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Measurement of ^{226}Ra, ^{228}Ra, ^{137}Cs and ^{40}K in edible parts of two types of leafy vegetables cultivated in Tehran province-Iran and resultant annual ingestion radiation dose

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Background: The sources of radioactivity in the environment have natural, terrestrial and extraterrestrial, and anthropogenic origins. Plants may get radioactive nuclides in two ways: (i) by the deposition of radioactive fallout, (ii) by absorption from the soil. ***Materials and Methods:*** The Concentrations of the natural radionuclides (^{226}Ra, ^{228}Ra, ^{40}K) and the artificial radionuclide (^{137}Cs) in leek and parsley in Tehran province-Iran were determined using HPGe. Also the effective dose due to the ingestion of such vegetables by the population of Tehran province was studied. ***Results:*** The average value of radionuclide concentrations in parsley samples were measured 177.69 ± 12.47 mBq kg^{-1} $_{fresh}$ for ^{226}Ra; 349.62 ± 28.42 mBq kg^{-1} $_{fresh}$ for ^{228}Ra; 187364.6 mBq kg^{-1} $_{fresh}$ for ^{40}K. The average value of radionuclide concentrations in leek samples were measured 94.31 ± 6.46 mBq kg^{-1} $_{fresh}$ for ^{226}Ra; 207.47 ± 19.46 mBq kg^{-1} $_{fresh}$ for ^{228}Ra; 174555 ± 1704.21 mBq kg^{-1} $_{fresh}$ for ^{40}K. The concentrations of ^{137}Cs in most of Parsley and Leek samples were below the minimum detectable activity (MDA). ***Conclusion:*** The Average ^{226}Ra and ^{228}Ra activities in 29 leek and parsley samples were about 2.63 and 6.78 times the reference values, respectively. The annual effective dose resulting from the studied radionuclides for the adult population in Tehran province were found to be safe in comparison with normal background areas. **Iran. J. Radiat. Res., 2010; 8 (2): 103-110**

Keywords: *^{226}Ra, ^{228}Ra, ^{40}K, ^{137}Cs, leafy vegetable, HPGe, effective dose.*

INTRODUCTION

The sources of radioactivity in the environment have natural, terrestrial and extraterrestrial, and anthropogenic origins [1] which are resulted from natural processes, medical and industrial uses of radioisotopes, nuclear weapon tests, nuclear accidents and the operation of nuclear power plants [2]. During the process of fallout, the airborne particles may be intercepted by plants or return to the top soil. Therefore, Plants may get radioactive nuclides in two ways (i) by the deposition of radioactive fallout on the plants directly and (ii) by absorption from the soil [3].

According to a report by UNSCER, the total exposure per person resulting from ingestion of terrestrial radioisotopes was 0.29 mSv, of which 0.17 mSv was from ^{40}K and 0.12 mSv was from thorium and uranium series. Exposure from inhalation of terrestrial radioisotopes contributes another 0.01 mSv [4].

There have been several studies of radionuclide concentrations in vegetables from normal background areas, such as those for ^{226}Ra [5, 6], ^{228}Ra [5, 7], ^{40}K [8-10] and ^{137}Cs [6, 11], also in vegetables from areas with high levels of natural radiation (HLNR), such as that for ^{226}Ra conducted in Ramsar- Iran [12].

The main goals of the present study were: 1) to determine ^{226}Ra, ^{228}Ra, ^{40}K and ^{137}Cs concentrations in two kinds of cultivated vegetables in Tehran province to establish baseline data for the concentration

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of the radionuclides in such vegetables in that area; 2) to assess the effective dose due to the ingestion of such vegetables by the population of Tehran province.

The Tehran province is approximately located between 34° and 36.5° N latitude and between 50° and 53° E longitude. Figure 1 shows the map of a part of Tehran province and the location of the sampling sites.

