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RADIOACTIVITY LEVELS IN PLANT SAMPLES IN TULKAREM DISTRICT, PALESTINE AND ITS IMPACT ON HUMAN HEALTH

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The activity concentrations of naturally occurring radioactive materials such as ^{226}Ra, ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs were measured for 44 plant samples collected from different locations in the northwestern region of the West Bank, Palestine, using high-resolution gamma ray spectroscopy. The activity concentrations of radionuclides in the investigated plant samples ranged from 7.5 to 157.6 Bq kg^{-1} for ^{226}Ra, 7.5 to 66.1 Bq kg^{-1} for ^{238}U, 1.8 to 48.5 Bq kg^{-1} for ^{232}Th, 14.3 to 1622 Bq kg^{-1} for ^{40}K and <0.1 to 4.7 Bq kg^{-1} for ^{137}Cs. The average values of these activities were 48.3, 26.5, 10.1, 288.0 and 2.2 Bq kg^{-1}, for ^{226}Ra, ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs, respectively. The study presents the total gamma radiation dose rate assessed from natural radionuclides, ^{137}Cs and cosmic radiation, the dose rate of each radionuclide and the effective dose for all the samples. The radiological health implication to the population that may result from these doses is found to be low, except in few cases. The measurements have been taken as representing a baseline database of values of these radionuclides in the plants in the area.

INTRODUCTION

Naturally occurring radioisotopes are the main sources of both external and internal radiation exposure in human beings. Natural radioactivity is omnipresent in the earth's crust in different amounts. Rocks and soil on the earth's crust are a source of continuous exposure to human beings. The amount of radioactivity in soil was transferred in minute quantities into plants. Radionuclide uptake of plants depends upon different factors such as soil type, texture, pH, conductivity, carbonate and sulphite contents[1]. Radioactivity is common in rocks and soil[2], air[3], food and in water[4]. When ingested or inhaled, radionuclides are distributed among body organs according to the metabolism of the element involved, which normally exhibits varying sensitivities to radiation. The contents of radio elements have been measured in various food and drinking water samples by several methods, but their concentrations differ from one place to another[5]. A fundamental challenge in radiation research related to human health is to predict the biological impact of exposure to ionising radiation[6], there are now myriad experimental reports that radiation alters the normal metabolic processes of living cells and tissues[7].

Plants absorb these radionuclides from soil with some others minerals during their growth. These dangerous isotopes enter the cells and tissues of human beings with food. Most of the non-edible parts in these components are returned to the soil as organic fertiliser where they may again be utilised in the soil–plant pathway and/or are mixed with feed for livestock[8]. The critical paths of radionuclides and the critical foods in the West Bank are different from those in European and North American countries because agricultural products, food customs, chemical and physical conditions of the soil system are different[9].

An important element appearing to be significant in these properties is potassium, which is associated with sodium and regulates the muscle system function and the cardiac rhythm[10]. Living organisms need potassium in their bodies, which is incorporated through alimentation. However, ~0.0118 % of the Earth's potassium is ^{40}K radioisotope. In this way, each organic material ingested will present a little quantity of radioactivity[10].

The purpose of the present study was to measure the activity concentrations of ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs in different plant leaves and grasses grown in the Tulkarem district, Palestine, and to determine the radiological effects resulting from their consumption. Since no previous study was done in the area under investigation, the present study would therefore constitute a contribution to the establishment of a standard database of the natural radioactivity of edible plants in Palestine.

MATERIALS AND METHODS

Study area

Tulkarem (Latitude: 21°32′48.3673″, Longitude: 39°11′29.4389″) is a Palestinian governorate located in the north of the West Bank with a population of 173 000 inhabitants www.pcbs.gov.ps. The

governorate is situated on the western edge of northern West Bank, in the foothills of the Samarian mountains ~15 km north west of Nablus and 15 km east of the coastal town of Netanya (Figure 1). It is bordered by the 1948 cease-fire line, with the Centre and Haifa Districts to the west, and Qalqiliya and Ramallah Districts to the south. The Tulkarem arable land allows the city inhabitants to produce citrus fruits, melons, olives, nuts, thyme, figs, grapes, almonds, tomatoes, potatoes, green beans, guava and other products. Tulkarem district was among the first districts targeted by the Israeli Segregation Wall plan that started in 2002. So far, ~42 km^2 of fertile agricultural lands have been destroyed, confiscated or segregated by the construction of the Wall in this district. The targeted land is considered among the most fertile in the West Bank where it served as the food basket for Palestinians in Tulkarem and elsewhere. http://en.wikipedia.org/wiki/Tulkarm.

