these kinds of plants as medical treatment. The aim of the present study is to measure the naturally occurring levels of radioactivity (^{238}U, ^{232}Th and ^{40}K) in some selected herbs usually used in Iraq, and calculate the radiological hazard of the use of these herbs such as radium equivalent activity and internal hazard indices.

MATERIALS AND METHODS

Preparation and collection of samples

In September 2015, forty different samples of medical plants were collected from the local markets in various places in Najaf city as shown in Table 1. It is consist of the cursor in front of each sample represents the sample code, trade name, scientific name, Part used and country of origin.

After collecting the samples, each one was put in a plastic bag and given a label to show its name. The samples have been dried, ground homogenization and sieved. The samples are dried before radioactivity measurement for (2-4) days at a temperature of (42-44)°C to avoid any humidity adsorption, and to maintain the actual weight. The dried samples under study were have been ground and milled using a blender to obtain an equal size particles. Later, equal weight (0.75) kg of each sample (using a high sensitive digital weighting balance with a percent of ±0.01%). Then the samples have been kept in the containers. These samples were put into a 1 liter polyethylene plastic Marinelli beakers of a fixed volume to reach a geometric homogeneity all around the detector, then the net weights were measured and recorded with a highly sensitive digital weighing balance at ±0.01%.

Laboratory procedure

Measurements are carried out by adopting systems of gamma spectrometry from ORTEC, equipped with a high efficiency scintillation detector, an NaI(Tl) detector of (3"×3") crystal dimension, with resolution 9.2% for ^{137}Cs (661.7 keV). A lead shield of ten cm thickness was put around the detector to lessen the background, with a 0.3 cm layer of copper to weaken x-rays emitted by the lead shield. The spectra are analyzed off-line using the ORTEC Maestro-32 data acquisition and analysis system. The activity concentration is expressed in (Bq.kg^{-1}) dry weight depending on the sample type. The detector is energy calibrated using the standard source of known energies like ^{22}Na, ^{60}Co and ^{137}Cs . The specific activity of ^{40}K was directly identified from the peak areas at 1460 keV. The activity concentrations of ^{238}U and ^{232}Th were measured presuming secular equilibrium with their decay products. To measure the activity concentration of radioisotope in the ^{238}U-series, gamma transition lines of ^{214}Bi (1765 keV) were employed. Also, radioisotope activity concentrations in the ^{232}Th-series were identified by applying gamma transition lines of ^{208}Tl (2614 keV). The average counting time is 24 hour for each sample, to ensure a good statistical significance.

Calculations

Activity concentration (A_c) can be determined as follows (Al-Hamidawi, 2014):

$$A_c = \frac{C - BG}{\varepsilon\% M.t.I_\gamma} \ldots\ldots \quad (1)$$

where Ac means the activity concentration, C refers to the spot under the photo peaks, BG is background, ε% is energy efficiency percentage, M means mass of sample, t is counting time and Iγ means the percentage of gamma-emission probability for the radionuclide under consideration.

Radium equivalent activity (Ra_{eq}) is utilized to evaluate the risks of materials that contain ^{238}U, ^{232}Th and ^{40}K in Bq.kg^{-1} (Nasim et al., 2012), which is identified by presuming that 370 Bq.kg^{-1} of ^{226}Ra or 260 Bq.kg^{-1} of ^{232}Th or 4810 Bq.kg^{-1} of ^{40}K produce the same gamma dose rate.

The Ra_{eq} of a sample in (Bq.kg^{-1}) can be achieved using the following relation (Ali Abid et al., 2014; Yu et al. 1992; El-Arabi, 2007; Quindos et al., 1987):

$$Ra_{eq.} = A_U + (1.43\, A_{Th}) + (0.077\, A_K) \ldots\ldots \quad (2)$$

The internal hazard index can be quantified by the internal hazard index (H_{in}). This is given by the following equation (Ali Abid et al., 2014; Yu et al., 1992; El-Arabi, 2007; Quindos et al., 1987):

$$H_{in} = \frac{A_U}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \ldots\ldots\ldots\ldots\ldots \quad (3)$$

The internal hazard index has to be less than one as well to provide safe radionuclide levels in medical plants.

