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Radiological Study on Soils, Foodstuff and Fertilizers in the Alexandria Region, Egypt

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Abstract

Radionuclides ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs were measured in foodstuffs (vegetables, fruits, seeds, cereals, meats, and milks), soil, and agricultural chemical fertilizers (phosphate and urea) using a high-resolution gamma ray spectrometer. The gamma Dose Rate Conversion Factors (DRCFs) for the radionuclides present in the surface layer of the investigated soil were developed based on the local soil conditions and the updated nuclear properties of radionuclides. Good agreement between the calculated DRCFs and those in the literature was found. The annual external and internal effective dose rates from soil and food were deduced. The annual average of the external effective dose rate was 195.28 μSv. The daily intake of radionuclides from food consumption reveals that vegetables are the biggest contributors, while meat is the lowest. The annual internal effective dose rates were 61.32 μnd 146.54 μSv for ^{226}Ra and ^{40}K, respectively. The levels of radionuclides in phosphate and urea fertilizers frequently applied to the agricultural soil of the study area indicated that the highest levels of ^{226}Ra and ^{232}Th were in super-phosphate and the lowest in urea. Finally, the transfer of radionuclides from soil to plants was discussed.

Key words: Effective dose, Fertilizer, Foodstuff, Radionuclide, Soil.

Introduction

Studies on radiation levels and radionuclide distribution in the environment provide vital radiological baseline information. Such information is essential in understanding human exposure from natural and man-made sources of radiation and necessary in establishing rules and regulations relating to radiation protection (Quindos et al., 1994).

There are many sources of radiation and radioactivity in the environment. The earth and atmosphere contain varied levels from naturally radionuclides such as ^{238}U and ^{232}Th decay chains as well as singly occurring types such as ^{40}K. Soil features, geological formations, and human activities related to radiation and radioactivity are important factors enhancing the background levels of natural radiation (Colmerero Sujo et al., 2004). The continuity in increasing of these radionuclides in the environment may be attributed to several factors such as the successive utilization of phosphate fertilizer, burning of fossil fuels (crude oil and coal), mining and milling operations, and building materials. Ingesting and inhaling such levels of radionuclides contribute significantly to the radiation dose that people receive (Martínez, 1989). In addition, exposure externally to enhanced levels of radiation can elevate the health hazard risk.

Several studies presented different methods to calculate the gamma radiation doses at 1 m from the ground surface from the levels of ^{238}U and ^{232}Th decay chains and ^{40}K in surface soil. The predictions of radiation dose using these methods are in close agreement with those obtained from experimental measurements (Quindós et al., 1990; Ortega and Rosell, 1991). Radionuclide Transfer Ratio (TR) is considered an important factor determining the uptake of radionuclides by plants. TR is defined as the ratio of radionuclide level in a plant to that in its soil. This ratio depends on the physical and chemical properties of soil, climatic conditions, and plant types (Narayana et al., 1995).

The objectives of the present study are: 1) To perform measurements on radioactivity in soils, foodstuffs, and fertilizers in the Alexandria region. 2) To develop external Dose Rate Conversion Factors (DRCFs) for assessing the external radiation exposure at 1 m above the ground based on the local soil properties and the updated nuclear properties of radionuclides. 3) To assess the ingested ^{226}Ra and ^{40}K via food consumption. This study is considered a part of the radiological baseline information of the Alexandria region.

Experimental Work

In the present study, 41 different samples from soil, foods, and fertilizers were investigated. Soil samples were taken from 10 different sites on the western side of the Nile delta of Egypt as shown in Figure 1. The

