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Assessment of natural radioactivity level and radiation hazards in soil samples of Wadi Al- Rummah Qassim province, Saudi Arabia

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Abstract

The present work deals with estimation and distribution of natural radionuclides in 40 soil samples collected from Wadi Al-Rummah (Qassim part) Central Saudi Arabia. Estimations were carried out using NaI (Tl) gamma-ray spectrometer. Activity concentrations ranged from 5.3 to 45.0 Bq kg^{-1}, from 4.3 to 33.8 Bq kg^{-1} and from 38 Bq kg^{-1} to 273.1 Bq kg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K, respectively. The average activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in soil samples was 15.5 Bq kg^{-1}, 14.1 Bq kg^{-1} and 143.1 Bq kg^{-1}. The results of this study were compared with other studies around the world. Radiation hazard parameters such as radium equivalent activity, annual dose, external hazard were calculated and compared with the recommended levels quoted from International Commission on Radiological Protection (ICRP-60) and United Nations Scientific Committee on the Effects of Atomic Radiation UNSCEAR reports.

Key words

Natural radioactivity levels, Radium equivalent, Radiation hazards soil

Introduction

Soil is the upper part of the earth's crust and is formed as a result of rock deformation by complex physicso-chemical processes, which include weathering decomposition, organic matter addition and water movement. Soil consists of minerals, organic matter, water and air where their percentage vary widely according to soil type, usage, and particle size (Chowdhury *et al.,* 2006; Smith *et al.*, 2015). Natural radioactivity in soil occurs mainly from ^{238}U series, ^{232}Th series and ^{40}K. Naturally occurring radioactive materials (NORMs) in soil are one of the components of external gamma-ray exposure to which persons are exposed regularly. Natural environmental radioactivity and associated external exposure due to gamma radiation depend primarily on geological and geographical conditions and exist at different levels in soil of each region of the world. The specific levels of terrestrial environmental radiation are related to geological composition for each lithologically separated area, and to uranium, thorium and potassium content in rocks from which the soil was formed in each area (Gbadebo and Amos, 2010; Alashrah and El-Taher, 2015).

High level of uranium and its decay products in rock and soil and thorium in monazite sands are the main source of high natural background areas that have been identified in several areas of the world like Yangjiang in China, Rasmar in Iran, Kerala coast of India, etc. (Kurnaz *et al.,* 2007). Therefore, estimation of natural radioactivity in soil is great interest for many researchers throughout the world, which led to worldwide national surveys for last two decades. El-Taher and Abdel Hlim (2014) studied the elemental analysis of Toshki soil in south Egypt by instrumental neutron activation analysis. El-Taher (2010) studied radioactivity in soil samples from Wadi Al-Assuity protective area in upper Egypt. El-Taher and Makhluf (2010) measured ^{226}Ra ^{232}Th and ^{40}K levels in phosphate fertilizer and its environmental implications in Assuit governorate, Upper Egypt. More specifically, natural environmental radioactivity due to gamma radiation depends primarily on the geological and geographical conditions, and appears at different level in

soils of each region in the world (Kurnaz *et al.,*2007). There are no existing databases for the natural radioactivity in soil and along the Wadi Al-Rummah. In view of above, the present study was carried out to assess natural radioactivity level in soil samples of Wadi Al-Rummah and calculate the radiation hazard parameters.

Materials and Methods

Samples collection and preparation : Forty surface soil samples were collected from eight locations in Wadi Al-Rummah in Qassim province. Each sample was taken with a coring tool within area of 1 m^2 and from five point (one in the center and four at the corner). These samples were then mixed to prepare a single sample after eliminating leaves or roots of the plants. To release humidity from samples, they were dried at 110°C for 24 hrs until constant dry weight sample (Veiga *et al.*, 2006). Dried samples were crushed and allowed to pass through micro sieves to maintain uniform grain size to obtain affine-grained homogenous soil sample. About 400 g of homogenized soil samples were transferred into cylindrical containers. They were carefully sealed and stored for at least 30 days before gamma ray analysis was performed to allow ^{226}Ra and its short-lived progenies to reach secular equilibrium.

