DOI Link: https://doi.org/10.61586/zyF3K
Vol.33, Issue.11, Part.1, Nov 2025, PP.2-14

Evaluation of radionuclide concentrations and average annual committed effective dose due to medicinal plants collected from Al-Qassim province, Saudi Arabia

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Received Feb 2025 Accepted Oct 2025 Published Nov 2025

Abstract:
Medicinal plants have historically served as some of the earliest therapeutic agents for managing a wide range of diseases and health conditions. Given their widespread use and potential for direct human consumption, it is essential to evaluate the concentrations of radionuclides in these plants to ensure both human safety and environmental protection. In the present study, the levels of natural radioactivity were assessed in 21 medicinal plant samples collected from local markets in the Al-Qassim region. The investigation focused on quantifying the activity concentrations and distribution patterns of naturally occurring radioactive isotopes within the samples. Gamma-ray spectrometry using a NaI (Tl) detector was employed to measure the activity concentrations of key radionuclides. The activity concentrations of 226Ra ranged from 19.3±0.2 to 54.2±0.5 $Bq.Kg^{-1}$ with average 29.4±0.3 $Bq.Kg^{-1}$. The activity of 232Th ranged from 14.8±0.2 to 38.8±0.3 $Bq.Kg^{-1}$ with average 23.5±0.2 $Bq.Kg^{-1}$. For 40K ranged from 197.5±2.7 to 474.8±3.9 $Bq.Kg^{-1}$ with average 309.5±3.3 $Bq.Kg^{-1}$. In addition to activity concentrations, several radiological hazard indices were calculated, including the absorbed dose rate, gamma representative level index (Iγ), radium equivalent activity (Raeq), annual effective dose equivalent (AEDE), and external and internal hazard indices (H_{ex} and H_{in}). The results demonstrate that the natural radioactivity levels detected in the analyzed medicinal plant species are within internationally recommended safety thresholds based on radiation risk assessments. To the best of our knowledge, this study provides the first comprehensive evaluation of natural radioactivity in medicinal plants originating from the Al-Qassim region. The data generated offer a valuable baseline for developing safety standards and regulatory guidelines concerning the use of these plants. Such measures are crucial to safeguard public health and to ensure the safe incorporation of medicinal plants in traditional therapeutic practices and related applications.
Keywords: Natural radioactivity levels; Radiation hazards; Absorbed dose rate; Gamma ray spectrometry

1. Introduction:

Medicinal plants represent one of the earliest therapeutic resources employed by humans for the management of various diseases and health conditions. Their use dates back to ancient civilizations, where herbs and plant-based preparations served as primary remedies long before the development of modern pharmaceuticals. Even today, medicinal plants continue to play a vital role in traditional healing systems, including Traditional Chinese Medicine, Ayurveda, and traditional Arabic medicine. In recent years, interest in medicinal plants has grown significantly, as they are increasingly sought after as natural alternatives or complementary options to conventional therapies, owing to their perceived therapeutic benefits and relatively fewer side effects compared to synthetic drugs. [1], [2]. Each and every element of the environment, including the air, water, soil, food, and people, contains naturally occurring radioactive materials (NORMS). The International Food Safety Authorities Network states that ^{40}K, ^{232}Th, and ^{238}U and their offspring are frequently found in plants used as food. Since plants are the main source of naturally occurring radionuclides that enter the human body through the food chain, it is expected that the same would be present in plants utilized for medical purposes. Natural radioactivity in the environment is caused by terrestrial primordial radio nuclides, which are derived from the earth's crust and include radionuclides and their decay products from the ^{238}U and ^{232}Th family along with ^{40}K [3], [4]. Different parts of medicinal plants—such as aerial components, roots, leaves, fruits, flowers, seeds, and their extracts—are widely employed in the development of pharmaceutical products for therapeutic use. It is estimated that approximately 25% of modern drugs are derived, either directly or indirectly, from traditional medicinal plants. Globally, nearly 80% of the population, particularly in developing countries, relies on herbal remedies as a primary source of healthcare. Research on medicinal plants has expanded considerably, generating extensive data that highlights their vast potential across diverse traditional medical systems. However, the therapeutic properties and effectiveness of these plants can be significantly influenced by environmental conditions, climatic variations, and geographical origin. They may also directly affect how well they thrive in the areas where they are found and grown [3], [5]. Due to the growing emphasis on primary health care or basic health care advocacy, a WHO poll shows that between 70-80% of people worldwide rely on traditional medicine, primarily derived from plant sources [6]. Plants have the ability to directly and indirectly introduce radioactivity into the human nutrient cycle and ecology through vegetable and animal food items. Common food plants have ^{40}K, ^{232}Th, and ^{238}U [7].Therefore, since plants are the main source of natural radionuclides that enter the human body through the food chain, these parallels would be present in plants utilized for treatment. Consequently, it is necessary to keep an eye on the degree of contamination in the medicinal plants and analyzed. Additionally, every human being has some level of exposure to natural radionuclides such ^{238}U, ^{232}Th, and ^{40}K to radiation [5].The aim of this study is to determine the natural radioactivity present in some selected medicinal plants commonly used in Al-Qassim region of Saudi Arabia and to assess the radiological risks associated with the use of these medicinal plants by measuring the radioactivity, activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K, the radium equivalent