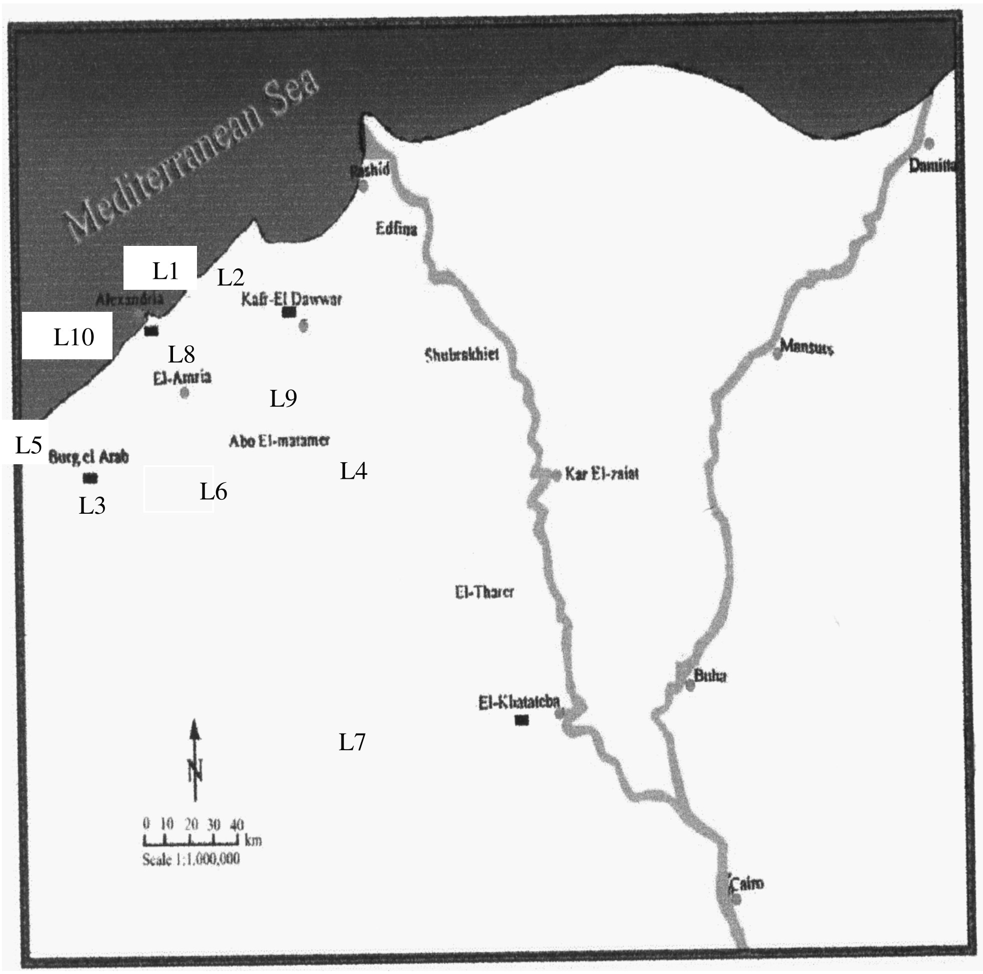

Figure 1. The locations of investigated soil samples.

land use of this area is mainly agricultural and residential. The soil texture varied gradually from approximately pure clay near the Nile River to approximately pure sand adjacent to the western desert. The fertilizer samples were selected from the types that are frequently applied in the study area. Soil and fertilizer samples were prepared for gamma ray spectroscopic analysis according to a method mentioned in Colmenero Sujo et al. (2004). Large stones and other objects were removed from the sample in the field. Two liters of soil were then taken to the laboratory, where it was powdered, sieved through a 2 mm mesh and dried in a vacuum drier at 101 °C, and finally was packed into Marinelli beakers and sealed. Then it was allowed to stand for at least 4 weeks so that the ^{226}Ra series was able to reach radioactive equilibrium.

Food samples were washed, peeled when necessary, and dried in air. After that, they were oven-dried at 80 °C for approximately 16 h (Santos et al., 2002). The prepared samples were weighed and transferred to 1000-ml Marinelli beakers (Alberto et al., 1996). The beakers were sealed and allowed to stand for at least 4 weeks so that the ^{226}Ra series was able to reach radioactive equilibrium.