Sample collection

A total of 44 plant samples (grass and leaves of old trees) were collected between 15th September and 14th October 2011 from 12 major supply locations of the Tulkarem district (Figure 1), where different types of plants exist whose consumption is very significant (for humans and animals), directly from the land farms, owned by some natives (as listed in Table 1).

In each location, some specific plant species were collected. The leaves were collected directly from the plants when the plants were about to shed them. The samples of plant leaves and grass for each location were thoroughly mixed and air-dried for a period of 30 d and they were oven-dried at a temperature of 110°C within 24 h, and were withdrawn, then crushed and sieved using a 2-mm sieve mesh. All the samples were placed thereafter in a vertical cylindrical plastic container named Marinelli beaker, previously washed, rinsed with diluted HCl, dried, and sealed for at least 4 weeks to allow sufficient time for ^{238}U and ^{232}Th to attain a state of secular radioactive equilibrium with their corresponding progenies prior to gamma spectroscopy[5, 11, 12].

Instrumentation

To determine the natural radioactivity of the samples using gamma ray spectrometry, a high-purity germanium detector (with an efficiency of 15 % compared with the standard NaI(Tl) detector and an energy resolution of 1.85 keV at 1332.5 keV) was connected to a multichannel analyzer[13]. To reduce the background effect, the detector was shielded in a 10-cm-wall lead covering lined with 2-mm copper and 2-mm cadmium foils[13]. Gamma ray spectrometry system was calibrated using a mixture of radioactive sources: ^{139}Ce (166 keV), ^{203}Hg (279 keV), ^{113}Sn (392 keV), ^{85}Sr (514 keV), ^{137}Cs (662 keV), ^{88}Y (898 and 1836 keV) and ^{60}Co (1173 and 1332 keV) in the energy range (186–1850) keV. The calibration efficiency curve beyond 1850 keV was constructed using different energy peaks of ^{226}Ra series (^{214}Bi has peaks at 2204 and 2448 keV) in order to cover the range from 60 up to 2500 keV[14]. The standard source packed in the Marinelli beaker had the same geometry as that used for measured samples.

Spectroscopic analysis

The quality assurance of the measurements was carried out daily for energy calibration and gathering sample measurement. A counting time of 70 000 s was used to obtain the gamma spectrum of the samples. The concentrations of radium, uranium, thorium, potassium and caesium was measured for each plant. The gamma ray transitions of energies of 186 keV (^{226}Ra), 295.2 and 351.9 keV (^{214}Pb), 609.3, 1120.3, 1764 and 2204 keV (^{214}Bi) were used to determine the concentration of the ^{238}U series. The gamma ray transitions of energies of 338.4 keV (^{228}Ac), 583.3 keV (^{208}Tl), 2614 keV (^{208}Tl) and 911.1 keV (^{228}Ac) were used to determine the concentration of the ^{232}Th series. The 1460 and 662 keV gamma ray transitions were used to determine the concentrations of ^{40}K and ^{137}Cs, respectively, in the samples[8].

RESULTS AND DISCUSSION

The activity concentration, *A*

The activity concentrations ($Bq\,kg^{-1}$)of natural and artificial radionuclides in different plant samples was calculated using the following equation[1]:

$$A = \frac{(c/s)_{\text{net}}}{I \times \varepsilon_{ff} \times M} \quad (1)$$

where $(c/s)_{\text{net}}$ is the number of counts per second $[(\text{cps})_{\text{sample}}-(\text{cps})_{\text{Background}}]$, I is the intensity of the gamma line in a radionuclide, ε_{ff} is the measured efficiency for each observed gamma line and M is the mass of the sample in kilograms[13, 15]. The activity concentrations of ^{226}Ra, ^{238}U, ^{232}Th and ^{40}K for different plant and grass samples are listed in Table 1. From the table, the activity concentration of ^{226}Ra is in the range 7.5–157.6 Bq kg^{-1} with an average value of 48.3 Bq kg^{-1}, while for ^{238}U, the maximum value is 66.1 Bq kg^{-1} and the minimum value is 7.5 Bq kg^{-1} with an average value of 26.5 Bq kg^{-1}, for ^{232}Th, the activity ranges from 1.8 to 48.5 Bq kg^{-1} with an average value of