activity (Ra_{eq}), hazard index, representative level index, Absorbed Dose, the annual effective dose equivalent (AEDE).

2. Materials and methods

2.1 Location of Study Area

This study was conducted in the Qassim Province, located in the central region of Saudi Arabia, approximately 400 km northwest of Riyadh, the capital city. Qassim is bordered by Riyadh Province to the south and east, Ha'il Province to the north, and Al-Madinah Province to the west. Geographically, the province is characterized by extensive sand dunes, particularly in the western and southern areas, and covers an area of about 73,000 km², representing approximately 3.2% of Saudi Arabia's total landmass (Figure 1). The region has a desert continental climate, with hot, arid summers and cold, rainy winters. Average annual rainfall is about 145 mm, while temperatures range between 45 °C in summer and −3 °C in winter. Although storms may occur throughout the year, they are most frequent during the spring season. [8]. It is rich in natural resources and agriculture and produces over 1.22 million tons of agricultural products produced per year.

Figure 1. Location of Al-Qassim region in Saudi Arabia.

2.2 Instrumentation and calibration

A gamma ray scintillation spectrometry NaI(Tl) detector model A320 and SN A3200829 was used to determine activity concentrations of radionuclides. The hermetically sealed assembly is coupled to a personal computer-multichannel analyzer (Canberra AccuSpec) model MCA2500R and serial 25,066. The detector was shielded to reduce background radiation using lead shield (100 mm thick) and copper shield (0.3 mm thick). Quantum Gold version 4.04.4 PGT (Princeton Gamma- Tech) was used to analyze gamma ray spectrum. An empty beaker was used in the same condition of samples measuring to estimate the background radiation around the work environment. The accumulated spectrum of background was subtracted from specified photo-peak energy of each sample to get accurate measured activity.

The naturally occurring radionuclides considered in the present analysis of the measured γ-ray spectra are : ^{212}Pb (with a main gamma energy at ~239 keV and a gamma yield of ~ 43.1%),^{214}Pb (~ 352 keV, ~ 37.1%),^{214}Bi (~ 609, 1120 and 1765 keV, ~ 46.1, 15 and 15.9% respectively), ^{228}Ac (~911 keV, ~ 29%), ^{208}Tl (~2615 keV , ~ 35.9%) and ^{40}K (~1461 keV, ~10.7%). Under the assumption that secular equilibrium was reached between ^{232}Th and ^{226}Ra and their decay products, the concentration of ^{232}Th was determined from the average concentrations of ^{212}Pb, ^{208}Tl and ^{228}Ac in the samples, and that of ^{238}U was determined from the average concentrations of the ^{214}Pb and ^{214}Bi decay products. Gamma-spectrometric measurements was performed with sodium iodide detector. The detector had a photopeak efficiency of about30% and an energy resolution of 1.85 keV for the 1.33MeV γ -transition of ^{60}Co. The γ-ray spectrometer energy was calibrated using ^{60}Co and ^{137}Cs point sources.

