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Original Article

Determination of Radionuclides Concentration and Average Annual Committed Effective Dose Due to Ingestion for Some Medicinal and Edible Plants from Shazand (Markazi Province), Iran

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Abstract

Introduction
Natural and artificial radionuclides are the main sources of human radiation exposure, which enter the food chain from the environment. Radionuclides can affect human health by transferring from soil to plants and entering the human body. In this research study, we aimed to determine the activity concentrations of radionuclides and calculate the average annual committed effective dose (AACED) due to the ingestion of some medicinal and edible plants from Shazand, Markazi Province, Iran.

Materials and Methods
The specific activities of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs in eight medicinal and edible plant species and their surrounding soils were determined, using gamma-ray spectrometry and a high-purity germanium (HPGe) detector. The samples were collected from Shazand, situated in Markazi Province, Iran.

Results
The maximum soil-to-plant transfer factor (TF)for ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cswas estimated at 0.13, 0.13, 3.17, and 0.09, respectively. Based on the findings, the AACED for 1kg of edible plants varied from 0.63±0.16 to 13.71±0.14μSv/y. The annual gonadal dose Equivalent (AGDE) varied from 556 to 717 μSv/y for the soil samples (mean: 642μSv/y). The threshold consumption rate for edible plants ranged between21.86and 82.63 kg/y.

Conclusion
In this study, AACED due to the ingestion of radionuclides was lower than the global average (0.3 mSv/y). Therefore, the present results indicated no radiation health risks, associated with the use of these medicinal plants for disease treatment. However, it should be noted that all soil samples and some plants were polluted by radiocesium.

Keywords: Natural radiation, Medicinal plant, Radionuclide

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1. Introduction

A significant amount of radiation exposure to humans originates from natural and artificial sources. Uranium, thorium series, and potassium are the main elements, contributing to natural terrestrial radioactivity [1]. The average ^{238}U content in the Earth's crust has been estimated at 2.7 mg/kg. Moreover, the concentration of this element may be as high as 120 mg/kg in phosphate rocks [2].

According to a study by Firestone et al. (1996),the average ^{232}Th content in the Earth's crust is approximately 9.6 mg/kg [3]. Environmental pollution by artificial radioactive isotopes originates from nuclear weapon tests and accidents in nuclear power plants [4, 5]. Release of radionuclides from nuclear facilities to the environment under normal or unplanned circumstances can potentially lead to radiation exposure to the nearby biota and human populations.

Radionuclides, released and dispersed into the atmosphere, enter the terrestrial environment due to the dry and wet deposition of soil and plants, leading to exposure to humans or biota [6]. Consequently, the study of radionuclides in agricultural regions is a global concern, as most of these radionuclides can be recycled within the biota, similar to nutrients in the soil. Interactions between radionuclides and soil depend on the chemical form of the element and some soil properties, such as pH, mineralogical composition, organic matter content, and nutrient status [7-9].Plant uptake of radionuclides is dependent on these interactions and the metabolic and physiological characteristics of plant species. Therefore, radionuclide mobility needs to be determined in soil to define potential hazard
s, caused by the entrance of radionuclides into the food chain.

Absorbed elements by plants may be redistributed within the plants through the root system (indirect contamination) or aerial organs (direct contamination) [10].Considerable differences have been detected in the uptake and translocation of long-lived radionuclides among different plant species [11]. These radionuclides are transported to specific tissues, depending on their function in plant metabolism, which is reflected in their higher concentration in a particular part of the plant.

Radionuclides can be also picked up along with nutrients and may show similar chemical behaviors as essential nutrients. In order to study the translocation and preferred site of radionuclide accumulation, in some cases, the total content of radionuclides in the whole plant has been standardized for the dry weight fraction of each plant part [12].

With this background in mind, we aimed to determine the activity concentrations of radionuclides, using gamma-ray spectrometry and a high-purity germanium (HPGe) detector in different medicinal and edible plant species. The average annual committed effective dose (AACED) for the consumption of 1 kg of all the samples and the maximum permissible consumption rates of edible plants were estimated. Radionuclide concentration was measured in cultivated soils. Also, the transfer of radionuclides from soil to plant was evaluated, and the radiological hazard was assessed in all the soil samples.