MATERIALS AND METHODS

Sites and vegetables selection

The criteria for the vegetable selection were commonly consumed edible vegetables by the people of Tehran province and time limitation of project. Therefore, among the edible vegetables (herbs), parsley and leek were selected. Most of the lands in which vegetables were cultivated are located in the west and south of Tehran province; therefore, the vegetables were taken from the vegetable cultivation lands in Karaj, Shahriyar, Islamshahr, Robat karym, Ray, Varamin and Pakdasht townships.

Sampling procedure

The geographical coordinates of the sampling locations were recorded using a GPS device. The samples were cut 2-3 cm above the soil surface, so that soil contamination of the samples would be minimized. Approximately 3kg of parsley and 5 kg of leek were collected from each point that equals to 330 grams (according to the volume of our Marinelli beakers) of the dried and grinded sample (table 1).

29 samples (13 parsley samples and 16 leek samples) were collected during April and May 2008 (table 2).

Sample preparation

The fresh vegetables were hand-cleaned to remove soil particles and then were weighed. The samples were washed with tap water, oven dried at 80 °C, ground, sieved and then reweighed. 330 grams of each of the 29 samples were sealed in Marinelli beaker (capacity 1000 cm^3) and stored for about 4 weeks prior to gamma spectrometry analysis to allow secular equilibrium between ^{226}Ra and ^{222}Rn and their decay products.

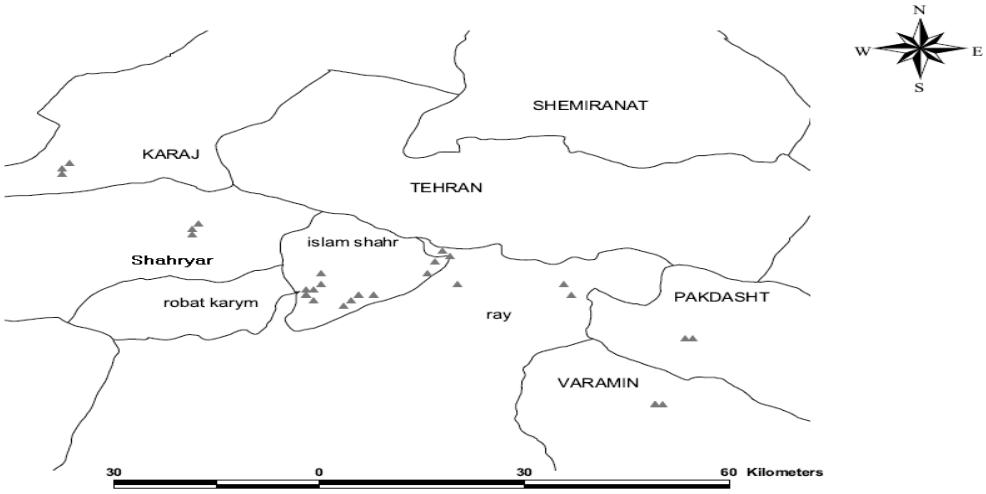

Figure 1. The map of part of Tehran province; and the location of the sampling sites (▲).

Table 1. Types of vegetables with dry-to-fresh weight ratios.

Type of vegetable	Scientific name	Weight ratio (dry/fresh)
Leek	Allium sp	0.07
Parsley	Petroselinum crispum Mill.	0.11

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Table 2. Geographical coordinates of the sampling points.

