Assessment of Agriculture Soil Primordial Radionuclide Concentrations in Aden Governorate South Of Yemen Region

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Abstract: Thirty six samples of farm soils, taken to a depth of up to 30 cm in cultivated land, were collected over the Aden governorate south of Yemen region. Activity concentration of background radionuclides such as 226Ra, 232Th and 40K of these samples were determined by gamma-ray spectrometry with NaI (Tl) detector. The average concentrations in Bq.kg−1 dry weight of determined radionuclides 226Ra, 232Th and 40K were 48.01±3.84, 58.11±4.65 and 624.80±49.98 Bq.kg−1 for Beer Ahmed- Beer Fadle area farm soil. For Daar-saad -Al-Masabian area farm soil the corresponding values were 70.78±5.66, 84.75±6.78 and 771.53±61.72 Bq.kg−1, respectively. Radium equivalent (Raeq), dose rate, annual effective dose, and hazard indices (Hex and Hin) average values were (179.23, 251.38) Bq.kg−1, (82.98, 114.07) nGy h-1, (0.10, 0.14) mSv and (0.48, 0.68 and 0.61, 0.87). The results of the present study were discussed and compared with the results of similar investigations and internationally recommended values.

Keywords: Activity concentration, farm soil, NaI (TI), dose rate, annual effective dose.

1 Introduction

Natural radioactivity origins are from the decay of natural radionuclides and their products in the earth's crust and cosmic radionuclide from outer space (UNSCEAR, 1988, 2000). The high geochemical mobility of these radionuclides in the environment allows them to move easily and to contaminate the environment which human come into contact (Egunyinka et al., 2009). Radionuclides (uranium, radium, thorium, and potassium) are found naturally in soil. The amount of radioactivity in soil depends upon the type and at different levels of the soil of each different geological region. It is important to study the distribution and specification of the encountered radioelements and their impact on the environment. Uranium, thorium, and their decay products are radionuclides that represent a potential risk to human health due to the emission of ionizing radiation (Safia, 2014), (El Aassy Ibrahim, 2011). The presence of natural radioactivity in soil and other building materials results in internal and external exposure to the occupants. NORM existing in soil could pose potential health risk. Terrestrial radioactivity and the associated external exposure due to the gamma radiation depend primarily on the geological and geographical conditions and appear at different levels in the soils of each region (Alaamer, 2012). Measurement of natural radioactivity in rocks and soils is very important to determine and monitor the amount of change of the natural background activity with time for environmental protection. (Najam Laith A., et al, 2011).

The aim of this study is to measuring the natural radioactivity contents, annual effective radiation dose, and external radiation hazard indices in the farm soils of Beer Ahmed- Beer Fadle and Daar-saad -Al-Masabian area. The data generated in this study will provide base line values of natural radioactivity in soils and may be useful for studies on this issue.

2 Experimental procedures

2.1 Sampling and samples preparation

For the measurement of the natural radioactivity in soil, surface-soil samples were collected randomly from different locations in the Aden governorate south of Yemen region as shown in figure (1). The soil samples were collected from an auger hole at a depth of about 0-30 cm so as to sample the natural soil. After collection, samples were crushed into fine powder by using mortar and pestle. The final grain-sizes of the samples were obtained by straining through a 100 micron-mesh size.
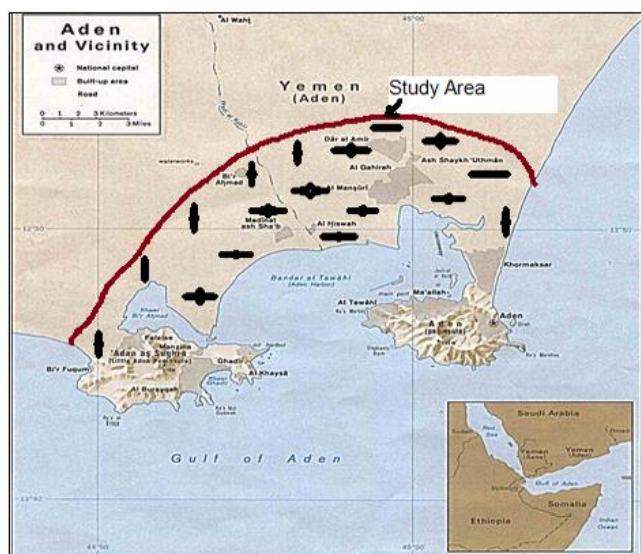
Figure 1: Map showing the location of the area studied.