2.3 Plant collection and identification

To assess the natural radioactivity levels, medicinal plant samples were collected from the Al-Qassim region of the Kingdom of Saudi Arabia (Table 1). A total of 21 samples were analyzed, with each plant species represented by an average of three measurements. The samples were first ground and sieved to achieve uniform particle size, after which they were transferred into 200 mL polyethylene containers. The containers were tightly sealed and stored for a period exceeding one month to allow secular equilibrium to be established between radium and its progeny, thereby ensuring the complete retention of radon prior to analysis. Using an NaI (Tl) detector, nuclear spectrometry measurements were conducted. The background distribution resulting from the natural radioactive isotopes present in the vicinity of the detector was determined by counting an empty polystyrene container in the same way as the samples were counted. The activity concentration was determined by measurement and background subtraction. The measuring time for gamma-ray spectra 24 hours. The formula below was used to determine the concentrations of radioactivity for both natural radioactive isotopes in the analyzed samples [9], [10], [11]

$$\boldsymbol{A}\left(\frac{\boldsymbol{Bq}}{\boldsymbol{Kg}}\right) = \frac{\boldsymbol{N_P \times 100}}{\boldsymbol{e \times \eta \times m}} \qquad (1)$$

Where: Np: is the net count rate (cps), e: is the abundance of the γ-peak in a radionuclide, m: is the mass of sample(kg), η: is the measured efficiency for each gamma-ray peak observed for the same number of channels.

Table (1): The average activity concentration of Ra-226 and Th-232 series and K-40 in ($Bq.Kg^{-1}$) of the studied samples

Sample Name	Sample number	Activity concentration ($Bq.kg^{-1}$)		
		Ra-226	Th-232	K-40
Fenugreek	1	31.4±0.4	26.3±0.3	326.9±3.6
Calligonum Comosum	2	36.0±0.3	29.9±0.3	323.7±3.0
Caraway	3	35.3±0.4	29.6±0.3	350.8±3.6
Moringa Oleifera	4	54.2±0.5	38.8±0.3	474.8±3.9
Foeniculum Vulgare	5	34.3±0.4	28.1±0.3	357.3±3.6
Eruca Vesicaria Ssp Sativa	6	22.8±0.3	18.5±0.3	221.4±3.1
Alum	7	22.1±0.3	18.2±0.3	197.5±2.7
Vitex Agnus-Castus	8	44.1±0.3	35.7±0.3	473.1±3.4
Pomegranate Peel	9	32.4±0.4	26.8±0.3	348.4±3.8
Anethum Graveolens	10	35.7±0.2	29.4±0.3	390.1±3.8
Celery	11	34.3±0.3	28.2±0.3	358.3±3.5
Garden Cress Seeds	12	25.4±0.4	20.9±0.3	275.7±3.8
Commiphora Myrrha	13	21.0±0.3	17.0±0.3	214.4±3.3
Ajwain	14	29.5±0.3	23.5±0.3	355.3±3.7
Prunus Mahaleb	15	23.3±0.3	17.5±0.2	268.3±2.9
Thyme	16	22.8±0.3	17.9±0.2	246.7±3.1
Saussurea Costus	17	26.7±0.3	21.4±0.2	317.4±3.4
Common Fig	18	21.1±0.3	16.3±0.2	255.8±3.5
Mung Bean	19	19.3±0.3	14.8±0.2	233.4±3.2
Alkanna Tinctoria	20	22.5±0.3	17.8±0.2	230.9±2.5
Pimpinella Anisum	21	23.3±0.3	17.3±v	280.2±3.0
	Maximum	54.2±0.5	38.8±0.3	474.8±3.9
	Minimum	19.3±0.2	14.8±0.2	197.5±2.7
	Average	29.4±0.3	23.5±0.2	309.5±3.3

3. Results and discussion

3.1 Activity Concentrations of ^{226}Ra, ^{232}Th and ^{40}K

The concentrations of radioactivity for the isotopes ^{226}Ra, ^{232}Th, and ^{40}K in medicinal plant samples were determined, and their concentrations were measured using gamma spectroscopy. The activity concentrations of ^{226}Ra ranged from 19.3±0.2 to 54.2±0.5 Bq.Kg^{-1} with average 29.4±0.3 Bq.Kg^{-1}. The activity of ^{232}Th ranged from 14.8±0.2 to 38.8±0.3 Bq.Kg^{-1} with average 23.5±0.2 Bq.Kg^{-1}. For ^{40}K ranged from 197.5±2.7 to 474.8±3.9 Bq.Kg^{-1} with average 309.5±3.3 Bq.Kg^{-1} as shown in table 1 and figure 2.