Sample label	Location	Geographical coordinates (Degree)	Sample label	Location	Geographical coordinates (Degree)
LP01-1	Varamin	N35.33276E51.67236	LP09-5	Isalmshahr	N35.53131E51.29741
LP01-2	Varamin	N35.33184E51.67108	L09-6	Isalmshahr	N35.53160E51.30382
LP01-3	Varamin	N35.33231E51.67142	LP10	Ray	N35.57192E51.36953
LP02	Varamin	N35.33331E51.67726	LP11-1	Ray	N35.54572E51.41299
LP03-1	Islamshahr	N35.52508E51.21264	P11-2	Ray	N35.58977E51.38364
LP03-2	Islamshahr	N35.52992E51.20985	L11-3	Ray	N35.60762E51.38753
LP03-3	Islamshahr	N35.53301E51.20790	LP11-4	Ray	N35.60380E51.39733
LP03-4	Islamshahr	N35.51960E51.22108	LP12-1	Shahriar	N35.65231E51.05868
LP04-1	Islamshahr	N35.53222E51.20506	LP12-2	Shahriar	N35.66175E51.06905
LP04-2	Islamshahr	N35.52716E51.20917	L13	Shahriar	N35.63651E51.06315
L05	Islamshahr	N35.53571E51.21506	L14	Karaj	N35.76692E50.90496
P06	Islamshahr	N35.53986E51.21407	L15	Karaj	N35.75229E50.88785
L07	Islamshahr	N35.55025E51.23346	P15	Karaj	N35.75581E50.88563
LP08	Islamshahr	N35.57411E51.22761	LP16-1	Ray	N35.53040E51.55520
LP09-1	Islamshahr	N35.51314E51.26263	LP16-2	Ray	N35.55355E51.55046
LP09-2	Islamshahr	N35.52100E51.26923	LP17-1	Pakdasht	N35.44890E51.70997
L09-3	Islamshahr	N35.52993E51.27917	LP17-2	Pakdasht	N35.45307E51.71650
P09-4	Islamshahr	N35.53115E51.27778			

Measurement techniques

The Samples were measured using a p-type coaxial (HPGe) gamma spectrometer with high resolution (1.98 keV at 1.33 MeV), relative efficiency (38.5%) and low background to determine gamma-ray emitters. The detector was calibrated using certified gamma-ray plant-based standard sources. The counting time for each sample was 24 h. The background spectra were measured frequently under the same conditions of sample measurements and were used to correct the calculated sample activities. The characteristic gamma peaks selected for the determination of the different radionuclides were 352 keV for the ^{226}Ra (^{214}Pb), 609 keV for the ^{226}Ra (^{214}Bi), 911, 969 and 338 keV for the ^{228}Ra (^{228}Ac), 661 keV for the ^{137}Cs and 1461 keV for the ^{40}K. The Minimum Detectable Activity for the measured natural radionuclides was derived from the background measurements at 81564 s (table 3).

Table 3. Minimum Detectable Activity (MDA) of the gamma counting system.

Parent	Daughter	Energy (keV)	MDA (mBq)
^{228}Ra	^{212}Pb	238.63	110±10
	^{228}Ac	338.32	310±50
	^{228}Ac	911.2	260±30
	^{228}Ac	968.97	330±170
^{222}Rn	^{214}Pb	351.93	120±20
	^{214}Bi	609.31	130±10
^{228}Th	^{208}Tl	583.41	60±10
^{40}K	^{40}K	1461	1880±60
^{22}Na	^{22}Na	511	80±3

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The efficiency equation for plant-based standards was generated and had an analytic form of:

$$\varepsilon(\%) = \frac{(a + c \times E + e \times E^2 + g \times E^3)}{1 + b \times E + d \times E^2 + f \times E^3}$$

These plant-based standards consist of ^{152}Eu, ^{137}Cs+ ^{241}Am and ^{133}Ba. In order to perform the quality control for making relevant samples, a sample with a defined activity was analyzed using the generated efficiency curve.

RESULTS AND DISCUSSION

Table 4 shows the concentrations of ^{226}Ra and ^{228}Ra via their daughters in each sample. The activities are given in mBqkg^{-1} fresh weight and the measurement errors shown represents one-sigma uncertainties.