An average 400g of soil is used per sample. Before measurement samples were dried in an oven at a temperature of 100°C for 24 hours. Each sample was packed and sealed in an airtight PVC container and kept for about a four-week period to allow radioactive equilibrium among the daughter products of radon (^{222}Rn), thoron (^{220}Rn) and their short lived decay products (Dabayneh et al., 2008)

2.2 Detection Technique

Each sample was measured with a gamma-ray spectrometer consisting of a NaI (Tl) (2x2) inch setup and multi-channel analyzer 8192 channel, with the following specifications: resolution (FWHM) at 1.33 MeV ^{60}Co is 60keV- relative efficiency at 1.33MeV ^{60}Co is 7.5 %. The detector is shielded in a chamber of two layers starting with stainless steel (10 mm thick) and leads (30 mm thick). This shield serves to reduce different background radioactivity.
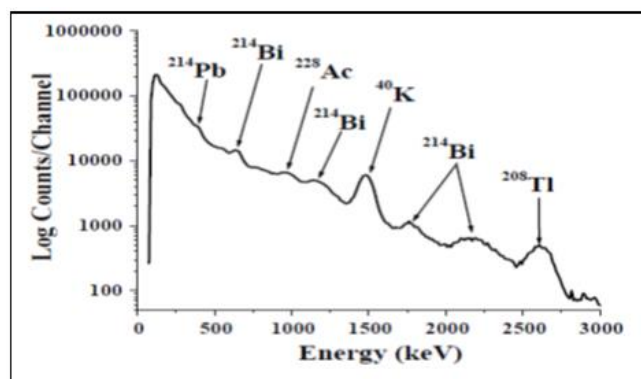

Figure 2: The energy spectrum recorded for soil sample by scintillation detector NaI(Tl).

The detector was calibrated using standard source QCYB41 from Physikalisch Technische Bundesanstalt PTB, Germany; which has ten radionuclides with twelve γ- ray emitters ranged from 230 to 1836 keV.(A mixed source containing (^{133}Ba, ^{57}Co, ^{139}Ce, ^{133}Ba, ^{85}Sr, ^{137}Cs, ^{54}Mg, ^{88}Y, ,^{65}Zn and ^{88}Y,). For calibration, the standard source is placed above the detector, and the measurement started. Figure (2) shows the energy spectrum recorded for soil sample by scintillation detector NaI (Tl) for 24 h the time of counts. The dependence of the efficiency on the radiation energy was determined at (zero) mm sample detector distance. The absolute efficiency of the NaI (Tl) detector was determined. The detector efficiency decreases continuously with energy. Each sample was placed in plastic containers of the same size as that of the multi-element standard.

The spectra were either evaluated with the computer software program Maestro (EG and G ORTEC) or manually with the use of a spread sheet (Microsoft Excel) to calculate the natural radioactivity. ^{226}Ra activity of the samples was determined via its daughters (^{214}Pb and ^{214}Bi) through the intensity of the 351.93 keV, for ^{214}Pb and, 1120 and 1764.49 keV, for ^{214}Bi gamma-line. ^{232}Th activity of the sample was determined from the daughters (^{228}Ac), (^{212}Pb) and (^{208}Tl) through the intensity of 911.2 keV gamma -line for (^{228}Ac), and (^{208}Tl) emission at 2614 keV gamma line. ^{40}K activity was determined from the 1460.7 keV emission gamma-lines. The samples were counted for 12-24 h depending on the concentration of the radionuclides.

2.3 Measurement of natural radioactivity

Through calculating the area under the peak (net area) and by means of the detector efficiency curve, the specific activity (activity concentration) A_{Ei} was determined using the formula.

$$A_{Ei} = \frac{NP}{t_c . I_\gamma (E_\gamma) . \varepsilon (E_\gamma) . M} \quad (1)$$

Where NP is the number of count in a given peak area corrected for background peaks of a peak at energy E,ε (E_γ) the detection efficiency at energy E, t_c is the counting lifetime, I_γ (E_γ) the number of gammas per disintegration of this nuclide for a transition at energy E, and M the mass in kg of the measured sample (S.Harb 2004).