The activity concentrations must be less than 33, 45, and 400 Bq.Kg^{-1} for ^{226}Ra, ^{232}Th, and ^{40}K, respectively [12]. The activity concentrations of medicinal plants in Al-Qassim region were below the recommended level. Table 2 shows a comparison of the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in medicinal plant samples in this study and other studies around the world. Where the ^{226}Ra score was higher than that of Iraq (1), South India, Jordan, Turkey, Nigeria (1), Bangladesh, Egypt and Serbia but it was lower than Iraq (2) and Ghana. In case ^{232}Th, the result was higher than Iraq (1), Iraq (2), South India, Jordan, Bangladesh, Turkey, Nigeria (1), Egypt and Serbia but it was lower than Ghana and Nigeria (2). Regarding to ^{40}K, our result was lower than the most countries except Iraq (1) and Turkey.

Table 2. The mean activity concentrations (Bq.Kg^{-1}) of the natural radioactivity of medicinal plant samples in the present study were compared with those from similar investigations performed in other countries.

Country	Ra-226	Th-232	K-40	Reference
Iraq (1)	4.953 ± 0.37	2.916 ± 0.12	219.134 ± 2.24	[23]
South India	6.34 ± 0.81	5.05 ± 0.7	1895.24 ± 103.95	[24]
Iraq (2)	38.12 ± 1.619	12.95 ± 0.896	570.70 ± 31.453	[20]
Ghana	31.8 ± 2.8	56.2 ± 2.3	839.8 ± 11.9	[25]
Bangladesh	12.65 ± 5.20	7.38 ± 3.45	661.1 ± 202.6	[26]
Jordan	2.63 ± 0.30	1.44 ± 0.18	593.97 ± 63.47	[27]
Turkey	4.48	1.83	259.2	[28]
Nigeria (1)	5.79 ± 1.51	4.13 ± 0.55	630.03 ± 52.9	[29]
Nigeria (2)	25.02 ± 3.18	35.09 ± 0.71	324.18 ± 8.69	[30]
Serbia	2.82	0.63	984.32	[11]
Egypt	7.25 ± 0.54	7.78 ± 0.633	471.4 ± 11.33	[10]
World	33	45	400	[16]
Present study	29.4±0.3	23.5±0.2	309.5±3.3	

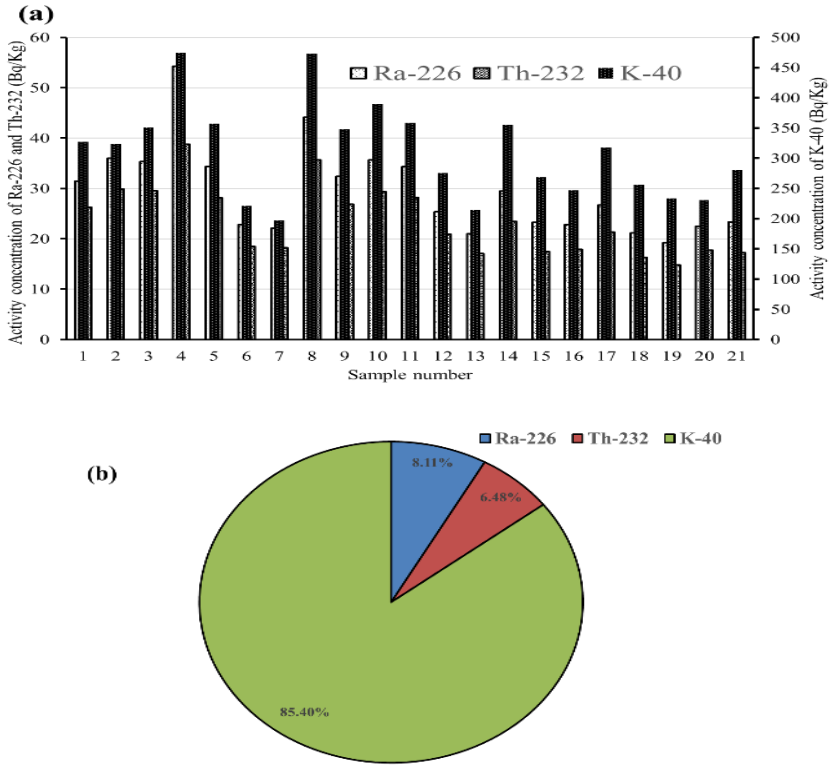

Figure 2. (a)The average activity concentration of Ra-226, Th-232 and K-40 and (b) the percentage contribution of Ra-226, Th-232 and K-40 in the selected samples.