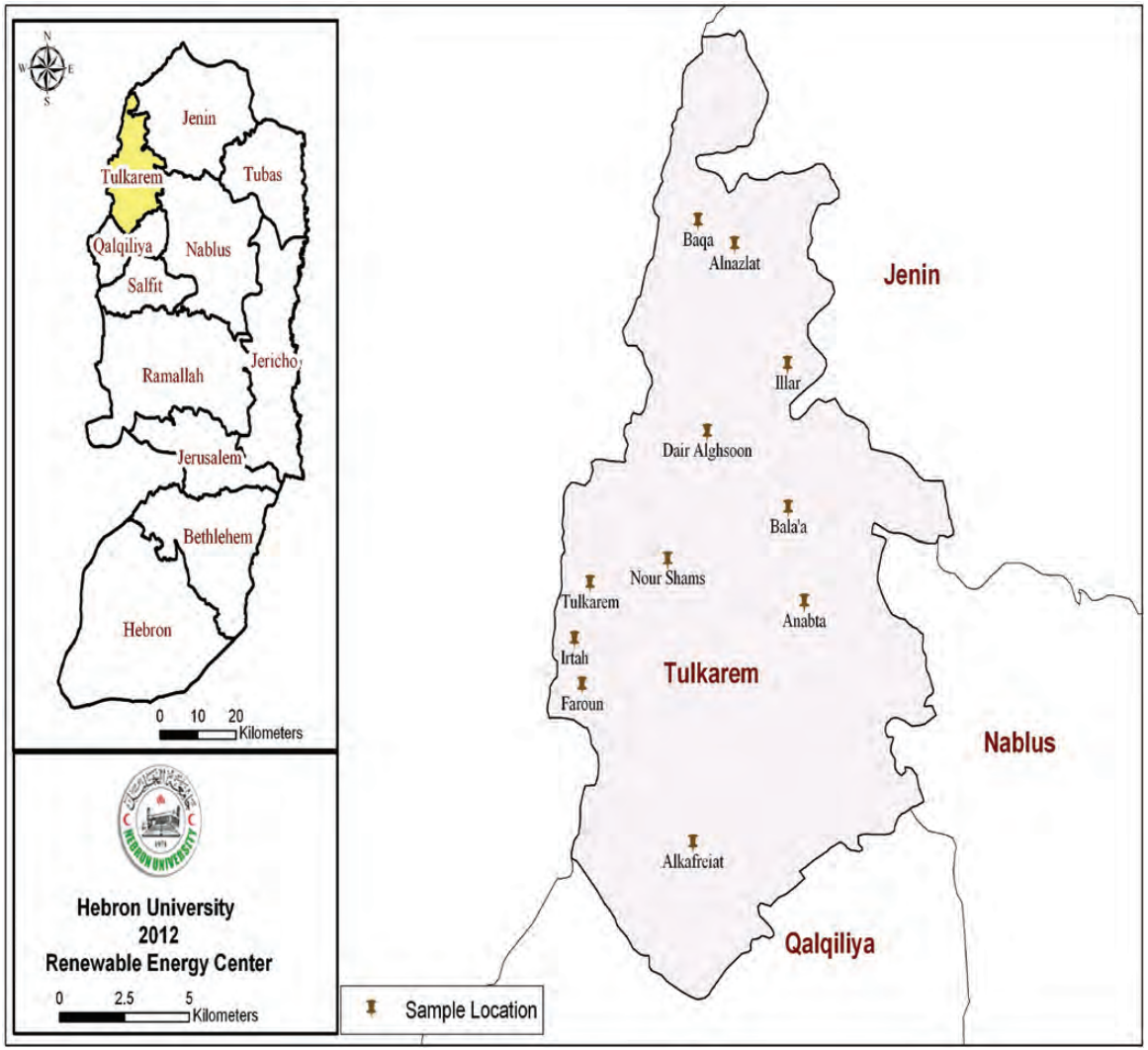

Figure 1. West Bank geographical map and sample location of the Tulkarem district.