RESULTS AND DISCUSSION

The activity concentration due to ^{238}U, ^{232}Th, and ^{40}K in different kinds of medical plant samples has been measured (Table 2). The ^{238}U activity concentration was turned to be within the scope of (<BDL) Bq.kg^{-1}to (12.59±0.43) Bq.kg^{-1}with an average of (4.68±0.26) Bq.kg^{-1}, ^{232}Th from (<BDL) Bq.kg^{-1}to (14.63±0.24) Bq.kg^{-1}with an average of (2.91±0.12) Bq.kg^{-1} and ^{40}K from (78.56±1.15) Bq.kg^{-1}to (579.32±5.21) Bq.kg^{-1}within an average of (219.13±2.24 Bq.kg^{-1}. The radium equivalent and internal hazard indices were calculated for the whole samples in this research as in Table 3. The Ra_{eq} and (H_{in}) varied from (0.016) to (0.135) with (0.060) average and from (6.08±0.09) Bq.kg^{-1} to (44.61±1.08) Bq.kg^{-1} at an average of (20.27±0.38) Bq.kg^{-1} respectively. When

Table 1. Medical plant samples.

S/N	Sample code	Traditional name	Scientific name	Part used	Country of origin
1	H1	Senna	*Cassai senna* L.	Leaves	Saudi Arabia
2	H2	Safflower	*Carthamus tinctorius*	Flowers	Iran
3	H3	Ziziphus	*Ziziphus spina-Christi* L.	Leaves	Iraq
4	H4	Hops	*Humulus Lubulus* L.	Peduncle	Iran
5	H5	Pepper mint	*Mentha piperita* L.	Leaves	Iraq
6	H6	Balanitea	*Balanites aegyptiaca* (L.) Del.	Fruits	Egypt
7	H7	Aelchenan	*Anabasis*	Leaves	Iraq
8	H8	Green tea	*Camellia sinensis*	Leaves	China
9	H9	Fenugreek	*Trigonella foenum-graecum* L.	Seeds	India
10	H10	Sweet marjoram	*Origanum majvrana*	Aerial parts	Middle east
11	H11	Ginger	*Zingiber officinale* Roscoe.	Roots	India
12	H12	Grea plantain	*Plantago major* L.	Peel fruits and seeds	India
13	H13	Hawthorn	*Crataegus spp.*	Leaves	USA
14	H14	Mahleb	*Prunus vinginiana*	Seeds	Azerbaijan
15	H15	Myrtle	*Myrtus Communis* L.	Leaves	Iraq
16	H16	Barberry	*Thuja occidentalis* L.	Fruits	Syria
17	H17	Rosemary	*Rosmarinus officinalis* L.	Aerial parts	Mediterranean sea
18	H18	Chicory	*Cichorium intybus* L.	Roots,stalk and leaves	Iraq
19	H19	Chamomile	*Matricaria chamomilla* L.	Flowers	Syria
20	H20	Sage	*Salvia Officinalis*	Leaves	India
21	H21	Maidenhair fern	*Abiantum capillus-Veneris* L.	Leaves and stalk	USA
22	H22	Leaf mustard	*Brasica nigra* (L.) Koch	Seeds	China
23	H23	Cyperus	*Cyperus* esculentus	Seeds	Egypt
24	H24	Rose-Mallow	*Althaea rosea* L.	Flowers	India
25	H25	Blinko	*Ocimumba silicum*	Seeds	Iran
26	H26	Bay leaves	*Laurus nobilis*	Leaves	Syria
27	H27	Corn Mint	*Mentha hapolcaltt*	Aerial parts	India
28	H28	Black cumin	*Nigella sativa* L.	Seeds	India
29	H29	Roselle	*Hibiscuc sabddariffa* L.	Flowers	Iraq
30	H30	Horse tail	*Equisetium arvense* L.	Aerial parts	Egypt
31	H31	African rue	*Ruta chalepensis* L.	Aerial parts	Saudi Arabia
32	H32	Flax	*Linum Usitatissimum* L.	Seeds	Iran
33	H33	Stout bien	*Angelica archangelica* L.	Each herb	China
34	H34	Yarrow	*Achillea nillefolium* (Forssk)Sh-Bip	Aerial parts	Iran
35	H35	Nutgrass	*Cyperus rotundus L.*	Roots and leaves	Saudi Arabia
36	H36	Colocynth	*Citrullus colocynthis* (L.) *Shradc*	Fruits	Iraq
37	H37	Primrose	*Primula vulgris* L.	Flowers	west Asia
38	H38	Alkanet	*Borago officinalis*	Flowers	Iran
39	H39	Coltsfoot	*Tassilago Farfar*	Leaves and flowers	North Asia
40	H40	Rose of jericho	*Anastatica Hierochuntica* L.	Branches	Palestine