3.2. Radiological Parameters

3.2.1. Radium Equivalent Activities (Ra_{eq})

The radiation hazard index that is most frequently used is the radium equivalent activity (Ra_{eq}). The source of Ra_{eq} is

$$Ra_{eq}\left(\frac{Bq}{Kg}\right) = A_{Ra} + 1.43 \times A_{Th} + 0.077 \times A_{K} \quad (2)$$

The average value calculated for Raeq was 84.7 Bq.Kg^{-1} as shown in Table 3. This value was below the 370 Bq kg^{-1} permissible maximum value [13], [14], [15].

3.2.2. Absorbed Dose rate

The absorbed dose rate for the uniform distribution of radioactive isotopes in the air at a height of one meter above the ground surface has been calculated in accordance with the criteria [15]. The following equation represents it.

$$D(nGy/h) = 0.427\ A_{Ra} + 0.662\ A_{Th} + 0.043 A_{K} \quad (3)$$

where A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively. The average calculated absorbed dose rate is 41.4 nGy/h. As shown in Table 3, this information was calculated from the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K, which are below the recommended limits [16]. Figure 3 shows the specified absorbed dose rate and the radium equivalent activity of the samples.

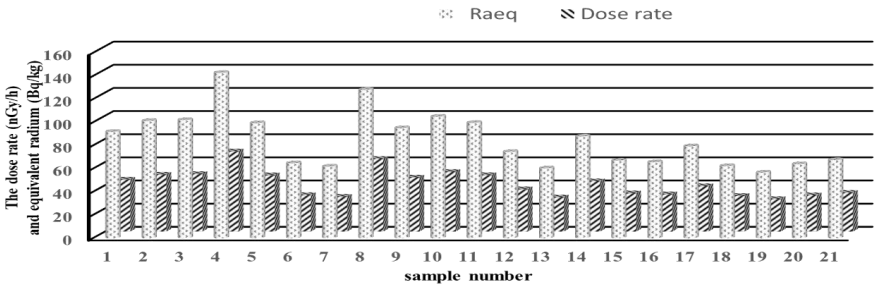

Figure 3: average activity radium equivalent ($Bq.Kg^{-1}$), absorbed dose rate of medicinal plants.

3.2.3 The Annual Effective Dose Equivalent (AEDE)

Using an outdoor occupancy factor of 0.2 and an indoor occupancy factor of 0.8, the absorbed dose was converted to the annual effective dose equivalent (AEDE), and a dose conversion factor of 0.7 Sv Gy^{-1} is provided by

$$E_{eff}\left(\frac{\text{mSv}}{\text{y}}\right) = \text{Dose rate} \times 8766 \times 0.7 \times 0.2 \times 10^{-6} \quad (4)$$

The annual effective dose equivalent for the location under study, as given in Table 3. The comparable global average is 1 $mSvy^{-1}$ [17], [18], [19].

3.2.4 Hazard Index (H)

The external hazard index (H_{ex}) and internal hazard index (H_{in}) for samples is given by the following equations [20], [21], [22]

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (5)$$

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (6)$$

Where A_K, A_{Ra} and A_{Th} are the activity concentrations (Bq kg^{-1}) of the specific radiation. The internal hazard index has to be less than one as well to provide safe radionuclide levels in medical plants[15]. As shown in Table 3, all of which are below the crucial value of unity.

Table (3): The average external hazard index (H_{ex}) and internal hazard index (H_{in}) and Representative Level Index of the samples studied