Gamma-spectrometric measurements were performed with NaI (Tl) detector. The measuring time for gamma-ray spectra range was 12 hrs. In order to determine the background distribution due to naturally occurring radionuclides in the environment around the detector, an empty polystyrene container was counted in the same manner as samples. After measurement and subtraction of the background, the activity concentration was calculated. The specific activity of ^{226}Ra was evaluated from gamma-ray lines of ^{214}Bi at 609.3,1120.3 keV and ^{214}Pb at 351 keV, while specific activity of ^{232}Th was evaluated from gamma-ray lines of ^{228}Ac at 338.4, 911.1 and 968.9 keV. The specific activity of ^{40}K was determined directly from its 1460.8 keV gamma-ray line. Activity calculations were carried out following the method of Lalit and Ramachandra (1978). The activity concentrations of natural radionuclides in the measured samples (AS) were computed by the following formula

$$A_S \ (Bq\ kg^{-1}) = Ca \ / \varepsilon\ Pr\ Ms \qquad (1)$$

where, Ca is the net gamma counting rate (counts per second); ε the detector efficiency of specific γ–ray, Pr is the absolute transition probability of gamma–decay and Ms the mass of the sample (kg) (Uosif *et al.*, 2008).

The external hazard index of the samples were calculated by the following equation (Beretka and Mathew, 1985).

$$H_{ex} = C_{Ra}/370 + C_{Th}/259 + C_K/4810 \qquad (3)$$

In addition, the external irradiation radon and its short-lived products are also hazardous to the respiratory organs. The internal exposure to radon and its daughter products was quantified by the internal hazard index (H_{in}), which is given by the following equation:

$$H_{in} = C_{Ra}/185 + C_{Th}/259 + C_K/4810 \qquad (4)$$

where, C_{Ra}, C_{Th} and C_K are activity concentrations (Bq kg^{-1}) of specific radiation.

Radiation hazards due to specified radionuclides of ^{226}Ra, ^{232}Th and ^{40}K were assessed by another index called representative level index, $I_{\gamma r}$. The following equation was applied to calculate Iγr for soil samples:

$$I\gamma r = (1/150)\ C_{Ra} + (1/100)\ C_{Th} + (1/1500)\ C_K \qquad (5)$$

where, C_{Ra}, C_{Th} and C_K are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K in Bq kg^{-1}, respectively. Iγr varied from 0.05 to 0.41 with a mean value of 0.16 as shown in Table 3.

The absorbed gamma dose rates DR (nGh^{-1}) in air at 1m above the ground surface for the uniform distribution of radionuclides were calculated based on guidelines provided by UNSCEAR (2000)

$$D_R\ (nG\ h^{-1}) = 0.427C_{Ra} + 0.623C_{Th} + 0.043C_K \qquad (6)$$

where, C_{Ra}, C_{Th} and C_K are the activity concentrations (Bq kg^{-1}) of ^{226}Ra, ^{232}Th and ^{40}K, respectively.

The annual effective dose equivalent (AEDE) was calculated from the absorbed dose by applying the dose conversion factor of 0.7 Sv Gy^{-1} with an outdoor occupancy factor of 0.2 and 0.8 for indoor UNSCEAR (2000)

$$(AEDE)_{outdoor} = D(nG.h^{-1}) \times 8760(h.y^{-1}) \times 0.7 \times (10^3 mSv/nGy\ 10^9) \times 0.2 \qquad (7)$$

Equation (7) simplifies into $(AEDE)_{outdoor} = D \times 1.226 \times 10^{-3}$ $(mSv.y^{-1})$

$$(AEDE)_{indoor} = D(nG.h^{-1}) \times 8760(h.y^{-1}) \times 0.7 \times (10^3 mSv/n\ Gy 10^9) \times 0.8 \qquad (8)$$

Equation (8) simplifies into $(AEDE)_{indoor} = D \times 4.905 \times 10\text{-}3$ $(mSv.y^{-1})$

To estimate annual effective doses, account must be taken of (a) the conversion coefficient from absorbed dose in air to effective dose and (b) the indoor occupancy factor. The change in the average values of the radiological parameters depends on the population age. In addition, it changed with climate at the location considered. From UNSCEAR (2000) , the outdoor dose conversion coefficient was 0.7 Sv Gy^{-1} and it was used to change the absorbed dose in air to effective dose

received by adults. Furthermore, the conversion coefficient was 0.8 Sv Gy^{-1} for indoor occupancy factor, *i.e.,* the percentage of time spent indoor and outdoor was 80% and 20%, respectively. The average of worldwide annual effective dose was 0.48 mSv (0.3 - 0.6 mSv). For children and infants, the values were about 10% and 30% high, in direct proportion to an increase in the value of conversion coefficient from absorbed dose in air to effective dose (UNSCEAR, 2000).