Table 1. The activity concentrations of radionuclides in plant leaves and grass samples collected from the Tulkarem district, the West Bank, Palestine.

Zone	Sample code	Kind of samples	No. of samples	^{226}Ra (Bq kg^{-1})	^{238}U (Bq kg^{-1})	^{232}Th (Bq kg^{-1})	^{40}K (Bq kg^{-1})
Tulkarem (City)	TSS-1	Almond	2	62.8	24.8	3.6	140.0
	TSS-2	Olive	2	60.1	27.0	9.8	153.0
Nour-Shams	TSS-3	Olive	2	15.6	8.1	3.5	104.0
	TSS-4	Fig	2	81.4	38.1	6.6	15.0
DairAlghsoon	TSS-5	Olive	2	28.1	14.7	4.0	61.0
	TSS-6	Grapes	2	29.2	13.0	5.3	139.0
Illar	TSS-7	Citrus	2	7.5	7.8	14.8	61.4
	TSS-8	Nut	2	87.7	34.3	3.2	51.0
	TSS-9	Olive	2	30.1	15.1	6.2	14.3
Irtah	TSS-10	Grass	2	48.6	38.8	48.5	309.0
	TSS-11	Thyme	2	55.6	24.2	9.0	1622
Bala'a	TSS-12	Olive	2	12.0	7.5	4.2	15.0
Chemical factory	TSS-13	Grass	2	18.0	19.8	20.9	140.3
Baqa	TSS-14	Grass	2	13.0	14.2	8.4	93.6
	TSS-15	Thyme	2	85.2	41.4	9.3	1413
Anabta	TSS-16	Olive	2	21.0	11.5	5.9	31.0
	TSS-17	Thyme	2	63.5	26.3	1.8	1376
Alkafreiat	TSS-18	Grass	2	155.2	59.6	3.5	61.0
	TSS-19	Grass	2	25.0	17.1	12.0	87.0
Faroun	TSS-20	Grass	2	55.6	25.5	5.9	111.5
	TSS-21	Grass	2	157.6	66.1	31.6	244.0
Alnazlat	TSS-22	Grass	2	129.0	47.3	4.2	84.0
	Range	—	—	7.5–157.6	7.5–66.1	1.8–48.5	14.3–1622
	Average	—	—	48.3	26.5	10.1	288.0

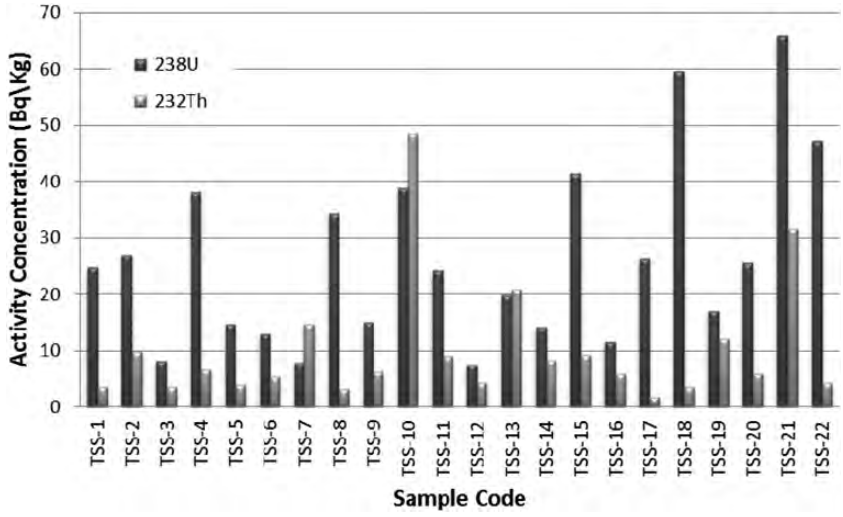

Figure 2. Comparison between the ^{238}U and ^{232}Th radionuclides for plant samples in different locations in the Tulkarem district.