The prepared samples were measured by gamma spectrometer system. The measuring system consists of a p-type coaxial HPGe with an efficiency of 24.5% and a resolution of 1.7 keV at 1.33 MeV. The gamma spectrum was recorded using a PC based 4096 channel analyzer and processed using MCA-Super software.

The activity concentration of ^{40}K was determined using the 1460 keV gamma line. The lines 186.2, 295.2, 351.9, 609.3, and 1120.3 keV were used for ^{226}Ra activity determination. The lines 338.4, 911, and 583.1 keV were used to determine the activity of ^{232}Th. The activity concentration of ^{173}Cs was determined at 661.7 keV (IAEA, 1989). The spectrometer was calibrated for energy using a set of certified standard sources (^{137}Cs, ^{60}Co, ^{57}Co, and ^{241} Am). The absolute detection efficiencies were calibrated using certified standard source (^{152}Eu) and reference materials prepared in geometrical shape and composition to simulate the investigated samples' matrix (El-Tahawy et al., 1992). The measurements were carried out in the radiation laboratory of the Department of Environmental Studies, Institute of Graduate Studies and Research, Alexandria University. The detector was shielded using a cylindrical lead castle of 0.1 m thickness with an internal wall made of copper. For quality control requirements, reference soil samples (IAEA-326/327, 1996 Soil) and a cockle flesh sample (IAEA-134, 1993 Cockle flesh) were analyzed during the measurements.

Results and Discussion

Radionuclides in soil

The radionuclides detected in soil were ^{226}Ra, ^{232}Th, and ^{40}K in addition to ^{137}Cs, and their levels are shown in Table 1. The average concentrations were 16.43 ± 2.89, 18.31 ± 5.25, and 268.18 ± 81.65 Bq kg^{-1} for ^{226}Ra, ^{232}Th, and ^{40}K, respectively. ^{137}Cs was detected only in 3 locations. The maximum level of 7.24 Bq kg^{-1} was observed in the sample collected from location number 7. The averages of the detected levels were compared with those listed in Table 2. It is clear that the levels of radionuclides in the present study are much closer to those of the soil of Middle and Upper Egypt (Ibrahim et al., 1993) and lower than the world averages reported in UNSCER (1988). The variation among the levels in soils of different countries may be attributed to the wide variations in geological formations of different types of soil.

The gamma absorbed dose rate in air

In this part the absorbed Dose Rate Conversion Factors (DRCFs) R_{Ra},R_{Th}, and $R_k$for ^{238}U and ^{232}Th decay chains and ^{40}K were deduced in (nGy h^{-1} per Bq kg^{-1}). The obtained factors and the concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in the surface layer of soil were used to calculate the gamma radiation dose rate at a height of 1 m above the ground. Equation (1) was used for these calculations after the DRCFs had been deduced.

$$D = R_{Ra}C_{Ra} + R_{Th}C_{Th} + R_K C_K \qquad (1)$$

where C$_{Ra}$, C$_{Th}$, and C$_k$are the concentrations in Bq kg^{-1} of ^{226}Ra, ^{232}Th, and ^{40}K, respectively.

The calculations of these factors were carried out based on the data of DRCFs published in Kocher and Sjoreen (1985) for mono-energetic sources (in the range between 0.01 and 3 MeV). A quadratic regression for the energy range between 0.01 and 0.1 MeV and a linear regression between 0.1 and 3 MeV were fitted and Eqs. (1) and (2) were obtained.

Table 1. The levels of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in dry soil (Bq kg^{-1}).

Location	^{226}Ra	^{232}Th	^{40}K	^{137}Cs
L1	23.94 ± 0.71	9.84 ± 0.03	175.88 ± 5.55	ND
L2	16.8 ± 0.64	15.97 ± 0.84	242.67 ± 6.45	ND
L3	15.14 ± 0.76	15.62 ± 0.78	344.02 ± 9.10	ND
L4	16.65 ± 0.73	17.02 ± 0.86	218.93 ± 7.35	ND
L5	17.45 ± 0.79	26.79 ± 1.21	382.55 ± 9.73	ND
L6	17.26 ± 0.76	26.5 ± 1.19	378.48 ± 9.63	ND
L7	15.38 ± 0.61	17.84 ± 0.74	244.72 ± 6.74	7.24 ± 0.52
L8	14.82 ± 1.13	20.08 ± 0.05	315.4 ± 8.75	ND
L9	13.74 ± 1.24	14.07 ± 0.66	156.00 ± 4.72	0.73 ± 0.23
L10	16.38 ± 0.61	19.39 ± 0.74	222.91 ± 6.04	1.77 ± 0.23