Excess lifetime cancer risk (ELCR) was calculated by the following using equation :

$$ELCR=AEDE \times DL \times RF \quad (9)$$

Where, DL is duration of life (70 year) and RF is risk factor (Sv^{-1}), fatal cancer risk per Sievert. For stochastic effects, ICRP-60 (1991) uses values of 0.05 for the public. Using equation 6 for AEDE= 22.03 $nGyh^{-1}$ total annual effective gamma dose (terrestrial plus cosmic) was calculated as 0.22 mSv.

Results and Discussion

The concentrations of ^{226}Ra, ^{232}Th and ^{40}K radionuclides in soil samples collected from different parts of the studied area of Wadi Al-Rummah in Qassim province is shown in Table 1. ^{226}Ra activity concentration in soil samples ranged from 4.3 Bq kg^{-1} (Al Badaya) to 33.8 Bq kg^{-1} (Al-Rass) with a mean of 14.1±0.9 Bq kg^{-1}. Furthermore, ^{232}Th activity ranged between 5.3 Bq kg^{-1} (Al Badaya) and 45 Bq kg^{-1} (Al-Rass) with a mean of 15.5±1. ^{40}K ranged from 38.5 Bq kg^{-1} (Al Badaya) to 273 Bq kg^{-1} (Qassr Ibn Okial) with a mean of 546±23 Bq kg^{-1}, respectively. The concentration of ^{232}Th and ^{226}Ra in all the measured soil was lower than the concentration of ^{40}K. According to the recommended reference level of 30, 25 and 370 Bq kg^{-1} for ^{226}Ra, ^{232}Th, and ^{40}K respectively, for the World average concentrations published by UNSCEAR (2000), it is noted that the obtained results in most samples were lower than the recommended reference level.

A comparison between the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the present study with similar studies carried out in other countries are presented in Table 2. The mean values of ^{232}Th, ^{226}Ra and ^{40}K in the present study were lower than the recommended values for soil in Yemen, Jordan, Thailand, Algeria, China, Turkey and Sweden. However, ^{232}Th was higher than the reported for Jeddah in Saudi Arabia.

However, the mean values of ^{232}Th, ^{226}Ra and ^{40}K were lower than reported from Albaha, Saudi Arabia. A comparison of ^{40}K activity concentration showed that the value of this radionuclide in soil of United State, Yemen, Nigeria, Algeria and China were lower than the mean value of present study. Variation in the concentrations of radioactivity in soil of various locations of the world depend on the geological and geographical conditions of the area and the extent of fertilizer applied on the agriculture lands (Tzortzis *et al.,* 2003).

Variations in the concentrations of radioactivity in the soil of various locations of the world depend upon the geological and geographical conditions of the area and the extent of fertilizer applied to the agriculture lands (El-Taher and Abbady, 2012; El-Taher and Makhluf, 2011).

The concentration of naturally occurring radionuclides in soil depends on the rock type from which the soil is formed. Soil is either contaminated by radionuclide deposition originally discharged into the atmosphere, or on the land surface by direct discharge of the wastes. The concentration of radionuclides in soil increases by adsorption with soil particles and their precipitation on soil. The concentration decreases by leaching process and also dilutes when organic matter and soil water content increases behavior of radionuclides in soil on site characteristics, rate and amount of rain-fall and soil drainage (NCRP, 1991).

The physico-chemical form of radionuclide strongly affects its retention by soil particles and its availability for uptake by plants. The soil type strongly affects the behavior of radionuclides in soil, and soil retention characteristics (Schulz, 1965). Sandy soils do not have retention capacity of clay soils. Clay soils are composed of smaller particle sizes with larger surface area and negative charge surfaces (Nelson *et al.,* 1966).