Sample number	Hex	Hin	Iγ	Raeq (Bq.Kg^{-1})	Dose rate (nGy/h)	annual effective doses (mSv/y)
1	0.3	0.3	0.2	91.9	44.9	0.06
2	0.3	0.4	0.2	101.4	49.1	0.06
3	0.3	0.4	0.2	102.2	49.7	0.06
4	0.4	0.5	0.4	142.9	69.2	0.08
5	0.3	0.4	0.2	99.5	48.6	0.06
6	0.2	0.2	0.2	64.8	31.5	0.04
7	0.2	0.2	0.1	62.0	30.0	0.04
8	0.4	0.5	0.3	128.2	62.8	0.08
9	0.3	0.4	0.2	95.2	46.6	0.06
10	0.3	0.4	0.3	105.1	51.5	0.06
11	0.3	0.4	0.2	99.7	48.7	0.06
12	0.2	0.3	0.2	74.6	36.5	0.04
13	0.2	0.2	0.1	60.4	29.5	0.04
14	0.2	0.3	0.2	87.9	43.4	0.05
15	0.2	0.2	0.2	67.1	33.1	0.04
16	0.2	0.2	0.2	65.7	32.2	0.04
17	0.2	0.3	0.2	79.5	39.2	0.05
18	0.2	0.2	0.2	62.4	30.8	0.04
19	0.2	0.2	0.1	56.7	28.0	0.03
20	0.2	0.2	0.2	64.1	31.3	0.04
21	0.2	0.3	0.2	67.6	33.4	0.04
Maximum	0.4	0.5	0.4	142.9	69.2	0.08
Minimum	0.2	0.2	0.1	56.7	28.0	0.03
Average	0.2	0.3	0.2	84.7	41.4	0.05

3.2.5 Representative Level Index (I_γ)

Representative Level Index (I_γ) was calculated using the following equation [14], [21].

$$I_\gamma = (1/300)\, A_{Ra} + (1 / 2200) A_{Th} + (1 / 3000) A_K \quad (7)$$

Where A_{Ra}, A_{Th} and A_K are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K in Bq/Kg, respectively. The average representative level index, I_γ and external hazard index, internal hazard index as shown in Figure 4

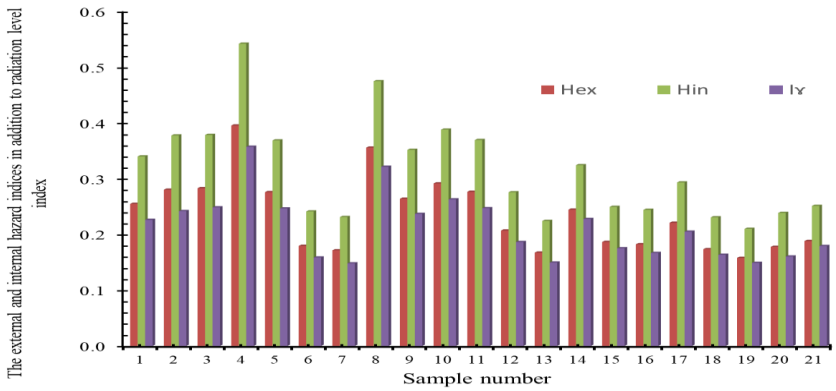

Figure 4: Average radiological hazards and representative level index (H_{ex}, H_{in}, I_{γ}) of medicinal plants.

Conclusion

The average radioactivity concentrations of the isotopes ^{226}Ra, ^{232}Th, and ^{40}K for various medicinal plants commonly used in Al-Qassim region were estimated as follows 29.4±0.3 (54.2-19.3) $Bq.Kg^{-1}$, 23.5±0.2 (38.8-14.8) $Bq.Kg^{-1}$ and 309.5±3.3 (474.8-197.5) $Bq.Kg^{-1}$, respectively. It was found that the concentrations of ^{226}Ra, ^{232}Th, and ^{40}K were lower than the concentrations reported in the literature. The average annual equivalent dose that can be exposed due to the consumption of medicinal plants was assessed as 0.05(0.08-0.03) $mSvy^{-1}$. The values of radium equivalent activity (Ra_{eq}) fall within the internationally permissible maximum limit of 370 $Bq.Kg^{-1}$, and the absorbed does rate is below the permissible limit. The internal and external hazard indices, along with the representative level index, were calculated for the samples analyzed, and all values were found to be less than unity. This indicates that the consumption of these medicinal plants does not pose any significant radiological risk to public health. Accordingly, the findings of this study provide a reliable baseline dataset that can support future investigations on natural radioactivity in medicinal plants from the Al-Qassim region and contribute to the development of region-specific safety guidelines.

Acknowledgment

The authors gratefully acknowledge Qassim University, represented by the Deanship of Graduate Studies and Scientific Research, on the financial support for this research under the number (QU-J-PG-2-2025-53219) during the academic year 1446 AH/2024 AD.

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