ND: Not detected: detection limit was about 0.5 Bq kg^{-1} based on the 95% confidence

Table 2. Comparison of the average concentrations with those published data in soil.

Radionuclides concentration (Bq kg^{-1})				
^{226}Ra	^{232}Th	^{40}K	Region	Reference
16.43	18.31	268.16	Present work	
77.10	146.00	370.00	Hong Kong	Yu et al., 1992
16.60	18.10	316.00	Egypt	Ibrahim et., 1993
38.80	41.00	653.00	Spain	Baeza et al., 1994
29.20	47.80	704.00	Brazil	Alberto et al., 1996
114.60	43.20	274.60	India	Selvolseka et al., 1999
25.00	25.00	370.00	World average	UNSCEAR, 1988

$$DRCF = 0.0073\mathrm{X}E^2 - 9X10^{-5}E + 6\mathrm{X}10^{-7} \text{ for}$$

$$0.01MeV \leq E \leq 0.1MeV, \qquad (2)$$

$$DRCF = 1.73\mathrm{X}10^{-3}E - 6\mathrm{X}10^{-5} \text{ for}$$

$$0.1MeV\mathrm{gg} \leq \mathrm{g}MeV. \qquad (3)$$

The correlation coefficients were calculated to be 0.998 and 0.999 for relations 2 and 3, respectively.

Using Eqs. (2) and (3), assuming the existence of secular equilibrium in the radioactive decay chains, and taking into account all gamma lines and X-rays emitting by radionuclides, the DRCFs were calculated for all radionuclides that are members in ^{238}U and ^{232}Th decay chains as well as ^{40}K.

The gamma DRCF for a particular radionuclide is given by

$$DRCF = \sum_i f_i DRCF_i(E_i) \qquad (4)$$

where f_i and $DRCF_i$ are the branching ratio and the gamma dose rate conversion factor for the line of energy E_i, respectively.

The following assumptions were considered in the calculations: uniform distribution of radionuclides in a slab parallel to the ground surface to a depth of 1.20 m and homogeneous dry soil of average density (1.4 g cm^{-3}) in the study area, despite the fact that only the top 20 cm had been verified experimentally. This is because the mineralogical composition of the soil does not change significantly with depth; therefore, the concentrations remain approximately constant with depth.

The comparison between the results obtained and those listed in Table 3 revealed good agreement.

Table 3. Comparison of DRCFs with those published data (pGy h^{-1} per Bq kg^{-1}).

Radionuclide		DRCFs		
	Present work	UNSCEAR, 1988	UNSCEAR, 1993	Barisic, 1996
^{40}K	42.04	43.00	41.4.00	42.86
^{238}U	495.00	427.00	461.00	475.78
^{232}Th	669.30	662.00	623.00	665.44

The external dose rate at 1 m from the ground surface from ^{238}U and ^{232}Th decay chains in addition to ^{40}K as obtained from Eq. (1) ranged from 22.78 to 42.65 nGy h^{-1} with an average of 31.82 ± 6.40 nGy h^{-1}. The annual effective dose was calculated using the conversion factor 6.13 (μSv/year)(nGy/hour)$^{-1}$ as recommended by UNSCEAR (1988). The values obtained ranged from 139.64 to 261.44 μSv with an average value of 195.06 ± 39.23 μSv. It is clear that the results of the present study are much lower than the world average of the terrestrial background source, which is 410 μSv (UNSCEAR, 1988).