10.1 Bq kg^{-1}; and for ^{40}K, ranges from 14.3 to 1622 Bq kg^{-1} with an average value of 288.0 Bq kg^{-1}. From these results, it has been shown that, the grass samples generally have higher concentration levels for ^{226}Ra, ^{238}U and ^{232}Th radionuclides, than other plant samples. It has also been shown that for ^{40}K,

the activity concentration is high in some samples (Thyme samples)and low in others (Olive and Fig samples).

Figure 2 shows a comparison between ^{238}U and ^{232}Th for plant samples in different locations in the Tulkarem district. The presence of each radionuclide in the plant and the variation in concentration are dependent on the level of these radionuclides itself in the soil, and this reveals the nature of the soil presented in the study area. The variation observed in the concentrations ^{226}Ra, ^{238}U and ^{40}K may also be attributed to the use of fertilisers in some of the cultivated lands from which plant samples were collected and the presence of radionuclides in the soil in which the plant was grown.^{40}K is an essential mineral nutrient needed for the growth of the plant and is transported together with water from the roots to the leaves through the stem.

The ranges of concentrations of ^{226}Ra and ^{238}U were nearly similar, while ^{232}Th exhibited the lowest values and ^{40}K showed the highest values. These findings may be attributed to the genetic differences, i.e. certain plant species have the ability to accumulate uniquely high concentrations of particular elements and this phenomenon can be explained on the basis that there is wide variability in the bioaccumulation of trace elements among plant species[16]. In general, the highest radioactivity concentration in plants was found in those collected in the area with the highest radioactive concentration in the soil substrate and the lowest in those with the lowest concentration in the substrate[17].

Table 2. Activity concentrations of ^{137}Cs and total dose rate of natural radionuclide's, ^{137}Cs and cosmic radiation in plant leaves and grass samples collected from the Tulkarem district, the West Bank, Palestine.

Zone	No. of samples	^{137}Cs (Bq kg^{-1}) range	D_γ (nGy h^{-1}) range
Tulkarem (City)	4	ND–0.1	53.0–58.6
Nour-Shams	4	0.2–1.9	44.2–55.3
DairAlghsoon	4	1.9–2.2	45.6–49.1
Illar	6	ND–1.6	45.2–53.0
Irtah	4	ND–4.7	96.1–120.0
Bala'a	2	ND–1.1	38.2–43.7
Chemical factory	2	ND–1.1	57.2–66.7
Baqa	4	ND– 4.0	49.8–118.6
Anabta	4	ND–0.6	44.2–105.6
Alkafreiat	4	4.0–4.6	55.3–71.1
Faroun	4	ND–3.0	53.6–93.7
Alnazlat	2	1.1–1.4	55.4–66.6
Total no.	**44**	—	—
Total range		<0.1–4.7	38.2–120.0
Average		2.2	64.8

ND, not detected.

Calculation of radiological effects

The total gamma radiation dose rate, D_γ

The total gamma radiation dose rate is modified to include the contributions from natural radionuclides,^{137} Cs and cosmic radiation according to the following equation[9]:

$$D_\gamma(\text{nGy h}^{-1}) = 0.427C_U + 0.662C_{Th} + 0.043C_K + 0.03C_{Cs} + 34 \quad (2)$$

where C_U, C_{Th}, C_k and C_{Cs} are the activity concentrations of ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs radionuclides, respectively in Bq kg^{-1}. The number 34 is a factor included to ensure that the effects of cosmic rays are implemented.

The activity concentrations of ^{137}Cs and the total gamma radiation dose rates of natural radionuclides (^{238}U, ^{232}Th and ^{40}K), ^{137}Cs and cosmic radiation are presented in Table 2. As it can be seen from the table, the activity concentrations are found to range from <0.1 to 4.7 Bq kg^{-1} with an overall average value of 2.2 Bq kg^{-1} and the total gamma radiation dose rates range from 38.2 to 120.0 nGy h^{-1} with an overall average value of 64.8 nGy h^{-1}. It is clear that some places in the area under investigation lie in a higher radiation background compared with the worlds' average (see Table 3). The slightly higher value of the dose in the plant samples may be due to the use of fertilisers for cultivation and fall out ^{137}Cs from the atmosphere. The lesser values and the absence of fallout in some samples may be due to erosion of ^{137}Cs by rain the study area[18].

