Uptake of U- and Th-series radionuclides by cereal crops in Upper Egypt

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Abstract The main aim of this study is to determine the activity concentrations of uranium series and thorium series radionuclide in crops cultivated in Upper Egypt. Fifteen types of cereal crops were collected from local markets and analyzed by γ-ray spectrometry. The mean activity concentrations of the natural radionuclides ^{238}U, ^{226}Ra and ^{232}Th were 0.67, 1.27 and 0.56 Bq·kg^{-1}, respectively. The annual effective doses from these radionuclides were estimated for different age groups.

Key words Cereal crops, Gamma spectrometer, HPGe detector, Annual committed effective dose, Age groups

1 Introduction

The concentration of natural radionuclides in the environment and man is especially important in areas where the radionuclide concentrations are elevated or enhanced as a result of human activity. The purpose of this study was to evaluate the intake of ^{238}U, ^{226}Ra and ^{232}Th with crops by inhabitants of the Qena region in Egypt. In this area the mean values of ^{232}Th and ^{226}Ra in soil are 13.7±7, and 12.3±4.6 Bq kg^{-1} respectively[1]. Cereal crops play a central role in human nutrition. Radionuclides from the U and Th series present in food and drinking water consumed by man lead to ingestion dose. It is important to determine the radionuclide concentrations in the environment because they constitute a source of exposure of humans, plants and animals. All living organisms are exposed to ionizing radiations by terrestrial radionuclides in the earths crust, building materials, air, water, foods and the human body itself. Assessments of such radiological consequences to the environment generally include consideration of the dose from ingestion of contaminated foodstuffs. Measured or predicted activity concentrations are used in conjunction with ingestion rates to provide an estimate of the overall intake of activity. However, while measurements or predictions generally apply to the raw foodstuff, ingestion rates relate to the form actually eaten[2]. Very few authors have given consideration to the radioactivity in foodstuffs in Egypt[3,4].

2 Materials and methods

2.1 Sampling and preparing

In April 2006, fifteen different kinds of cereal crops were collected from the local market selected according to the local production and use (Qena and Qift, Upper Egypt). The crop samples were washed with water, as for human consumption, weighed, dried in an oven at 80℃ for 24 h, ground into powder[5], and filled in 250 mL polypropylene bottles, which were sealed and left for at least 4 weeks before counting by gamma spectrometry in order to ensure that radioactive secular equilibrium.

2.2 Gamma spectrometry

2.2.1 Instrumentation

Direct determination of radionuclides in crops without any chemical treatment was performed at the Centre for Radiation Protection and Radioecology (ZSR), Hanover University, Germany, using a p-type HPGe coaxial detector (GEM 50198-P) of 35% relative

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Received date: 2008-10-09

efficiency, with a resolution of 1.78 keV at 1.332 MeV. It is shielded with 10 cm lead and 2 mm copper, and coupled to an 8192-channel analyzer[6].

2.2.2 Efficiency and calibration

The counting efficiencies of the γ-ray peaks were measured by using QCY48 and QCY40 standard solutions (Physikalisch-Technische Bundesanstalt PTB Germany) and were determined using a certified standard solution containing ^{210}Pb, ^{57}Co, ^{60}Co, ^{85}Sr, ^{88}Y, ^{109}Cd, ^{113}Sn, ^{139}Ce, ^{137}Cs, and ^{241}Am. The geometry of the experimental samples was the same as that of the standard samples[7].

2.3 Measurements and calculation

2.3.1 Calculation of radionuclides

Following the spectrum analysis, counting rates for each detected photopeak and activity per mass unit for each of the detected nuclides are calculated. The specific activity (in Bq·kg^{-1}) is given by

$$A_{\text{specific}}=(N/t-N_0/t_0)/(I_\gamma\cdot\varepsilon\cdot m) \quad (1)$$

where N is the net counts of a given peak for a sample, t=18–24 h is the counting time for the sample, N_0 is the background of the given peak, t_0=72 h is the counting time for background, ε is the detection efficiency, I_γ is the number of gamma photons per disintegration, and m is the mass in kg of the measured sample.