2. Materials and Methods

2.1. Sampling and plant identification

Eight medicinal and edible plant samples were collected from Shazand, Markazi Province, Iran. Shazand is located at a latitude of 33°N and longitude of 49°E, as presented in Figure 1. All the collected plant samples were transferred to the Plant Systematic Laboratory of Arak University and were identified according to the literature [13, 14].

2.2. Sample preparation

The collected medicinal and edible plants included Salvia nemorosa L., Triticum aestivum L., Peganum harmala L., Vitis vinifera cv. Shirazi, Medicago sativa L., Gundelia Tournefortii L.,Descurainia sophia L. Webb et Berth, and Achillea vermicularis Trin. In this study, by using random integration and experimental sampling, a combined approach was implemented to collect the samples.

Figure 1. Sampling location (Bagh Baraftab village in Shazand), Markazi Province-Iran.

The samples were obtained at various stages with an average fresh weight of 5 kg for the complete plant, including the root and aerial parts, with the exception of *V. vinifera*, the aerial part of which was examined. The root samples were placed in distilled water for 15 min and washed to remove the soil and other unwanted materials. Then, the samples were chopped into small pieces with a plastic knife and spread with special care for preliminary drying.

The dried plants were placed at oven temperature of 80℃for two days. All the collected samples were crushed, homogenized, and passed through a 50-mesh screen to prepare fine powder in the laboratory. The prepared samples were maintained in standard sealed Marinelli beakers for a minimum of 50 days. This period was required for reaching radioactive equilibrium, where the decay rate of daughters became equal to that of the parent.

The net weight of each sample was 330g. The soil samples were obtained from the samilar area at a soil depth of 5 cm by employing the template method. The soil samples were dried at oven temperature of 200℃ for 12 h. In order to obtain homogeneous samples, the dried soil samples were pulverized by a grinder into fine powder and passed through 10- and 50-mesh screens. Also, the same procedure was applied for packing and sealing the samples in Marinelli beakers. The net weight of each sample was 950g [15].

2.3. Radionuclide activity measurement

Gamma-ray spectrometry was performed, using an HPGe detector (model GCD30195, Baltic Science Instruments Ltd.) with a relative efficiency of 30% and full width at half maximum (FWHM) of 1.95keV for Co gamma ray energy line at 1332.520 keV. The detector was shielded in a two-layered chamber (10 cm thick lead and 2 mm thick copper); this shield served to reduce background radiation.

The soft components of cosmic ray, consisting of photons and electrons, were reduced to a very low level by 100 mm of lead shielding. X-ray (73.9 keV), emitted from the lead through its interaction with external radiation, was suppressed by the copper layer [16]. To minimize the effect of scattering radiation from the shield, the detector was located in the center of the chamber. Then, the sample was placed in a face-to-face geometry over the detector.

The obtained spectra were registered, using LRSM-BSI software with a multi-channel

analyzer (8192 channels). The registered gamma ‑ ray spectra were analyzed, and specific activities were calculated using EG&G Ortec Mastro II Gamavision 32 software (Tennessee 37831 USA). The ^{226}Ra activity of the samples was determined with 351.9 keV and 609.3 keV gamma ray lines of ^{214}Pb and ^{214}Bi, respectively.

The ^{232}Th activity was calculated, using ^{228}Ac gamma ray line at 911.21 keV and 968.97 keV with an intensity of 26.6% and emission percentage of 17.4%. The ^{40}K and ^{137}Cs activities were measured, using1460.7 and 661.66keV gamma ray lines, respectively. The background radiation was measured under similar conditions with an empty Marinelli beaker and was deducted from all the spectra.

The standard sample sources, including ^{137}Cs, ^{152}Eu, and ^{241}Am with specific activities, were used for the calculation of absolute efficiency. The efficiency calibration of the detector was obtained, based on the International Atomic Energy Agency (IAEA)-154instruction [17]. Geometrical conditions for gamma-ray spectra measurement of samples, standard mixed source and background radiation were the same. The counting time of all the spectra was86, 400 s.