Radionuclides in vegetables

Thirteen types of vegetables were investigated for ^{226}Ra and ^{40}K and the results are listed in Table 4. It was found that the maximum level of ^{226}Ra was 2.11 ± 0.01 Bq kg^{-1} detected in garden rocket, while the minimum level of <0.32 was detected in carrot, cucumber, green haricot, green bean, and spinach. ^{40}K was detected in all samples with reasonable activity levels. The levels ranged from 42.38 ± 3.93 Bq kg^{-1} in carrot to 217.10 ± 22.57 Bq kg^{-1} in spinach. It is well known that the existence of potassium in all living things with different levels leads to a wide variety of ^{40}K radioisotope levels in plants.

Radionuclides in fruits

Seven types of fruit were investigated for the levels of ^{226}Ra and ^{40}K and the results are presented in Table 5. ^{226}Ra was detected only in 4 samples. The highest concentration was 1.25 ± 0.35 Bq kg^{-1}, recorded in apple, and the lowest was < 0.32 Bq kg^{-1}, detected in cantaloupe, guava, and mango. The levels of ^{40}K were distributed from the lowest level of 7.34 ± 0.41 Bq kg^{-1}, in mango, to the highest level of 78.19 ± 4.33 Bq kg^{-1}, which was observed in cantaloupe.

Radionuclides in nuts, cereals, milk and meat

The levels of radionuclides in nuts, cereals, milks, and meats are listed in Table 6. The highest level of ^{226}Ra was detected at 1.31 ± 0.8 Bq kg^{-1}, in coconut, while the lowest level was below the detection limit (0.32 Bq kg^{-1}) in raisins, stewed beans, sprouted beans, lentils, and rice. ^{40}K was detected in all samples. The levels detected in milk showed wide variation between the 2 investigated types. ^{226}Ra was detected only in the powdered milk with the commercial name Nido, with a level of 0.44 ± 0.23 Bq kg^{-1}. ^{40}K levels were 222.11 ± 7.94 and 47.25 ± 3.22 Bq kg^{-1} in powdered milk with the commercial name Miro and Nido milk, respectively.

Table 4. The levels of ^{226}Ra, and ^{40}K in fresh vegetables (Bq kg^{-1}).

Type of sample	^{226}Ra	^{40}K
Carrot	* < 0.32	42.38 ± 3.93
Cucumber	< 0.32	68.55 ± 4.34
Garden rocket	2.11 ± 0.01	181.03 ± 21.21
Green haricot	< 0.32	79.44 ± 4.72
Green bean	< 0.32	67.11 ± 4.72
Mallow	0.63 ± 0.17	197.00 ± 3.16
Lettuce	1.05 ± 0.48	166.00 ± 16.69
Green okra	0.86 ± 0.05	117.98 ± 7.32
Potato	0.80 ± 0.49	118.75 ± 2.34
Spinach	< 0.32	217.10 ± 22.57
Squash	0.62 ± 0.25	87.66 ± 9.10
Tomato	0.96 ± 0.30	49.48 ± 3.45

* < 0.32 is the detection limit based on 95% confidence intervals

Internal dose from ingested foods

The annual intakes (Q) for ^{226}Ra and ^{40}K were determined in foodstuffs by using the activity levels (C) in foods and the annual food consumption rates (F) by the Egyptian population according to the Food

Balance Sheet of Egypt (2002). The calculated annual intakes and the Internal Dose Conversion Factors (*IDCF*) of 0.28 and 6.2 × $10^{-3}\mu$Sv Bq^{-1} of ^{226}Ra and ^{40}K, respectively, were used to estimate the annual internal effective dose (H) (IAEA, 1998), using the following equations:

$$Q = C \times F \tag{5}$$

$$H = Q \times IDCF \tag{6}$$

Table 5. The levels of ^{226}Ra and ^{40}K in fresh fruits (Bq kg^{-1}).