If there is more than one peak in the energy analysis range for a nuclide, an average of the peak activities is made and the result is then the weighted average nuclide activity. Based on the measured γ-ray peaks, emitted by specific radionuclides in the ^{232}Th and ^{238}U decay series, and ^{40}K, their concentrations in the samples collected were determined. Calculations relied on establishment of secular equilibrium in the samples, due to the much smaller lifetime of daughter radionuclides in the decay series of ^{232}Th and ^{238}U.

The γ-rays of ^{212}Pb (238.63 keV), ^{208}Tl (583.2 keV) and ^{228}Ac (338.4, 911 and 969 keV) were used to determine the ^{232}Th concentration. The γ-rays of ^{234}Th (92.37 and 62.28 keV) were used for the ^{238}U, and the γ-rays of ^{214}Bi (609.3, 1120.3 and 1764.5 keV) and ^{214}Pb (295.2 and 351.9 keV), for ^{226}Ra. The total uncertainty[8] value (Table 1) is composed of the random and systematic errors in all the factors involved in producing the final nuclide concentration result. The error is written as:

$$\frac{u^2(A_s)}{A_s^2}=\frac{u^2(NP)}{(NP)^2}+\frac{u^2(t_c)}{(t_c^2)}+\frac{u^2 I_\gamma(E_\gamma)}{I_\gamma^2(E_\gamma)}+\frac{u^2\varepsilon(E_\gamma)}{\varepsilon^2(E_\gamma)}+\frac{u^2(M)}{M^2} \quad (2)$$

where N and P is net count, t_c is the counting time and N_0 is counting of background.

Table 1 Radioactivity concentrations (Bq·kg^{-1} dry weight) of ^{238}U, ^{226}Ra and ^{232}Th in different edible seeds in Upper Egypt

Samples	Mass / g	^{238}U	^{226}Ra	^{232}Th
Rice	321	0.17±0.37	0.77±0.51	0.59±0.41
Wheat	274	0.79±0.45	1.27±0.52	0.52±0.34
Barley	192	0.88±0.79	1.36±0.58	0.31±0.50
Phaseolus	252	1.38±0.77	0.61±0.58	0.29±0.42
Lentils	204	1.16±0.64	1.80±0.67	0.32±0.43
Bean	216	0.41±0.64	1.38±0.52	0.75±0.53
Chick pea	217	0.30±0.61	1.53±0.83	0.93±0.49
Chick pea	257	0.34±0.53	2.28±0.91	0.16±0.31
Sorghum	231	–	0.99±0.51	0.96±0.60
Maize	279	1.32±0.48	0.59±0.28	0.55±0.43
Maize	244	0.058±0.48	0.92±0.56	0.50±0.25
Lupins	179	0.58±0.81	1.00±0.59	0.41±14.03
Lupins	191	0.19±0.64	0.94±0.57	0.59±0.33
Kidney beans	197	0.88±0.76	1.58±0.91	1.00±0.36
Pea	208	0.88±0.64	2.02±0.81	0.52±0.43
Average		0.67±0.6 (0.058–1.38)	1.27±0.62(0.59–2.28)	0.56±1.33(0.16–1.00)

2.4 Calculation of radiation exposure from ingestion of foodstuffs (crops)

Doses from intakes of radionuclides in cereals (crops) by population groups can be calculated from measurements of the concentrations of radionuclides in cereals, annual consumption rates and dose coefficients. This can be described by Eq.(3) according to BfS modeling[9]:

$$H_j = U_{n.j} \cdot \sum_{\gamma} C_{n,\gamma}\, g_{\mathrm{Ing},\gamma,j} \tag{3}$$

where H_j is annual effective dose by ingestion in Sv for age group j, $C_{n,\gamma}$ is concentration of radionuclide γ in crop type n (Bq·kg^{-1}), $U_{n,j}$ is annual consumption of the food n by the reference person j in kg, and $g_{r,j}$ is ingestions dose coefficient for the radionuclide r for age group j in Sv·Bq^{-1} ICRP[10].

The annual effective ingestion dose to man due to consumption of cereal crops is computed based on annual intake of cereals and the standard dose conversion factors BfS[8].