Radioactive ^{137}Cs, has a half-life of 30 y. These can stay in the environment for many years and could continue to have long-term harmful effects on food products and hence a threat to human health. If ^{137}Cs enters the body, it is distributed fairly uniformly throughout the body's soft tissues, resulting in exposure of those tissues. Compared with some other radionuclides, ^{137}Cs remains in the body for a relatively short time. Like all radionuclides, exposure to radiation from ^{137}Cs results in an increased risk of cancer[19].

By comparing the activity concentration for plant samples with those in different locations in the world as shown in Table 3, it is clear that, the activity levels for ^{238}U, ^{232}Th and ^{40}K for the area under investigation were higher than the activity levels in most of the other countries. According to the UNSCAR report (2000)[20], the world average values of activity concentration for ^{238}U, ^{232}Th and ^{40}K are 20, 15 and 350 Bq kg^{-1}, respectively. The measured values of the activity concentration for ^{238}U and ^{40}K radionuclides for some samples for the area under investigation were larger than the world

Table 3. Comparison of radioactivity concentration levels in the plant and grass samples of the Tulkarem region, Palestine with those from other locations and the world average.

Country	^{238}U (Bq kg^{-1})	^{232}Th (Bq kg^{-1})	^{40}K (Bq kg^{-1})	^{137}Cs (Bq kg^{-1})	Reference
Egypt	2.5–9.8	0.6–7.4	46.7–943.0	0.1–2.1	(22)
	5.5	3.6	207.3	—	
India	9.6–11.3	14.3–18.4	27.0–206.4	—	(23)
Taiwan	—	—	—	0.1–1.2	(24)
Brazil	—	<11–43.0	666.0–1216.0	<2.8–3.1	(10)
Nigeria	11.9–98.3	24.4–38.9	49.7–83.0	ND	(25)
	25.7	33.4	64.5		
Hebron-Palestine	17.5–134.1	4.5–66.8	160.8–2339.2	0.85	(9)
	66.2	36.6	1588.2		
World average	20	15	350	—	(20)
Tulkarem-Palestine	7.5–66.1	1.8–48.5	14.3–1622.0	<0.1–4.7	Present work
	26.5	10.1	288.0	—	

Table 4. The gamma absorbed dose rates (D_r), the effective dose (H_E) and the ELCR in plant leaves and grass samples collected from the Tulkarem district, the West Bank, Palestine.

	^{238}U	^{232}Th	^{40}K	Total	
				Range	Average
D_r (nGy h^{-1})				5.2–127.4	30.3
Range	3.5–30.5	1.1–29.3	0.6–67.6		
Average	12.1	6.1	12.0		
H_E (μSv y^{-1})				6.4–156.1	37.2
Range	4.3–37.4	1.4–35.9	0.7–82.8		
Average	15.0	7.5	14.7		
ELCR ($\times 10^3$)				0.03–0.55	0.13
Range	—	—	—		
Average	—	—	—		

average value and for most samples, ^{232}Th is also lying within the world averages. The use of fertilisers in large amounts has affected radionuclide concentration, especially potassium-containing fertilisers is one of the causes of the presence of high activity of ^{40}K in some samples[21].

Dose rate and effective dose rate, D_r

The decay of naturally occurring radionuclides in soil and plants produces radiation to which human beings are exposed. Outdoor exposure arise from terrestrial radionuclides present at trace levels in all plant types. The external gamma dose rate in air is calculated from measurement of concentrations of the relevant radionuclides in plants. For uniformly distributed radionuclides, dose rate can be calculated using the following relation[18]:

$$D_r = A_C \times C_F \quad (3)$$

where D_r is the dose rate in nGy h^{-1}, A_C is the activity concentration in Bq kg^{-1} and C_F is the dose conversion factor in nGy h^{-1} per Bq kg^{-1} (absorbed dose rate in air per unit of activity concentration). The dose conversion factors (0.462 for ^{238}U, 0.604 for ^{232}Th and 0.0417 for ^{40}K) were taken from the UNSEAR (2000) report, which are based on the Monte Carlo technique.