2.4 Calculation of air-absorbed dose rate

The external, terrestrial γ-radiation, absorbed dose rate in air at a height of about 1 m above the ground is calculated by using the conversion factor of 0.042 nGy h^{-1}/ Bq kg^{-1} for ^{40}K, 0.455 nGy h^{-1}/Bq kg^{-1} for ^{226}Ra, and 0.583 nGy h^{-1}/Bq kg^{-1} for ^{232}Th (UNSCEAR, 1993) assuming that the ^{235}U decay series can be neglected. They contribute very little to the total dose from the environmental background (Kocher and Sjoreen, 1985; Jacob et al., 1986; Leung et al., 1990).

$$D\ (nGy.h^{-1}) = (0.4551)\ A_U + (0.583)\ A_{Th} + (0.042)\ A_K \quad (2)$$

Table 1: Activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in cultivated soil samples from Beer Ahmed-Beer Fadle and Daar-Saad-Al-Masabian areas Aden Governorate, South of Yemen.

	Beer Ahmed-Beer Fadle area				Daar-Saad-Al-Masabian area		
S.No.	Activity concentration Bq kg^{-1}			S.No.	Activity concentration Bq kg^{-1}		
	^{226}Ra	^{232}Th	^{40}K		^{226}Ra	^{232}Th	^{40}K
1	82.12±6.57	94.75±7.58	738.4±59.07	1	104.22±8.33	129.48±10.35	967.2±77.37
2	83.66±6.69	95.77±7.66	893.9±71.51	2	43.70±3.49	59.13±4.73	738.4±59.07
3	88.05±7.04	98.70±7.89	895.9±71.67	3	35.73±2.85	43.82±3.50	652.8±52.22
4	20.18±1.61	33.45±2.67	375.5±30.04	4	32.79±2.62	41.86±3.34	609.6±48.76
5	25.96±2.07	37.31±2.98	543.8±43.504	5	37.14±2.97	47.76±3.82	624.0±49.92
6	48.14±3.85	52.09±4.16	666.5±53.32	6	92.31±7.38	101.54±8.12	709.6±56.76
7	15.39±1.23	30.26±2.42	297.9±23.832	7	104.48±8.35	123.65±9.89	836.4±66.91
8	46.25±3.70	50.83±4.06	727.6±58.21	8	89.21±7.13	109.47±8.75	790.65±63.25
9	39.35±3.14	46.23±3.69	643.7±51.49	9	48.48±3.87	62.32±4.98	580.9±46.47
10	34.12±3.22	42.75±3.42	565.9±45.27	10	52.83±4.22	65.22±5.21	621.78±49.74
11	40.24±3.49	56.82±4.54	647.7±51.81	11	68.80±5.50	75.86±6.06	765.67±61.25
12	43.74±3.49	49.16±3.93	580.2±46.41	12	62.10±4.96	71.40±5.71	734.3±58.74
13	73.68±5.89	89.12±7.13	768.4±61.47	13	60.94±4.87	70.63±5.65	690.22±55.21
14	24.84±1.98	36.56±2.92	289.5±23.16	14	106.52±8.52	120.01±9.60	960.6±76.84
15	31.76±2.54	41.17±3.29	634.8±50.78	15	108.18±8.65	132.12±10.57	1120.23±89.62
16	64.06±5.12	72.71±5.81	695.3±55.62	16	80.56±6.44	98.2±7.85	854.34±68.35
17	31.66±2.53	41.11±3.28	490.4±39.23	17	76.8±6.14	92.7±7.41	843.54±67.48
18	70.91±5.67	77.27±6.18	781.5±62.52	18	69.2±5.53	80.3±6.42	787.43±62.99
Mean	**48.01±3.84**	**58.11±4.65**	**624.27±49.94**	Mean	**70.78±5.66**	**84.75±6.78**	**771.53±61.72**

Where: A_{Ra}, A_{Th} and A_K are the mean activity concentrations of ^{238}U, ^{232}Th and ^{40}K, respectively, in (Bq kg^{-1}).