Gamma- ray radiation hazards due to specified radionuclides ^{226}Ra, ^{232}Th and ^{40}K were assessed. The most widely used radiation hazard index Ra_{eq} is called the radium equivalent activity Ra_{eq} Beretka and Mathew (1985). Ra_{eq} is a weight sum of activities of the above three radionuclides based on the estimation that 370 Bqkg^{-1} of ^{226}Ra , 259 Bqkg^{-1} of ^{232}Th and 4810 Bqkg^{-1} of ^{40}K produced same ?-ray dose rates. Ra_{eq} is given by

$$Ra_{eq}=A_{Ra}+(A_{Th} \times 1.43) + (AK \times 0.077) \quad (2)$$

A value of 370 Bqkg^{-1} corresponds to 1 mSv y^{-1}. The radium equivalent concept allows a single index or number which is widely used hazard index to describe the gamma output from different mixtures of uranium, thorium and potassium in soil samples from different locations. The calculated values varied from 15.43 Bq kg^{-1} (Al Badaya) to 105.34 (Al-Rass) Bq kg^{-1} with an average of 45.53 Bq kg^{-1} (Table 3). These values were lower than the permissible maximum value of 370 Bq kg^{-1}.

Table 1 : Average activity concentrations (^{226}Ra, ^{232}Th and ^{40}K) in soil samples collected from Wadi Al-Rummah in Qassim province, Saudi Arabia

Location	Average (rage) in (Bq kg^{-1})		
	^{226}Ra	^{232}Th	^{40}K
Odia	11.2(10.5 - 11.9)	11.4(10.6-12.4)	143.2(127.9 -97.4)
Al Badaya	4.4(4.3-4.6)	5.5(5.3- 5.6)	41.5(38.5- 43.8)
Alkhabra	13.2(12.8- 13.5)	14.0(13.7–14.5)	125(120.2–130.6)
Thuayrat	6.1(5.9 – 6.2)	6.6(6.4 – 6.6)	61.9(61.9 -61.9)
Unaizah	11.1(10.8 - 11.5)	11.5(11.4–11.6)	99.4(99.4 – 99.4)
Qassr Ibn Okial	17(13.6 -20.5)	15.2(9.2 -21.5)	235.8(213.7-273.1)
Al-Batin	17.8(17.3 -18.3)	20(19.4 – 20.3)	137.1(131.2–140.1)
Al-Rass	30.2(28.1-33.8)	39.9(36.8-45.0)	239.5(226.6-254.0)
All Areas	14.1(4.3 -33.8)	15.5(5.3– 45.0)	143.1(38.5-273.1)

Table 2 : Comparison of natural radioactivity concentration (Bq kg^{-1}) in soil samples of present study with previous reports of different countries of the world

Location	Average activity concentration			References
	^{226}Ra	^{232}Th	^{40}K	
Qassim, KSA	14.1	15.5	143.1	Present study
Riyadh KSA	14.5	11	225	(Alaamer *et al*., 2008)
Albaha, KSA	37	32	343	(Al-Zahrani, 2012)
Tourbh, KSA	4.35	3.3	71.74	(Alharbi, 2012)
Makkah,KSA	13.3	15.6	514.3	(Hamidalddin *et al*., 2012)
Oman	14.42	9.95	158.21	(Saleh, 2012)
Yemen Juban	44	58	823	(Abd El-Mageed *et al*., 2011)
Egypt Assiut	17	18	320	(UNSCEAR, 2000)
Sweden	42	42	680	(UNSCEAR, 2000)
Japan	33	28	400	(UNSCEAR, 2000)
Nigeria Minna	7.8	29.4	229	(Kolo *et al*., 2011)
Nigeria Agbabu	9.38	8.64	65.75	(Isinkaye, 2008)
China	32	41	440	(UNSCEAR, 2000)
India	41	29	400	(UNSCEAR, 2000)
United State	40	35	370	(UNSCEAR, 2000)
World	32	45	420	(UNSCEAR, 2000)

The maximum value of H_{ex} to be less than unity corresponds to the upper limit of Ra_{eq} (370 Bqkg^{-1}). If the highest radium concentration value was less than the normal acceptable by factor half, then H_{in} value would be less than 1. The first goal of this index due to calculate dose equivalent less than 1 mSv per year. The values of indoor radiation hazard index (H_{in}) and outdoor radiation hazard index (H_{ex}) were from 0.01 to 0.05 and from 0.01 to 0.32, respectively. In addition, the average values of H_{in} and H_{ex} were 0.02. The values were less than the permissibly limit value, of unity. Therefore, these results showed that there was no health hazard due to gamma radioactivity. External and internal hazard indices calculated for soil samples are presented in Table 3.