The radionuclide concentrations in parsley samples ranged from 120 ± 50 to 260 ± 52 mBq kg$^{-1}_{fresh}$ (average value=177.69 ± 12.47 mBq kg^{-1}) for ^{226}Ra; from 240 ±110 to 440 ± 110 mBq kg$^{-1}_{fresh}$ (average value=349.62 ± 28.42 mBq kg^{-1}) for ^{228}Ra; from 120590 ± 4720 to 243940 ± 9540 mBq kg$^{-1}_{fresh}$ (average value=187364.6 mBq kg^{-1})for ^{40}K. The radionuclide concentrations in leek samples ranged from 45 ± 18 to 210 ± 20 mBq kg$^{-1}_{fresh}$ (average value= 94.31 ± 6.46 mBq kg^{-1})for ^{226}Ra; from 100 ± 70 to 330 ± 130 mBq kg$^{-1}_{fresh}$(average value= 207.47 ± 19.46 mBq kg^{-1}) for ^{228}Ra; from 124450 ± 4830 to 223990 ± 8650 mBq kg$^{-1}_{fresh}$ (average value= 174555 ± 1704.21 mBq kg^{-1}) for ^{40}K.

The concentrations of ^{137}Cs in most of the Parsley and Leek samples were below the MDA, except in 2 samples of Parsley that were 70±20 and 30±20 mBqkg$^{-1}_{fresh}$ and 3 samples of Leek that were 30±10, 40±20 and 30±10 mBqkg$^{-1}_{fresh}$. For leafy vegetables, reference values of 50 and 40 mBq kg^{-1}, have been reported for ^{226}Ra and ^{228}Ra, respectively [4]. Therefore, the average values of ^{226}Ra and ^{228}Ra activities in parsley samples in this study were about 3.55 and 8.74 times the reference values, respectively. The average values of ^{226}Ra and ^{228}Ra activities in Leek samples were about 1.89 and 5.19 times the reference values. Generally, the average values of ^{226}Ra and ^{228}Ra activity in these leafy vegetable samples (131.69 and 271.19) were about 2.63 and 6.78 times the reference values, respectively (table 5). As shown in this table, our values for ^{226}Ra were greater than those reported for the USA, China, Italy, Poland and U.K. [4], whereas they were smaller to the average values reported for Germany [4].

The ^{226}Ra concentrations measured for parsley and leek samples in this study were 82.33 and 41.57 times smaller than those measured in parsley and leek samples from Talesh Mahalleh, a district of the city Ramsar in Iran with a high level of natural radiation (HLNR), with ^{226}Ra mean concentrations of 14630 and 3920 mBqkg^{-1} fresh weight, respectively [12] . In table 6, data obtained from present study concerning ^{40}K and ^{137}Cs concentrations in Parsley and Leek samples (leafy vegetables) have been compared with those reported for leafy vegetables in some other countries, including Egypt [9] and Korea [10].

As shown in table 6 the average ^{40}K activity in these leafy vegetable samples (180297.23 ± 1316.41) is approximately equal to the mean value of Egypt and Korea samples (182145±37320). The average ^{137}Cs activity in these leafy vegetable samples (40.80 ± 7.48) is about 66.3 times smaller than the mean value of Egypt. Among the natural radioisotopes in our samples, the concentration of ^{40}K was the highest. Similar findings have been published by other authors, [7, 10] possibly due to the concentrations of 40K in the soil and the transfer factor of 40K, higher than some natural radioisotopes [13]. However, ^{40}K is an essential biological element and its concentration in human tissue is under close metabolic control [1]. The present study revealed that the concentrations of the radionuclides in parsley samples were higher than leek samples in the same location. The only exceptions are the concentration of ^{40}K in 8th and 11th samples.

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Table 4. The concentrations of the radionuclides measured in each sample.