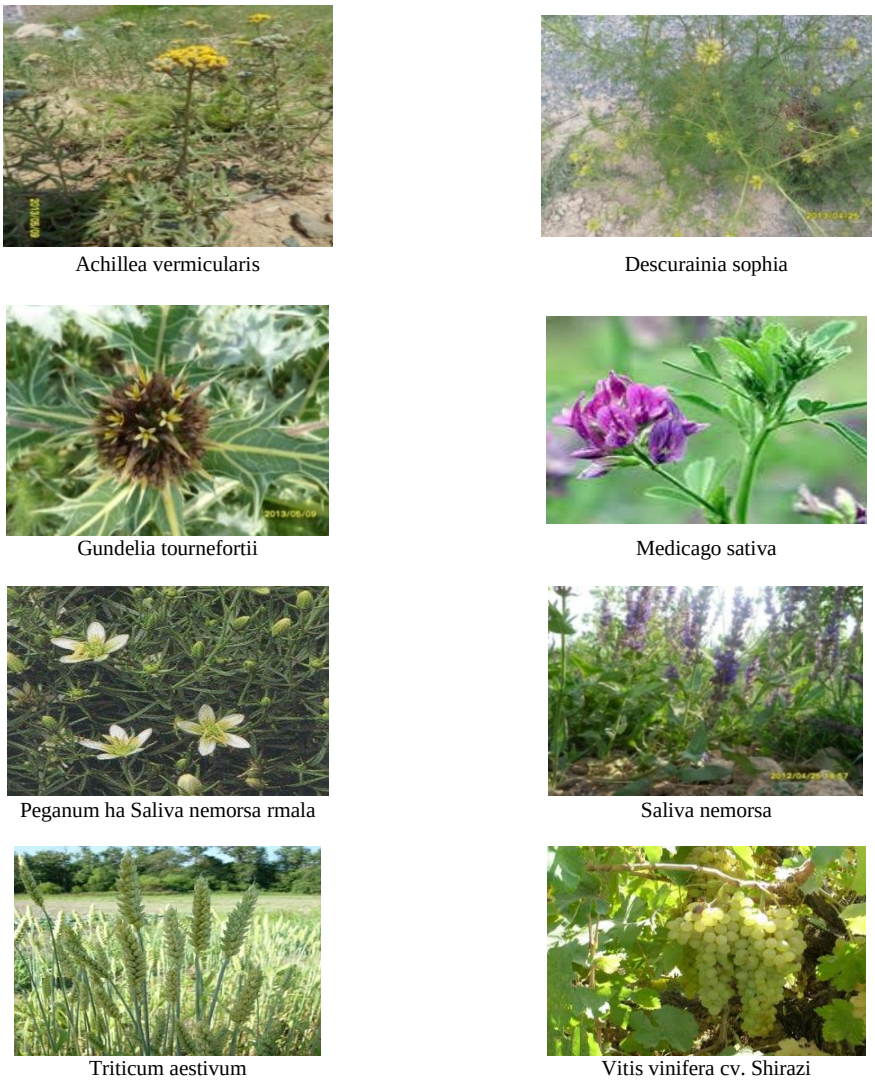

Figure 2. All of studied plant pictures

According to the registered gamma ray spectrum, the absolute efficiency of the detector configuration was measured as follows [17]:

$$\varepsilon = \frac{C_{net}}{Act \times P_n(E_i)\% \times T} \times 100 \quad (1)$$

Where ε is the efficiency for gamma ray energy E_i, C_{net} is the net count under the full-energy peak corresponding to E_i energy, Act denotes the radionuclide activity on the measurement date, $P_n(E_i)$ is the probability of E_i photon emission, and T refers to the counting time. The experimental data were fitted by polynomial approximation, using the Table Curve software version 2D 5.01 and Equation (2):

$$Y=a+b(Lnx)+c(Lnx)^2+d(Lnx)^3+e(Lnx)^4+f(Lnx)^5 \quad (2)$$

Where Y is the efficiency, a, b, c, d, e and f are constant values, and x denotes the gamma ray energy, expressed in keV. The calibration curve was fitted to the experimental data, as presented in Figure 2. The nuclear activities were calculated as follows [17]:

$$Act = \frac{C_{net}}{\varepsilon \times (B.R.) \times T \times m} \times 100 \quad (3)$$

Where $C_{net}$is the net count under the peak, Act denotes the activity concentration of the sample (Bq/kg), ε is the energy efficiency for the gamma ray by the detector, B.R. is the branching ratio of gamma ray intensity percentage, T denotes the time of spectra (s), and m refers to the mass of the sample (kg).