3 Results and discussion

The study was aimed at determining activity concentrations of ^{238}U, ^{226}Ra and ^{232}Th in cereal crops for inhabitants of Qena and Qift. Fifteen kinds of crops (Table 1) from markets were analyzed to estimat annual effective dose from ^{238}U, ^{226}Ra and ^{232}Th due to the food consumption. The effective dose coefficients for ingestion was used[9].

3.1 Concentrations of natural radionuclides ^{238}U, ^{226}Ra, and ^{232}Th

Table 1 gives the radioactivity concentration of the cereal crops collected from Qena and Qift. The ^{238}U concentration ranged from 0.058 to 1.38 Bq·kg^{-1}, and ^{232}Th concentration from 0.16 to 1.0 Bq·kg^{-1}. The mean radionuclide concentrations in the cereals considered is 1.27±0.62 Bq·kg^{-1} for ^{226}Ra, with the chick pea (Sample 8) having the highest radionuclide concentration (2.28±0.91Bq·kg^{-1} for ^{226}Ra) of all the crops and radionuclides. Large differences were observed between the activities of ^{226}Ra in different crops due to use of phosphate fertilizers, which may contain considerable levels of radionuclides from the U and Th series[11]. The activity concentrations of radionuclides from this study are comparable with those reported for other countries (Table 2).

The comparison of the activity concentrations of natural radionuclides in different cereals from various countries indicates that the activity concentration of natural radionuclides in Qena, Egypt is different from that in other regions of the world. In Egypt and Spain, similar values can be found with respect to ^{226}Ra for lentils, bean and chickpea crops, but bean and chickpea in Egypt have less ^{238}U concentration than in Spain. Table 2 also shows that the ^{226}Ra concentration of all the cereals in Egypt is higher than that in Refs.[5,13,14,16,20], and the ^{232}Th concentrations of wheat and barley from Egypt are less than from Morocco[19].

3.2 Annual effective dose due to ingestion of different crops for different age groups.

To estimate the annual effective doses due to ingestion of radionuclides in cereal crops, a set of standard food consumption rates for relevant crops was derived from data from the ICRP, applying the conversion factor of for U and Th series which converts to human effective dose BfS model, the average annual effective dose due to γ-rays from the U and Th series sources was determined for each crops with different age groups assessed according to Publication 60 of ICRP[10].

It has been emphasized that it is possible to use radioactivity concentration of natural radionuclides in cereals (Table 1) to give an estimate of the annual effective doses for different age groups from the food chain (cereal crops). However, the annual dose to an average consumer of all the foodstuffs (only crops) studied at the two sites can be estimated, and the results for age groups are summarised in Table 3, with the estimated annual cereals consumption rates of 12, 30, 80, 95, 110 and 110 kg for age groups of <1 a, 1–2 a, 2–7 a, 7–12 a, 12–17 a and > 17 a, respectively, using the dose coefficients in Ref.[8]. ^{210}Pb and ^{226}Ra were important contributors to the overall doses. This notes the considerable variability in results for different crops.

Table 2 Concentration of U and Th in crops (cereals) in Egypt and other countries (Bq·kg^{-1})