compared to the obtained results with the world wide average recommended by UNSCEAR (2000), we have found that ^{238}U and ^{232}Th are lower but higher in the case of ^{40}K in some samples such as H18, H30, H36 and H38 UNSCEAR (2000). The highest allowable activity concentration in some samples because increase in the concentration of potassium nuclide in some areas of the reason is due to the existence of agricultural land and areas containing phosphate fertilizers in which the focus increasingly peer-potassium (^{40}K). The values for the radium equivalent activity (Ra_{eq}) are found to be within the world average allowed maximum value of 370 $Bq.kg^{-1}$ UNSCEAR (2000). All values of internal hazard indices are lower than the international permissible value of unity UNSCEAR (2000). However, comparing these results with earlier studies (Table 4) indicates that the levels of

Table 2. Activity concentration of ^{238}U, ^{232}Th and ^{40}K in medical plant samples.

Sample code	Activity concentration in ($Bq.kg^{-1}$)		
	^{238}U	^{232}Th	^{40}K
H1	3.58±0.25	BLD	139.90±1.63
H2	BLD	BLD	274.51±2.925
H3	4.09±0.26	BLD	261.73±2.19
H4	2.96±0.23	BLD	108.79±1.49
H5	2.61±0.23	BLD	234.57±2.33
H6	1.98±0.13	BLD	133.96±1.18
H7	4.42±0.23	BLD	289.03±1.94
H8	7.11±0.34	1.86±0.10	167.49±1.72
H9	2.45±0.17	BLD	144.60±1.40
H10	BLD	BLD	186.02±2.02
H11	3.13±0.23	3.17±0.14	213.77±2.00
H12	12.59±0.43	2.24±0.12	187.05±1.75
H13	BLD	BLD	119.03±1.84
H14	BLD	BLD	84.10±1.28
H15	1.08±0.14	BLD	154.74±1.80
H16	BLD	BLD	87.46±1.08
H17	BLD	BLD	116.19±1.79
H18	BLD	BLD	579.32±5.21
H19	BLD	BLD	338.94±3.57
H20	BLD	BLD	208.82±3.03
H21	11.47±0.62	2.10±0.16	237.860±2.95
H22	1.17±0.14	BLD	106.05±1.40
H23	BLD	1.43±0.09	78.56±1.15
H24	BLD	1.29±0.10	308.44±2.78
H25	BLD	1.46±0.08	136.353±1.43
H26	BLD	BLD	135.561±1.61
H27	BLD	1.35±0.10	297.128±2.74
H28	BLD	BLD	130.36±1.53
H29	2.38±0.16	3.02±0.11	283.65±1.91
H30	BLD	1.62±0.13	449.231±3.96
H31	BLD	BLD	108.94±1.39
H32	BLD	BLD	78.98±1.17
H33	BLD	1.49±0.11	370.10±3.18
H34	BLD	BLD	296.57±3.21
H35	8.89±0.31	14.63±0.24	150.24±1.34
H36	BLD	BLD	440.43±3.12
H37	BLD	2.739±0.23	211.11±3.48
H38	BLD	BLD	409.26±4.02
H39	4.95±0.37	2.39±0.15	185.68±2.40
H40	2.75±0.22	BLD	320.99±4.09
Average	4.68±0.26	2.91±0.12	219.13±2.24