2.4. Evaluation of the radiological effects of soil samples

The external hazard index is a relation that quantifies the exposure factor. Radiation hazards due to the specific activities of radionuclides, including Ra, Th, and K, were determined by two different indices. In general, the most widely used radiation hazard index is known as the radium equivalent activity (Ra_{eq}). Ra_{eq} is the weighted sum of Ra, Th, and K radionuclide activities, based on the estimation that 370 Bq/kg of Ra, 259 Bq/kg of Th and 4810 Bq/kg of K produced the same gamma-ray dose rates at a 1 m distance above the rocks and soil. Ra_{eq} activity was calculated as follows [18]:

$$Ra_{eq}\ (Bq/kg) = A_{Ra} + 1.43\ A_{Th} + 0.077\ A_K \quad (4)$$

Where A_{Ra}, A_{Th}, and A_K are the specific activities of ^{226}Ra, ^{232}Th, and ^{40}K, respectively, expressed in Bq/kg.

The external hazard index (H_{ex}) only incorporates the external exposure risk due to gamma rays and corresponds to a maximum Ra_{eq} activity of 370 Bq/kg for the soil and rocks. The internal exposure to radon and its daughter products was quantified by the internal hazard index (H_{in}). H_{ex} and H_{in} were calculated as follows [19]:

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

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

The values of H_{ex} and H_{in} should be below the unity for the radiation risk to be negligible [20]. Also, the total absorbed dose rate in air (nGy/h) was determined as follows [20]:

$$D\ (nGy/h) = 0.427\ A_{Ra} + 0.662\ A_{Th} + 0.0432\ A_K \quad (7)$$

where A_{Ra}, A_{th}, and A_K are the mean specific activities of ^{226}Ra, ^{232}Th, and ^{40}K in rock or soil samples, respectively.

2.5. Annual gonadal dose equivalent (AGDE)

The organs of interest by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) include thyroid, lungs, bone marrow, bone surface cells, gonads, and female breast. AGDE due to the specific activities of ^{226}Ra, ^{232}Th, and ^{40}K in the soil was determined as follows [21]:

$$AGDE\ (\mu Sv/year) = 3.09\ A_{Ra} + 4.18\ A_{Th} + 0.314\ A_K \quad (8)$$

2.6. Transfer Factor (TF)

The soil-to-plant TF indicated the transfer of radionuclides from soil to plants through the plant roots. The TF values were determined based on the measured activity concentrations of radionuclides in the plant and the surrounding soil [22]:

$$TF = \frac{\text{Activity of radionuclides in plant } (\frac{Bq}{kg} \text{ dry weight})}{\text{Activity of radionuclides in soil } (\frac{Bq}{kg} \text{ dry weight})} \quad (9)$$

2.7. *Average annual committed effective dose* (*AACED) measurements*

The AACED due to the ingestion of naturally occurring radioactive materials (NORMs) in medicinal plants was estimated as follows [23]:

$$E_{ave} = C_r \times DCF_i \times A_i \quad (10)$$

where E_{ave} denotes AACED, C_r is the consumption rate of radionuclides, and DCF_i is the dose conversion factor for each radionuclide (2.8×10^{-7}, 2.3×10^{-7}, 6.2×10^{-9}, and 1.3×10^{-8}Sv/Bq for ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs, respectively), and A_i is the activity concentration of each radionuclide. According to Equation 11, AACED for an individual is directly proportional to the consumption rate of the components of medicinal plants. By using the same equation, the threshold consumption rate for a medicinal plant was measured as follows [12]:

$$\mathbf{C_r} = \frac{E_{ave}}{\sum_{i=1}^{4}(DCF_i \times A_i)} \quad (11)$$

where E_{ave}= 0.3 mSv/y is the threshold AACED due to the ingestion of NORM in the medicinal plants, A_i denotes the activity concentration of radionuclide i, and DCF_i is the dose conversion factor for the radionuclides.

3. Results

The results of plant identification showed that the evaluated plants, with the exception of *A.*vermicularis and G. tournefortii, belonged to different families. Table 1 demonstrates the calculated radionuclide concentrations (Bq/kg) in the eight studied plant species and the surrounding soil, soil-to-plant TF values of radionuclides, AACED, and the annual acceptable criteria for the consumption of edible plants (C_r).The radiological parameters of the studied soil, including external and internal hazard indices, absorbed dose rate in the air, Ra_{eq}, indoor and outdoor annual effective dose rates (AED_{indoor} and $AED_{outdoor}$), and AGDE values are also presented in Table 2.