Isotopes	Rice	Wheat	Barley	Lentils	Bean	Chickpea	Country	References
^{238}U	0.17±0.37	0.79±0.45	0.88±0.79	1.16±0.64	0.41±0.64	0.3±0.6	Egypt	Present work
	–	0.05±0.008	–	–	–	–	India	[12]
	–	0.0023	–	–	0.005	–	Poland	[13]
	–	0.013	–	1.52	1.02	1.26	Spain	[14]
	0.0026	–	–	–	–	–	Brazil	[15]
	0.005	0.0106	–	–	–	–	Hong Kong	[5]
	–	–	0.0106	–	–	–	Poland	[16]
	2.75±1.2	0.0049	–	–	9.7±3	–	Nigeria	[17]
	0.005	2.3±0.12	–	0.0009	–	–	Iran	[18]
	–	–	3.69±0.25	–	–	–	Morocco	[19]
^{226}Ra	0.77±0.51	1.27±0.52	1.36±0.58	1.80±0.58	1.38±0.52	1.53±0.83	Egypt	Present work
	0.11	0.23	–	–	1.4	–	Brazil	[20]
	–	0.7±0.1	–	–	–	–	India	[12]
	–	0.048	–	–	0.048	–	Poland	[13]
	–	–	–	1.8	3.23	2.75	Spain	[14]
	0.011	0.025	–	–	–	–	Brazil	[15]
	–	–	–	–	0.748±0.06	–	Brazil	[21]
	0.006	–	–	–	–	–	Hong Kong	[5]
	–	0.0871	–	–	–	–	Poland	[16]
	0.097	1.153	–	0.006	–	–	Iran	[18]
^{232}Th	0.59±0.41	0.52±0.34	0.31±0.50	0.32±0.43	0.75±0.53	–	Egypt	Present work
	–	1.1±0.02	–	–	–	–	India	[12]
	–	0.00213	–	–	0.0021	–	Poland	[13]
	0.0032	0.0015	–	–	–	–	Brazil	[15]
	–	0.0211	0.0059	–	–	–	Poland	[16]
	10.5 ± 2	–	–	–	–	–	Nigeria	[17]
	–	2.75±0.16	2.54±0.16	–	–	–	Morocco	[19]

Table 3 Ingestions dose coefficients for each radionuclide by age group in SvBq^{-1}[9]

Age groups / a	< 1	1–2	2–7	7–12	12–17	>17
^{226}Ra	4.70×10^{-6}	9.60×10^{-7}	6.20×10^{-7}	8.00×10^{-7}	1.50×10^{-6}	2.80×10^{-7}
^{238}U	3.40×10^{-6}	1.20×10^{-7}	8.00×10^{-8}	6.80×10^{-8}	6.70×10^{-8}	4.50×10^{-8}
^{232}Th	4.60×10^{-6}	4.50×10^{-7}	3.50×10^{-7}	2.90×10^{-7}	2.50×10^{-7}	2.30×10^{-7}

The ingestion doses due to each radionuclide are presented in Table 4 and compared with the global dose due to ingestion of naturally occurring U and Th series nuclides excluding radon and thoron reported by UNSCEAR[22]. This work indicated that the main contributors to intakes of activity were foods (such as cereals), which would normally originate from a wide area rather than being produced locally. The general food categories considered in this study, i.e. wheat, rice, or other crops, together contributed from the intake of U and Th series. From Table 4 and Fig.1, it can be seen that the total annual effective doses for the <1 a group for all crops is higher than those for other age groups.

Table 4 Annual effective dose due to ingestion of different kind of cereals (in mSv) by different age (years) groups