2.5 Calculation of annual effective dose

To estimate annual effective doses, the following must be considered: (a) the conversion coefficient from absorbed dose in air to effective dose and (b) the indoor occupancy factor. The annual, estimated, average, effective- dose equivalent received by a member is calculated using a conversion factor of 0.7 Sv Gy^{-1}, which is used to convert the absorbed rate to human effective-dose equivalent with an outdoor occupancy of 20% and 80% for indoors (UNSCEAR, 1993).

The annual effective doses are determined as follows:

Effective dose rate (mSv.yr^{-1})
= Absorbed dose (nGy h^{-1}) × 8760 h.yr^{-1} ×0. 7× (10^3 mSv /10^9) × 0.2 (nGy^{-1}) (3)

2.6 External hazard index

External hazard index due to the emitted gamma-rays of the samples are calculated and examined according to the following criterion (Beretka and Mathew, 1985):

$$H_{ex} = A_{Ra}/370 + A_{Th}/259 + A_K/4810 \leq 1 \quad (4)$$

Where; A_{Ra}, A_{Th}, A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively.

2.7 Internal hazard index

In addition to external hazard index, radon and its short-lived products are also hazardous to the respiratory organs. The internal exposure to radon and its daughter products is quantified by the internal hazard index (Hin), which is given by the equation. (Beretka and Mathew, 1985).

$$H_{in} = A_{Ra}/185 + A_{Th}/259 + A_K/4810 \leq 1 \quad (5)$$

The values of the indeces (H$_{ex}$, H$_{in}$) must be less than unity for the radiation hazard to be negligible.

3 Results and Discussion

From the gamma spectrometric analysis, three naturally occurring radionuclides were determined ^{226}Ra, ^{232}Th and ^{40}K. Table 1 illustrates the specific activities in Bq/.kg dry weight of the natural radionuclides ^{226}Ra, ^{232}Th, and ^{40}K in the collected samples. The average activity concentration in the collected soil samples from Beer Ahmed-Beer Fadle area ranged from 15.39±1.23 to 88.05±7.04 Bq kg^{-1} with an average value of 48.01±3.84 Bq kg^{-1} for ^{226}Ra. The ^{232}Th specific activities ranged from 30.26±2.42 to 98.70±7.89 Bq kg^{-1} with an average value of 58.11±4.65 Bq kg^{-1}. The ^{40}K specific activities ranged from 297.9±23.832 to 895.9±71.67 Bq kg^{-1} with an average value of 624.27±49.94 Bq kg^{-1}. While the average activity concentration in the collected soil samples from Daar-Saad-

Al-Masabian area ranged from 32.79±2.62 to 108.18±8.65 Bq kg^{-1} with an average value of 70.78±5.66 Bq kg^{-1} for ^{226}Ra. The ^{232}Th specific activities ranged from 41.86±3.34

to 132.12±10.57 Bq kg^{-1} with an average value of 84.75±6.78 Bq kg^{-1}. The ^{40}K specific activities ranged from

Table 2: Radium equivalent activity, Ra$_{eq}$ (Bqkg^{-1}), representative level index, I$_{\gamma r}$, external hazard index, H$_{ex}$, and internal hazard index, H$_{in}$, in all cultivated soil samples.