4. Discussion

The specific activities of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs in plant samples varied from 2.27±0.45 to 7.43±0.60, from the minimum detectable activity (MDA) to 7.79±1.40, from MDA to 2750±10, and from MDA to 1.02±0.35 Bq/kg, respectively. The corresponding specific activities of these radionuclides in the soil samples ranged from 53.58±0.50 to 61.60±0.59, 49.97±1.01 to 60.98±1.08, 579.90±5.21 to 866.58±6.16,and 7.7±0.22 to 20.46±0.43 Bq/kg, respectively. These values were found to be higher than the mean global concentration for natural radionuclides (35,40, and 400 Bq/kg, respectively) [24].

The highest^{226}Ra activity concentration was determined for T. aestivum seeds, V. vinifera,andA. vermicularis. Also, thehighest ^{232}Th activity concentration was determined for A.vermicularis, S. nemorosa, and V. vinifera. The specific activity of ^{40}K was obviously greater than other radionuclides, except for D. sophia.

Since potassium channels are involved in the treatment of respiratory system complications, such as asthma, cough, and chronic obstructive pulmonary disease, higher concentrations of potassium in A. vermicularis Trin.andG. Tournefortii L.support their medicinal application for the treatment of these ailments. This finding also shows that P. harmala has maximum ^{40}K activity concentration in comparison with other plants; therefore, it could be a suitable option for absorption of ^{40}K.

G.tournefortii, S. nemorosa, and A. vermicularisshowed the highest^{40}K activity concentrations, respectively, while the lowest specific activity of ^{40}K was observed in the aerial parts and seeds of D. sophiaand T. aestivum, respectively. This study showed that the specific activity concentration of ^{40}K in T. aestivum chaff was higher than its seed.

Table 1. Specific activities of radionuclides (Bq/kg) and their transfer factors, Average annual committed effective dose (AACED) and maximum acceptable of consumption value (C_r).

Plant name	Measurable Quantity	^{226}Ra	^{232}Th	^{40}K	^{137}Cs
Achillea vermicularis Trin.	Activity in soil	61.60±0.59	60.98±1.08	866.58±6.16	9.49±0.24
	Activity in plant	4.07±0.57	7.79±1.40	1150 ±10	0.71± 0.24
	TF	0.06	0.13	1.32	0.07
	AACED (μSv/y)	10.07 ± 0. 36			
	C_r (kg/y)	29.78			
Descorainia Sophia L.Webb et Berth	Activity in soil	61.60±0.59	60.98±1.08	866.58±6.16	9.49±0.24
	Activity in plant	2.28±0.60	< MDA	< MDA	< MDA
	TF	0.04	----	----	----
	AACED (μSv/y)	0.63 ± 0. 16			
	C_r (kg/y)	469.92			
Gondelia tournefortii L.	Activity in soil	61.60±0.59	60.98±1.08	866.58±6.16	9.49±0.24
	Activity in plant	2.27±0.45	< MDA	2110 ± 10	< MDA
	TF	0.04	----	2.43	----
	AACED (μSv/y)	13.71 ± 0. 14			
	C_r (kg/y)	21.86			
Medicago sativa L.	Activity in soil	58.91±0.66	56.31±1.07	761.40±6.34	12.70±0.25
	Activity in plant	3.00±0.65	< MDA	898 ± 14	1.02±0.35
	TF	0.05	----	1.18	0.08
	AACED (μSv/y)	6.42 ± 0. 20			
	C_r (kg/y)	46.72			
Peganum harmala L.	Activity in soil	61.60±0.59	60.98±1.08	866.58±6.16	9.49±0.24
	Activity in plant	3.43±0.63	< MDA	2750 ±10	0.89 ±0.24
	TF	0.05	----	3.17	0.09
	AACED (μSv/y)	18.02 ± 0. 18			
	C_r (kg/y)	16.64 (not edible plant)			
Saliva nemorsa L.	Activity in soil	58.91±0.66	56.31±1.07	761.40±6.34	12.70±0.25
	Activity in plant	3.72±0.05	6.17±0.09	1460 ±10	0. 85 ± 0.25
	TF	0.06	0.11	1.91	0.06
	AACED (μSv/y)	11.52 ± 0.06			
	C_r (kg/y)	26.03			
Triticum aestivum L. (Aerial part)	Activity in soil	58.07±0.76	51.01±1.29	785.73±6.29	20.46±0.43
	Activity in plant	3.77±0.59	< MDA	491± 9	< MDA
	TF	0.06	----	0.63	----
	AACED (μSv/y)	4.09 ± 0. 17			
	C_r (kg/y)	73.17 (not edible part)			
Triticum aestivum L. (seed)	Activity in soil	58.07±0.76	51.01±1.29	785.73±6.29	20.46±0.43
	Activity in plant	7.43±0.60	< MDA	250 ± 6	< MDA
	TF	0.13	----	0.32	----
	AACED (μSv/y)	3.63 ± 0. 17			
	C_r (kg/y)	82.63			
Triticum aestivum L. (chaff)	Activity in soil	58.07±0.76	51.01±1.29	785.73±6.29	20.46±0.43
	Activity in plant	3.79±0.65	< MDA	721± 9	< MDA
	TF	0.06	----	0.92	----
	AACED (μSv/y)	5.53 ± 0. 19			
	C_r (kg/y)	54.23 (not edible part)			
Vitis vinifera cv. Shirazi	Activity in soil	53.58±0.50	49.97±1.01	579.90±5.21	7.71±0.22
	Activity in plant	4.15±0.57	4.51±1.18	837 ± 1	< MDA
	TF	0.08	0.09	1.44	----
	AACED (μSv/y)	7.38 ± 0. 31			
	C_r (kg/y)	40.60			