Sample label	^{228}Ra (mBq kg$^{-1}_{fresh}$)				^{226}Ra (mBq kg$^{-1}_{fresh}$)		
	^{214}Pb	^{214}Pb	^{214}Bi	^{214}Bi	^{228}Ac	^{228}Ac	^{228}Ac
	295.22 (keV)	351.93 (keV)	609.31 (keV)	1120.28 (keV)	911.2 (keV)	968.97 (keV)	338.32 (keV)
P01	160 ± 60	170 ± 40	200 ± 40	< MDA	520 ± 110	< MDA	480 ± 160
P02	220 ± 70	< MDA	150 ± 40	< MDA	390 ± 130	< MDA	< MDA
P03	200 ± 70	210 ± 40	240 ± 40	< MDA	370 ± 130	< MDA	350 ± 120
P04	180 ± 70	120 ± 40	160 ± 60	< MDA	320 ± 150	< MDA	< MDA
P06	< MDA	150 ± 40	130 ± 40	< MDA	270 ± 120	230 ± 110	< MDA
P08	170 ± 60	< MDA	200 ± 70	< MDA	< MDA	< MDA	290 ± 140
P09	330 ± 120	180 ± 60	270 ± 80	< MDA	380 ± 150	< MDA	370 ± 170
P10	< MDA	150 ± 70	< MDA	< MDA	240 ± 110	< MDA	< MDA
P11	250 ± 120	170 ± 60	200 ± 60	< MDA	380 ± 90	< MDA	< MDA
P12	< MDA	180 ± 50	200 ± 50	< MDA	440 ± 120	< MDA	430 ± 180
P15	< MDA	170 ± 60	< MDA	< MDA	410 ± 120	< MDA	330 ± 130
P16	< MDA	120 ± 50	< MDA	< MDA	390 ± 170	< MDA	< MDA
P17	< MDA	130 ± 40	150 ± 40	< MDA	340 ± 90	< MDA	400 ± 160
L01	< MDA	70 ± 30	110 ± 30	< MDA	140 ± 50	< MDA	190 ± 80
L02	130 ± 50	110 ± 30	100 ± 30	< MDA	220 ± 50	< MDA	190 ± 90
L03	< MDA	80 ± 20	140 ± 50	< MDA	280 ± 70	300 ± 140	270 ± 110
L04	< MDA	80 ± 30	90 ± 40	< MDA	< MDA	< MDA	100 ± 70
L05	< MDA	80 ± 30	100 ± 20	< MDA	< MDA	< MDA	180 ± 80
L07	220 ± 40	180 ± 20	240 ± 30	< MDA	140 ± 50	< MDA	< MDA
L08	< MDA	60 ± 30	< MDA	< MDA	240 ± 100	170 ± 80	< MDA
L09	< MDA	80 ± 30	110 ± 50	< MDA	< MDA	< MDA	260 ± 130
L10	110 ± 50	70 ± 30	140 ± 40	< MDA	110 ± 70	220 ±120	< MDA
L11	< MDA	< MDA	80 ± 30	< MDA	180 ± 50	< MDA	< MDA
L12	< MDA	110 ± 50	70 ± 30	< MDA	< MDA	< MDA	330 ± 130
L13	< MDA	60 ± 30	< MDA	< MDA	190 ± 80	< MDA	< MDA
L14	< MDA	110 ± 30	60 ± 20	< MDA	220 ± 80	< MDA	220 ± 70
L15	< MDA	80 ± 30	< MDA	< MDA	170 ± 60	240 ± 90	300 ± 100
L16	< MDA	40 ± 20	50 ± 30	< MDA	< MDA	< MDA	< MDA
L17	160 ± 70	80 ± 30	80 ± 30	< MDA	240 ± 80	< MDA	< MDA

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Table 5. Concentrations of ^{226}Ra and ^{228}Ra in leafy vegetables in other countries and the reference values [4].

Region /Country	**Concentration (mBqkg^{-1})**	
	^{226}Ra	^{228}Ra
North America (United States) [4]	56	
Asia (China)[4]	75	220
Europe		
Germany[4]	6-1150	
Italy [4]	27-44	
Poland [4]	37-43	
U.K [4]	2.2-170	
Reference value	50	40
Present study	45 – 260 (mean 131.69)	100 – 440 (mean 271.19)

Table 6. Concentrations of ^{137}Cs and ^{40}K in leafy vegetables in other countries and the present study.