	Beer Ahmed-Beer Fadle area					Daar-Saad-Al-Masabian area					
S.No.	Ra$_{eq}$ BqKg^{-1}	D (nGy/h)	D$_{eff}$ mSv/y	H$_{ex}$	H$_{in}$	S. No.	Ra$_{eq}$ BqKg^{-1}	D (nGy/h)	D$_{eff}$ mSv/y	H$_{ex}$	H$_{in}$
1	274.48	123.67	0.151	0.74	0.96	1	363.86	163.60	0.200	0.98	1.26
2	289.45	131.50	0.161	0.78	1.01	2	185.12	85.41	0.104	0.50	0.62
3	298.19	135.29	0.165	0.80	1.04	3	148.66	69.25	0.084	0.40	0.49
4	96.93	44.47	0.054	0.26	0.31	4	139.60	64.95	0.079	0.37	0.46
5	121.19	56.42	0.069	0.32	0.39	5	153.48	70.97	0.087	0.41	0.51
6	173.95	80.29	0.098	0.47	0.59	6	292.15	131.06	0.160	0.79	1.03
7	81.60	37.17	0.045	0.22	0.26	7	345.71	154.83	0.189	0.93	1.21
8	174.98	81.27	0.099	0.47	0.59	8	306.65	137.69	0.168	0.83	1.06
9	155.02	71.92	0.088	0.42	0.52	9	182.34	82.83	0.101	0.49	0.62
10	138.83	64.24	0.078	0.37	0.46	10	193.97	88.21	0.108	0.52	0.66
11	171.37	78.67	0.096	0.46	0.57	11	236.25	107.73	0.132	0.64	0.85
12	158.72	72.96	0.089	0.43	0.54	12	220.75	100.77	0.123	0.59	0.76
13	260.29	117.81	0.144	0.70	0.90	13	215.10	97.94	0.120	0.58	0.74
14	99.41	44.79	0.054	0.27	0.33	14	352.12	158.85	0.194	0.95	1.23
15	139.53	65.14	0.079	0.37	0.46	15	383.38	173.38	0.212	1.03	1.32
16	221.57	100.78	0.123	0.59	0.77	16	286.77	129.84	0.159	0.77	0.99
17	128.22	58.99	0.072	0.34	0.43	17	274.31	124.47	0.152	0.74	0.94
18	241.59	110.18	0.135	0.65	0.84	18	244.66	111.42	0.136	0.66	0.84
Mean	**179.23**	**82.98**	**0.100**	**0.48**	**0.61**	**Mean**	**251.38**	**114.07**	**0.14**	**0.68**	**0.87**

609.6±48.76 to 1120.23±89.62 Bq kg^{-1} with an average value of 771.53±61.72 Bq kg^{-1}. The average value of Ra-226 and Th-232 at Beer Ahmed- Beer Fadle and Daar-saad -Al-Masabian are closed. The average activity value of ^{226}Ra from Daar-Saad-Al-Masabian area is about 1.4 times higher than that of ^{226}Ra from Beer Ahmed-Beer Fadle area and two times higher than the worldwide value reported by (UNSCEAR, 2000). The average activity value of ^{232}Th from Daar-Saad-Al-Masabian area is about 1.4 times higher than that of ^{232}Th from Beer Ahmed-Beer Fadle area and 2.8 times higher than the worldwide value reported by (UNSCEAR, 2000). This narrow range of the activity concentrations is probably due to the fact that the sites studied cover an area with similar aquifer lithology's and consequent not large differences in radionuclide solubilities and mobilities. The average activity value of ^{40}K from Daar-Saad-Al-Masabian area is about 1.2 times higher than

that of ^{40}K from Beer Ahmed-Beer Fadle area and 1.9 times higher than the worldwide value reported by (UNSCEAR, 2000). The high value of ^{40}K in the area under study is may be due to the wide using of different quantities or species of fertilizers.

Table 2 gives the radium equivalent (Raeq), in addition to the dose rate (D), the annual effective dose (D$_{eff}$), and the external and internal hazard indices (H$_{ex}$ and

H$_{in}$). Raeq Values for samples from Beer Ahmed-Beer Fadle area ranged from 81.60 to 298.19 Bq kg^{-1} with an average value of 179.23 Bq kg^{-1}and for samples from Daar-Saad-Al-Masabian area ranged from139.60 to 383.38 Bq kg^{-1} with an average value of 251.38 Bq kg^{-1}. These values are lower than the recommended maximum value of 370 Bq kg^{-1} (UNSCEAR, 2000). The gamma dose rates due to naturally occurring terrestrial radionuclides ^{226}Ra, ^{232}Th and ^{40}K were calculated based on their activities in soil samples, determined by gamma-ray spectrometry. The absorbed gamma dose rate due to these radionuclides from Beer Ahmed-Beer Fadle area ranged from 37.17 to 135.29 nGyh^{-1} with an average value of 82.98 nGyh^{-1} and from 64.95 to 173.38 nGyh^{-1} with an average value of 114.07 nGyh–1 which is within the value given in UNSCEAR report 2000 (57 nGyh^{-1}) in Beer Ahmed-Beer Fadle samples and 1.9 times higher than the value given in UNSCEAR report 2000 in Daar-Saad-Al-Masabian samples.