Crops	Radionuclides	Concentration / Bq·kg^{-1}	Age group / a					
			<1	1-2	2-7	7-12	12-17	>17
Rice	^{226}Ra	0.77	0.0435	0.0222	0.0382	0.0586	0.1272	0.0237
	^{238}U	0.17	0.0070	0.0006	0.0011	0.0011	0.0013	0.0009
	^{232}Th	1.00	0.3610	0.1715	0.2728	0.3716	0.5847	0.0761
Wheat	^{226}Ra	1.27	0.072	0.037	0.063	0.096	0.209	0.039
	^{238}U	0.79	0.032	0.003	0.005	0.005	0.006	0.004
	^{232}Th	0.37	0.134	0.063	0.101	0.137	0.216	0.028
Barley	^{226}Ra	1.36	0.077	0.039	0.067	0.103	0.224	0.042
	^{238}U	0.88	0.036	0.003	0.006	0.006	0.006	0.004
	^{210}Pb	2.04	0.206	0.221	0.360	0.369	0.427	0.155
	^{232}Th	0.48	0.174	0.083	0.131	0.179	0.281	0.037
Phasols	^{226}Ra	0.62	0.035	0.018	0.030	0.047	0.101	0.019
	^{238}U	1.38	0.056	0.005	0.009	0.009	0.010	0.007
	^{232}Th	0.24	0.088	0.042	0.066	0.090	0.142	0.018
Lentils	^{226}Ra	1.79	0.101	0.052	0.089	0.137	0.297	0.055
	^{238}U	1.16	0.047	0.004	0.007	0.008	0.009	0.006
	^{232}Th	0.34	0.122	0.058	0.092	0.126	0.198	0.026
Bean	^{226}Ra	1.38	0.078	0.040	0.069	0.105	0.228	0.043
	^{238}U	0.41	0.017	0.001	0.003	0.003	0.003	0.002
	^{232}Th	0.62	0.222	0.105	0.168	0.228	0.359	0.047
Chick pea	^{226}Ra	1.53	0.087	0.044	0.076	0.117	0.253	0.047
	^{238}U	0.30	0.012	0.001	0.002	0.002	0.002	0.001
	^{232}Th	0.65	0.234	0.111	0.177	0.241	0.380	0.049
Sorghum	^{226}Ra	0.993	0.056	0.029	0.049	0.075	0.164	0.031
	^{238}U	0.000	0.000	0.000	0.000	0.000	0.000	0.000
	^{232}Th	1.100	0.396	0.188	0.299	0.408	0.641	0.000
Maize	^{226}Ra	0.595	0.034	0.017	0.029	0.045	0.098	0.018
	^{238}U	1.320	0.054	0.005	0.008	0.009	0.010	0.007
	^{232}Th	0.453	0.163	0.077	0.123	0.168	0.264	0.034
Lupines	^{226}Ra	1.003	0.057	0.029	0.050	0.076	0.166	0.031
	^{238}U	0.578	0.024	0.002	0.004	0.004	0.004	0.003
	^{232}Th	0.367	0.132	0.063	0.100	0.136	0.214	0.028
Kidney beans	^{226}Ra-	1.585	0.089	0.046	0.079	0.120	0.262	0.049
	^{238}U	0.883	0.036	0.003	0.006	0.006	0.007	0.004
	^{232}Th	0,50	0.022	0.006	0.009	0.007	0.005	0.004
Pea	^{226}Ra	2.016	0.114	0.058	0.100	0.153	0.333	0.062
	^{238}U	0.881	0.036	0.003	0.006	0.006	0.006	0.004
	^{232}Th	0.537	0.193	0.092	0.146	0.199	0.313	0.041

Using the obtained concentration values, the associated annual effective doses were estimated. Table 4 shows the results for each age group, as well as the weighted doses considering the age distribution. In the estimation of doses, the uncertainties considered were only those associated with concentration determination. From the evaluation results in Table 5, it is possible to appreciate that the main contribution to doses are produced by the U series (^{238}U and ^{226}Ra) and ^{232}Th. The annual effective dose values are comparable with reference values published by UNSCEAR[2], taking into account the specific alimentary habits and the age distribution of the Qena population.

With the obtained dose values, the annual effective dose that one average Qena individual receives in one year by the ingestion of food (cereal) was evaluated with different age groups.

The results obtained for the geometric mean of annual effective dose with different age are presented in Table 5. Age-dependent dose coefficients are now available from ICRP, and the number of age groups considered could be expanded to six: 3 months (from 0 to 1 a), 1 year (from 1 year to 2 a), 5 a (>2 to 7 a), 10 years (>7 to 12 a), 15 a (>12 to 17 a), and adult[9]. Unless otherwise stated, in Table 6 and Fig.1, the discussion is confined to >12 years to 17 years having higher than average consumption rates. However, the trends for 3 months, 1 year 5 years 10 years and >7 years to 12 years age groups were nearly similar for all cereals crops.

Table 5 The total annual effective dose (mSv) with age group (year) for different cereals crops

Crops	<1 a	1-2 a	2-7 a	7-12a	12-17 a	>17 a
Rice	0.412	0.194	0.312	0.431	0.713	0.101
Wheat	0.237	0.103	0.169	0.239	0.431	0.071
Barley	0.286	0.125	0.204	0.288	0.512	0.083
Phasols	0.179	0.064	0.105	0.146	0.253	0.044
Lentils	0.271	0.114	0.189	0.270	0.503	0.087
Bean	0.317	0.147	0.239	0.336	0.591	0.091
Chick pea	0.639	0.462	0.752	0.867	1.219	0.313
Sorghum	0.452	0.217	0.348	0.483	0.805	0.031
Maize	0.492	0.346	0.563	0.633	0.846	0.233
Lupines	0.212	0.094	0.153	0.216	0.384	0.062
Kidney Beans	0.148	0.054	0.093	0.133	0.273	0.057
Pea	0.343	0.153	0.252	0.358	0.652	0.107

The relative contributions from each radionuclide to the overall dose from a given cereals crops are summarized in Table 6; the overall doses are also given. For ^{210}Pb dose from cereals crops was generally very much greater than all radionuclides in the same crops.