natural radioactivity in this study are moderate, where Table 4 shows a comparison between the average value of the current work and the average values for medical plants sample in some countries.

Table 3. Radium equivalent internal hazard index in medical samples in this study.

Sample code	Ra_{eq} ($Bq.kg^{-1}$)	H_{in}
H1	14.35±0.37	0.048
H2	21.13±0.31	0.057
H3	24.25±0.43	0.076
H4	11.39±0.43	0.038
H5	20.67±0.49	0.062
H6	12.30±0.22	0.038
H7	26.68±0.47	0.084
H8	22.66±0.62	0.080
H9	13.68±0.28	0.043
H10	14.32±0.36	0.038
H11	24.12±0.59	0.073
H12	30.20±0.73	0.115
H13	9.16±0.25	0.024
H14	6.47±0.09	0.017
H15	13.00±0.28	0.038
H16	6.73±0.08	0.018
H17	8.94±0.13	0.024
H18	44.60±0.46	0.120
H19	26.09±0.27	0.070
H20	16.07±0.23	0.043
H21	32.79±1.08	0.119
H22	9.34±0.25	0.028
H23	8.08±0.21	0.021
H24	25.60±0.36	0.069
H25	12.59±0.23	0.033
H26	10.43±0.19	0.028
H27	24.81±0.36	0.067
H28	10.03±0.22	0.027
H29	28.54±0.47	0.083
H30	36.91±0.50	0.099
H31	8.38±0.15	0.022
H32	6.08±0.09	0.016
H33	30.62±0.41	0.082
H34	22.83±0.38	0.061
H35	41.42±0.76	0.135
H36	33.91±0.34	0.091
H37	20.13±0.59	0.054
H38	31.51±0.44	0.085
H39	22.66±0.78	0.074
H40	27.45±0.51	0.081
Average	20.27±0.38	0.060

Conclusions

The values of activity concentration of ^{238}U, ^{232}Th and ^{40}K in samples of medical plants are found to be lower than the world average allowed maximum values 32, 30 and 400 $Bq.kg^{-1}$ respectively, except the activity concentration

Table 4. Comparison of the activity concentrations in the medical plants.

Country	Activity concentration (Bq.kg^{-1})			Reference
	^{238}U	^{232}Th	^{40}K	
Iraq	4.68	2.91	219.1	This work
Italy	0.4	-	654.7	Desideri et al. (2010)
Brazil	-	21.7	976.3	Scheibel and Appoloni (2007)
Nigeria	-	35.1	171.7	Njinga et al. (2015)
Ghana	31.8	56.2	839.8	Tettey-Labri et al. (2013)
Serbia	2.6	7.4	589.6	Milutin et al. (2011)

of 40K that found to be higher in samples H18, H30, H36 and H38. This can be explained by the soil ails that come as a result of an abundance of this isotope concentration. The values for the radium equivalent activity (Ra_{eq}) are turned to be within the international average allowed maximum value of 370 Bq.kg^{-1}. This study could be of help as a data base for radionuclide concentration and radium equivalent activity. The value of hazard internal is lower than the international permissible value of unity. In general terms, it can be concluded that the implemented technique show good results when matched with other literature data. Also it can be concluded that samples under study, which have been analyzed, are safe for human consumption because their radioactivity levels are less than the maximum permitted level.

Conflict of Interests

The authors have not declared any conflict of interest.

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