The estimated annual effective dose for samples from Beer Ahmed-Beer Fadle samples ranged from 0.045 to 0.165 mSv with an average value of 0.100 mSv and for samples from Daar-Saad-Al-Masabian ranged from 0.079 to 0.212 mSv with an average value of 0.139 mSv. While the world wide average of annual effective dose is approximately 0.07mSv. The external hazard index (Hex) for samples from Beer Ahmed-Beer Fadle samples ranged from 0.22 to 0.80 with an average value of 0.48 and for

samples from Daar-Saad-Al-Masabian ranged from 0.37 to 1.03 with an average value of 0.68. The internal exposure to ^{222}Rn and its radioactive progeny is controlled by the internal hazard index (Hin), for samples from Beer Ahmed-Beer Fadle samples ranged from 0.26 to 1.04 with an average value of 0.61 and for samples from Daar-Saad-Al-Masabian ranged from 0.46 to 1.32 with an average value of 0.87. H_{ex} and H_{in} must not exceed the limit of unity for the radiation hazard to be negligible. In this

Table 3: Mean values of ^{226}Ra, ^{232}Th and ^{40}K for all cultivated soil samples under investigation beside other countries.

Country	Activity concentration Bq kg^{-1}			Reference
	^{226}Ra	^{232}Th	^{40}K	
Aden Governorate, South Yemen (Beer Ahmed-Beer Fadle)	48.01	58.11	624.27	Present work
Aden Governorate, South Yemen (Daar-Saad-Al-Masabian)	70.78	84.75	771.53	
Egypt (S.V.U.)	11	15	582	Nagwa, 2010
Egypt (Q.G.)	15	22	705	
Greece	27	36	496	Psichoudaki, 2008
Pakistan	28	51	589	Nasim Akhtar and M. Tufail, 2007
Vojvodina	40	53	554	I. Bikit et al., 2005
India	57	87	143	Surinder et al., 2005
India	39	49	348	Pulhani et al., 2005
Jordan	84	82	560	Al-Jundia et al., 2003
worldwide	35	30	400	UNSCEAR, 2000

study, the calculated average values of external and internal hazard indices were lower than unity, which means that this area is safe for the people live. The results shows for samples from Daar-Saad-Al-Masabian samples are higher than with the average of samples from Beer Ahmed-Beer Fadle. This refers to the addition in excessive rate of the inorganic phosphate fertilizers accompanied with the natural radioactivity nuclides in this area higher than Beer Ahmed-Beer Fadle area. The activity concentrations of radionuclides of ^{226}Ra, ^{232}Th and ^{40}K measured by gamma-ray spectrometry system for cultivated soil samples collected over the Aden governorate south of Yemen region. In this study, it is found that the value of ^{226}Ra, ^{232}Th and ^{40}K was higher than that all other comparable countries as shown in table 3. This refers to the addition in excessive rate of the inorganic phosphate fertilizers accompanied with the natural radioactivity nuclides in cultivated soil samples in Aden governorate farms.

4 Conclusion

The activity concentrations of Ra-226, Th-232 and K-40 in all area under study are higher than the worldwide mean values that recorded by UNSCEAR, 2000. Ra$_{eq}$ average value was less than that value identified by UNSCEAR (2000). Absorbed dose rate D (nGy/h) was higher than the world's average value of 60 nGy/h. The effective dose rate (D$_{eff}$ mSv/y was not exceed the recommended value 1 mSv. For the present work, the average value of the external hazard index H$_{ex}$ and internal hazard index H$_{in}$ was less

than unity. These data show that the activity concentration of naturally occurring radionuclides in soil samples were high. The use of fertilizers especially phosphate fertilizer in large extent have affected radionuclides concentration, potassium containing fertilizers are the one of the cause of presence of high activity of ^{40}K in soil. The application of these fertilizers has the effect of an accumulation of radio activity in soils that can be harmful for the population and the production. The enhancement of radioactivity in agricultural land can be controlled for the use of Phosphate Fertilizers.

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