4 Conclusion

Gamma-ray spectroscopy for rapid assessment of radiation exposure, identification of radionuclides and detection of changes in environmental radioactivity has been employed in the determination of the natural gamma radiation dose levels in 15 crops collected from Qena and Qift cities of Upper Egypt.

As a result of this study, the concentrations of radionuclides ^{238}U, ^{226}Ra, and ^{232}Th, are in the same order of magnitude as the results obtained by other authors and the reference values established by the several authors.

The natural gamma radiation dose levels in the crops samples have been determined from the measurement of specific activities of the Uranium series and Thorium series. The average annual effective doses due to gamma radiation in these crops were low and almost insignificant health burden on the population from radiation protection point of view.

References

1 Ahmed N, El-Arabi A G M. J Environ Radioact, 2005, **84:** 51–64.

2 UNSCEAR: Report to General Assembly, with Scientific Annexes, Sources and Effects of Ionizing Radiation. New York: United Nations, 2000.

3 Gomaa M A, Abdel-Fattah A T, Essa M W, *et al.* Appl Radiat lsot, 1995, **46:** 607–608.

4 Badran H M, Sharshar T, Elnimer T. J Environ Radioact, 2003, **67:** 181–190.

5 Yu K N, Mao S Y. Health Phys, 1999, **77:** 686–696.

6 Harb S. Thesis, ZSR, Hannover University, Germany, 2004.

7 Harb S, Salahel Din K, Abbady A. Efficiency Calibrations of HPGe Detectors for Gamma spectrometry levels of Environmental Samples, 3rd Environmental Physics Conference, Aswan, Egypt, Feb 2008, 19–23.

8 International Standards Organization (ISO/IEC 17025:1999), European Committee for Standardization, Brussels, 1999.

9 Bundesamt für Strahlenschutz (BfS), Berechnungsgrundlage Bergbau, Report, 1999 (in German).

10 USEPA. US Environmental Protection Agency, Washington D.C., 1999.

11 International Commission on Radiological Protection, ICRP publication 60, 1990.

12 Pulhani V A, Dafauti S, Hegde A G, *et al.* J Environ Radioact, 2005, **79:** 331–346.

13 Pietrzak-Flis Z, Rosiak L, Suplinska M M, *et al.* Sci Tot Environ, 2001, **273:** 163–169.

14 Hernandez F, Hernandez-Armas, Catalan J A, *et al.* Radia Prot Dosim, 2004, **111:** 205–210.

15 Santos E E, Lauria D C, Amaral E C S, *et al.* J Environ Radioact, 2002, **62:** 75–86.

16 Pietrzak-Flis Z, Suplinska M M, Rosiak L. J Radioanal Nucl Chem, 1997, **222:** 189–193.

17 Arogunjo A M, Ofuga E E, Afolabi M A. J Environ Radioact, 2005, **82:** 1–6.

18 Hosseini T, Fathivand A A, Abbasisiar F, *et al.* Radiat Prot Dosi, 2006, **118:** 1–3.

19 Misdaq M A, Bourzik W. J Radioanal Nucl Chem, 2002, **254:** 3551–3555.

20 Ventuini L, Sordi G A A. Health Phys, 1999, **76:** 311–313.

21 Povinec P P, Pham M K, Sanchez-Cabeza J A, *et al.* J Radioanal Nucl Chem, 2007, **273:** 383–393.

22 UNSCEAR: Report to General Assembly, with Scientific Annexes, Sources and Effects of Ionizing Radiation, New York: United Nations, 2000.