For an adult, the dose rates were converted into effective dose rates using the following relation[20]:

$$H_E = D_\gamma \times T \times F \quad (4)$$

where H_E is the effective dose (in μSv y^{-1}), D_γ is the estimated dose rate (in nGy h^{-1}), T is the outdoor occupancy time factor ($0.2 \times 24 \times 365.25$ d $\approx$ 1750 h y^{-1}) and F is the absorbed-to-effective dose conversion factor (0.7×10^{-3} μSv per nGy).

The gamma absorbed dose rates in air and the outdoor effective dose rates were estimated from the concentrations of each of the nuclides of ^{238}U and ^{232}Th series, and of ^{40}K using Equations (3) and (4), and the individual results were added in order to obtain the total absorbed dose rate in air and

effective dose rates due to gamma radiation from those radionuclides. The results of these calculations are listed in Table 4.

The estimated dose rates vary from 3.5 to 30.5 nGy h^{-1} with an average value of 12.2 nGy h^{-1} for ^{238}U, 1.1–29.3 nGy h^{-1} with an average value of 6.1 nGy h^{-1}, for ^{232}Th and 0.6–67.6 nGy h^{-1} with an average value of 12.0 nGy h^{-1}, for ^{40}K radionuclide. The total dose rate for the three radionuclides vary from 5.2 to 127.4 nGy h^{-1} with an average value of 30.3 nGy h^{-1}. The total values of the dose rates estimated for some plant samples in the study area were higher than that of the UNSCEAR (2000) values, and others were lower than that of the UNSCEAR (2000) values.

In the area under investigation, the total H_E ranges between 6.4 and 156.1 μSv y^{-1} with an average value of 37.2 μSv y^{-1}. For children and infants, the values are ~10 and 30 % higher, in direct proportion to an increase in the value of the conversion coefficient from absorbed dose in air to effective dose[20]. The values of the dose rates estimated for some plant samples in the study were higher than that of the UNSCEAR values and the average value was less than that of the UNSCEAR values.

Excess lifetime cancer risk

According to the Ministry of Health, Palestine (MOH), 1350 new cancer cases were reported in the West Bank in 2010. The Tulkarem district ranked second in Palestine with 134 new cases with an incidence rate of 80.8 per 100 000 population. Furthermore, the figures reported by MOH show a slight increase in cancer mortality in the West Bank in 2010 compared with 2007 from 10.3 to 10.8 % from the total deaths in the west bank[26].

The excess lifetime cancer risk (ELCR) is calculated using the following equation[27] and is shown in Table 4.

$$\mathrm{ELCR} = H_E \times \mathrm{DL} \times \mathrm{RF} \quad (5)$$

where H_E, DL and RF are the annual effective dose equivalent, duration of life (70 y) and risk factor (Sv^{-1}), fatal cancer risk per Sievert. For stochastic effects, ICRP 60 uses a value of 0.05 for the public[28]. The range of ELCR is 0.022×10^{-3} to 0.546×10^{-3} with a total average of 0.131×10^{-3}.

The ELCR values for some samples are higher than the world average and the total average value is less than the world average (0.29×10^{-3})[28]. Therefore, on average the cancer risk is low.

CONCLUSIONS

The concentration of natural radioactivity and the associated effective dose levels in plant leaves and grass samples were calculated for 12 different locations in the Tulkarem district, Palestine. The average concentrations of ^{226}Ra, ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs were 48.3, 26.5, 10.1, 288.0 and 2.2 Bq kg^{-1}, respectively. The calculated total gamma radiation dose rate for natural radionuclides, ^{137}Cs and cosmic radiation, the dose rate in each natural radionuclide and the effective dose in all samples are included. The total averages for these parameters were 64.8 nGy h^{-1}, 30.3 nGy h^{-1} and 37.2 μSv y^{-1}, respectively. The radiological health implication to the population that may result from these values is found to be low, except in some cases. The measurements taken in this study represent a baseline database of activity levels that can serve as a reference point for future studies.

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