Country	Vegetable type	concentration (mBqkg^{-1} fresh)				Ref.
		^{137}Cs		^{40}K		
		Mean	Range	Mean	Range	
Egypt	Jew's mallow	4250±4100	1040–8870	328000±147000	202000-489000	9
	Roquette	1160±1050	90–2560	118000±26000	72000-134000	
	Jew's mallow and Roquette	2705±2116		223000±74640		
Korea	Spinach			185700±690		10
	Lettuce			96880±440		
	Spinach and Roquette			141290±409		
Mean		2705±2116		182145±37320		
Present Study	Parsley	50±14	< MDA- 70 ±20	2055±187365	120590-243940	
	Leek	33±8	< MDA- 40 ±20	1704±174555	124450 - 223990	
	Leek Parsley and	40±7	< MDA- 70 ±20	1316±180297	120590 - 243940	

The uptake of radionuclides by plants from the soil into plants is highly complex and depends on several factors including the plant species, soil conditions, the concentration of radionuclides in soil and the radionuclides availability in soil [12, 14]. As ICRP (1999) reported, the extra use of phosphate fertilizer can be a factor causing the increase in Radium radionuclide concentration in the leafy vegetables samples of Tehran province [15].

The annual effective dose was calculated using mean concentration of radionuclide in Bq per kilogram fresh weight, annual consumption rate and dose coefficient using the equation [14]:

Annual effective dose (Sv/year) = Concentration (Bq/kg) × Annual consumption (kg / year) × Dose conversion (Sv/Bq)

The dose coefficient were 2.8×10^{-7}

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Sv Bq^{-1} for ^{226}Ra, 6.7×10^{-7} Sv Bq^{-1} for ^{228}Ra, 6.2×10^{-9} Sv Bq^{-1} for ^{40}K, 1.3×10^{-8} Sv Bq^{-1} for ^{137}Cs [16]. The Consumption rate was defined using FAO definition (FAO, 2000) [17]. The annual consumption rate was calculated to be 4.69 kg /year. The annual effective dose due to each radionuclide is shown in table 7. The annual effective dose for the adult population of Tehran province was estimated to be 0.1729 mSv/year of ^{226}Ra and 0.85216 mSv/year of ^{228}Ra.

Table 7. Annual effective dose due to consumption of parsley and leek by population of the Tehran province.

Radionuclide	Annual effective dose (mSv/year)
^{226}Ra	0.17
^{228}Ra	0.85
^{40}K	5.24
Total	6.27
^{137}Cs	2.49×10^{-3}
Total+ ^{137}Cs	6.27

The committed effective dose resulting from ingestion of ^{226}Ra and ^{228}Ra due to all foods and drinking water combined in normal background areas were 6.3 and 11mSv, respectively [4]. Therefore, the estimated annual effective doses of ^{226}Ra and ^{228}Ra were about 0.0274 and 0.0775 times smaller than them, respectively. The estimated annual effective dose of ^{40}K was 5.24268 mSv/year and it comprised 0.52% of the annual dose limit of 10^3 mSv/year for the general public [18]. The estimated annual effective doses of ^{137}Cs is 2.4875×10^{-3} mSv/year and it comprised 0.00025% of the annual dose limit.

The estimated total annual effective dose received from ^{226}Ra, ^{228}Ra, ^{40}K and ^{137}Cs, due to consumption of Parsley and Leek vegetables by population of Tehran province (6.268 mSv), was 46.27 times lower than the total exposure per person resulting from ingestion of terrestrial radioisotopes (290 mSv) as proposed by UNSCEAR[4]. ^{40}K contributed the highest to the mean annual effective dose. This result is in agreement with other published studies [7, 10].

In conclusion the average ^{226}Ra and ^{228}Ra activity in these leafy vegetable samples is about 2.63 and 6.78 times the reference values, respectively. The calculated total annual effective dose received from ^{226}Ra, ^{228}Ra, ^{40}K and ^{137}Cs in this study was 46.27 times lower than the total exposure per person resulting from the ingestion of terrestrial radioisotopes. ^{40}K is the gamma emitting radionuclide that contributed the highest to the mean annual effective dose.

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