Table 2. Calculated of radiological hazard assessment for soils samples

Soil code	H_{in}	H_{ex}	Ra_{eq}	D (nGy/h)	AEDindoor (mSv)	AEDoutdoor (mSv)	AGDE (μSv/y)
$Soil_1$	0.75	0.59	215.49	104.11	0.51	0.127	717.35
$Soil_2$	0.70	0.54	198.06	95.32	0.47	0.12	656.48
$Soil_3$	0.67	0.52	191.52	92.51	0.45	0.11	639.38
$soil_4$	0.60	0.45	169.96	81.01	0.40	0.10	556.53

Considering the necessity of ^{40}K for plant growth, ^{226}Ra and ^{40}K contamination in all the studied plants was greater than that observed in tea samples from Guilan Province, Iran [25].The specific activity of ^{137}Cs was very low in some of the studied samples. Also, the specific activity of ^{137}Csinthe majority of the samples was lower than MDA. The maximum value of $^{137}Cs$was observed in M.sativa, P. harmala, S. nemorosa and A. vermicularis.

The results of the evaluation of radiological parameters in the soil samples are listed in Table 2. External and internal hazard indices in soil samples obtained less than unit and the absorbed dose rate in air for all the soil samples was higher than the average global value (55 nGy/h).The activity concentration of Ra_{eq} was lower than the maximum acceptable value for all the studied soil samples (370 Bq/kg).Indoor and outdoor AED values varied from 397 to 511 μSv/y and 99 to 127 μSv/y, respectively, which were higher than the average global value (70 μSv/y).

AGED varied from 556 to 717μSv/y, with the mean value of 642μSv/y. The mean AGED for the soil samples was 439.73 μSv⁄y in Nigeria [26], 2398 μSv⁄y in Egypt [27], and 182.52 μSv⁄y in Saudi Arabia [28]. All the soil samples were polluted by ^{137}Cs, which was originated from countries with nuclear power and was transferred to these regions through atmospheric processes. The maximum soil-to-plant TF values of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs were obtained 0.13, 0.13, 3.17, and 0.09 respectively.

The AACED for 1kg of edible plants varied from0.63±0.16 to 13.71±0.14 μSv/y; the measured values were lower than the maximum global acceptable value (0.3mSv/y). Considering the maximum acceptable AACED for edible plants, the threshold consumption rate was within the range of 21.86-82.63 kg/y (Table 2).

5. Conclusion

In this research study, the specific activities of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs were determined in eight medicinal and edible plants and the surrounding soil, using gamma-ray spectrometry method and an employing HPGe detector system. The AACED for the studied edible plants was within the acceptable range. It is suggested that the specific activities of radionuclides in frequently used edible and medicinal plants be evaluated to assure their safety.

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