Type of sample	^{226}Ra	^{40}K
Apple	1.25 ± 0.35	32.27 ± 2.70
Cantaloupe	< 0.32	78.19 ± 4.33
Fresh fig	0.52 ± 0.27	50.93 ± 3.40
Grapes	0.33 ± 0.18	58.50 ± 3.71
Guava	< 0.32	26.38 ± 1.59
Mango	< 0.32	7.34 ± 0.41
Water-melon	0.85 ± 0.32	40.37 ± 3.07

Table 6. The activity concentrations of ^{226}Ra, and ^{40}K in nuts, cereals, milk and meat (Bq kg^{-1}).

Samples	^{226}Ra	^{40}K
Coconut	1.31 ± 0.8	73.89 ± 3.97
Raisins	< 0.32	281.35 ± 10.89
Stewed bean	< 0.32	188.64 ± 7.93
Sprouted bean	< 0.32	230.46 ± 8.22
Flour	0.68 ± 0.12	23.90 ± 1.33
Lentil	< 0.32	200.67 ± 7.51
Macaroni	0.9 ± 0.33	40.10 ± 3.05
Rice	< 0.32	13.97 ± 2.18
Miro Milk	ND	222.11 ± 7.94
Nido Milk	0.44±0.23	47.25 ± 3.22
Fresh Meat	0.96 ± 0.35	89.07±3.95
Dry Meat	1.87 ± 0.21	200.86 ± 0.67

The results are listed in Table 7. Vegetables are the highest contributors to the annual effective dose for both ^{226}Ra and ^{40}K and this may be attributed to their concentrations in vegetables. Furthermore, vegetables record the highest consumption rate with respect to the other investigated foods, as shown in Figure 2. The contribution percentages of the individual food items to the annual internal effective dose from ^{226}Ra were 48, 22, 12, 12, 5, and 1 from vegetables, cereals, fruits, milks, fishes, and meats, respectively, while for ^{40}K the percentages were 52, 20, 11, 10, 5, and 1 from the tested vegetables, cereals, fruits, milks, fishes, and meats, respectively. It is clear that the contribution percentages of ^{226}Ra and ^{40}K have the same ordering for all foodstuffs. The total annual effective doses from ingested foods were 61.32 and 146.54 μSv for ^{226}Ra and ^{40}K, respectively. These levels were lower than levels of the normal background areas reported by UNSCEAR (1988).

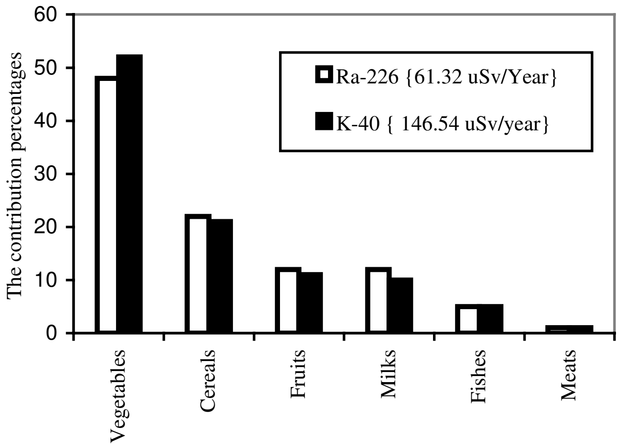

Figure 2. The contribution percentages of the individual food items to the annual internal effective doses from Ra-226 and K-40.

Radionuclides in fertilizers

Four types of the most frequently utilized agricultural fertilizers were investigated. Table 8 shows that the highest levels of ^{226}Ra and ^{232}Th are detected in super-phosphate. This finding might be attributed to the existence of high levels of uranium and thorium in phosphate ore, which is utilized in phosphate manufacturing.

Transfer ratio (TR)

TR for each radionuclide was calculated as mentioned before. The results obtained are listed in Table 9. The average values of TR for ^{226}Ra in vegetables and fruits are 0.066 and 0.013, respectively, and they are in good agreement with those reported by Antonio et al. (1988). It was also observed that the transfer in vegetables is much greater than that in fruits, which may indicate why vegetables have higher concentrations than fruits for the detected radionuclides.

Table 7. The estimated daily intake and annual internal effective dose from ^{226}Ra and ^{40}K.

Type of food	Consumption rate 10^{-3}(kg d^{-1})	Daily intake (Bq d^{-1})		Effective dose (μSv y^{-1})	
		^{226}Ra	^{40}K	^{226}Ra	^{40}K
Carrot	14.50	-	0.61	-	1.38
Cucumber	14.50	-	1.00	-	2.26
Garden rocket	13.70	0.03	2.48	3.07	5.62
Green haricot	6.00	-	0.48	-	1.09
Green beans	5.20	-	0.35	-	0.79
Mallow	20.30	0.013	4.12	1.33	9.33
Lettuce	13.70	0.01	2.27	1.02	5.14
Green okra	5.20	4.47×10^{-3}	0.61	0.46	1.38
Potato	52.60	0.042	6.25	4.30	14.13
Spinach	20.30	-	4.41	-	10.00
Squash	20.30	0.01	1.78	1.02	4.03
Tomato	190.10	0.18	9.41	18.40	21.31
Apple	16.40	0.020	0.53	2.05	1.20
Cantaloupe	21.60	-	3.83	-	1.690
Fresh fig	14.30	7.44×10^{-3}	0.73	0.76	1.60
Grape	37.80	0.01	2.21	1.02	5.00
Guava	14.30	-	0.38	-	0.86
Mango	6.60	-	0.05	-	0.11
Water melon	38.40	0.03	1.55	3.07	0.05
Stewed bean	17.50	-	3.30	-	7.47
Sprouted bean	3.60	-	0.83	-	1.86
Flour	5.80	0.004	0.14	0.40	0.31
Lentils	2.70	-	0.54	-	1.24
Macaroni	144.40	0.130	5.79	13.28	13.10
Rice	144.40	-	2.017	-	4.57
Meat	9.60	8.93×10^{-3}	1.39	0.91	2.02
Fish	28.80	0.03	7.128	3.07	7.86
Milk	64.73	0.07	6.43	7.16	14.64

Table 8. The levels of ^{226}Ra, ^{232}Th and ^{40}K in agricultural fertilizers.

Type of Fertilizer	Concentrations of radionuclides (Bq kg^{-1})		
	^{226}Ra	^{232}Th	^{40}K
Ammonium nitrate	0.52 ± 0.03	-	4.96 ± 1.04
Ammonium phosphate	0.32 ± 0.20	-	4.90 ± 1.16
Super phosphate	571.22 ± 26.19	6.12 ± 1.16	-
Urea	-	-	8.12 ± 2.57

Table 9. Transfer ratios from soil to different types of plants.

Vegetable	TR	Fruit	TR
Garden rocket	0.130	Apple	0.020
Mallow	0.040	Fresh figs	0.007
Lettuce	0.060	Grapes	0.010
Green okra	0.050	Water melon	0.030
Potato	0.050		
Squash	0.040		
Tomato	0.060		

Conclusions

Finally, some conclusions can be drawn from this work. Radionuclide surveying of soil indicated that very low levels of ^{137}Cs had been detected in some sampling sites and this may be as a result of nuclear detonations performed by nuclear counties and from nuclear power reactor accidents. The calculated DRCFs for the radionuclides in soil showed a good agreement with those in different studies; therefore, they will be suitable for assessing the radioactivity in local soil. The annual external effective doses ranged from 139.64 to 261.44 μSv, with a mean value of 195.06 μSv.

Radioactivity surveying of food showed that ^{226}Ra and ^{40}K were observed with significant levels in vegetables, fruits, seeds, cereals, meats, and milks. It was revealed that vegetables displayed the highest levels among the foods investigated. The annual internal effective doses are 61.32 and 146.54 μSv for ^{226}Ra and ^{40}K, respectively. This showed that vegetables are the greatest contributors, while meat gives the lowest rate of intake.

Super-phosphate fertilizer gives the highest levels for ^{226}Ra and ^{232}Th. Therefore, the high frequency of soil fertilization may lead to the accumulation of U and Th radionuclides in soil.

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