The absorbed dose rate expresses the received dose in open air from the radiations emitted from radionuclides concentration in environmental materials. Also, it is the first major step for evaluating the health risk and is expressed in gray (Gy). The calculated total absorbed dose and annual effective dose rates of samples are tabulated in Table 4. It was observed that the absorbed dose rate calculated from activity concentration of ^{226}Ra, ^{232}Th and ^{40}K ranged between 1.8 to 14.4, 3.5 to 29.8 and 1.7 to 11.8 nGy h^{-1}, respectively, where the upper limit was observed in Al-Rass and lower limit in Al Badaya location. The total absorbed dose in the study area ranged from 7.0 nGy h^{-1} (Al Badaya) to 55.2 nGy h^{-1} (Al-Rass) with an average value of 22.0 nGy h^{-1}, which was lower than the limits recommended by ICRP-65 report (ICRP-60, 1991). The relative contribution to dose due to ^{232}Th was 45.8 %, followed by the contribution due to ^{40}K and ^{226}Ra as 27.3 %, 26.9 % respectively.

As shown in Table 4, the annual effective dose equivalent from outdoor terrestrial gamma radiation ranged from 0.01 mSvy^{-1} (Thuayrat and Al Badaya) to 0.06 mSvy^{-1}

Table 3 : Average radiological hazards (external hazard index (Hex) , internal hazard index (Hin) , representative hazard index (Iãr) and radium equivalent activity Bq/Kg (Raeq)) in soil from Wadi Al-Rummah in Qassim province

Location	Radiological hazards			
	Raeq	Hex	Hin	Iãr
Odia	38.51	0.10	0.13	0.28
Unaizah	35.19	0.10	0.13	0.26
Thuayrat	20.26	0.05	0.07	0.15
Qassier Ibn Okial 56.88	0.15	0.20	0.42	
Al Rass	105.34	0.28	0.37	0.76
Alkhabra	42.86	0.12	0.15	0.31
Al Badaya	15.43	0.04	0.05	0.11
Al Batin	57.12	0.15	0.20	0.41
Average	46.45	0.13	0.16	0.16
Maximum	117.67	0.32	0.41	0.76
Minimum	14.85	0.04	0.05	0.11

Table 4 : Absorbed dose rates and annual effective doses calculated for surface soil samples collected from Qassim province

Location	Absorbed rates (nG/h)				Annual effective dose (mSv)	
	^{226}Ra	^{232}Th	^{40}K	Total	AEDEoutdoor	AEDE indoor
Odia	4.80	7.53	6.18	18.5	0.02	0.09
Unaizah	4.73	7.61	4.30	16.65	0.02	0.08
Thuayrat	2.60	4.36	2.67	9.63	0.01	0.05
Qassier Ibn Okial	7.24	10.08	10.19	27.51	0.03	0.13
Al Rass	12.85	26.30	10.34	49.49	0.06	0.24
Alkhabra	5.63	9.29	5.40	20.31	0.02	0.1
Al Badaya	1.87	3.63	1.80	7.30	0.01	0.04
Al Batin	7.61	13.28	5.95	26.84	0.03	0.13
Average	5.92	10.26	5.85	22.03	0.03	0.11
Maximum	14.4	29.8	11.8	55.2	0.06	0.24
Minimum	1.8	3.5	1.7	7.0	0.01	0.04

Table 5 : Annual effective gamma doses and excess lifetime risks of cancer

Location	Dose rate (mSv/h) 10^{-4}	Annual dose rate µSv/y	Life total dose mSv	Exess lifetime cancer risk (ELCR)		
				Min 10^{-5}	Max 10^{-5}	Mean 10^{-5}
Odia	0.19	22.7	1.59	7.3	8.6	7.9
Unaizah	0.17	20.4	1.43	7.1	7.3	7.1
Thuayrat	0.10	11.8	0.83	4.1	4.2	4.1
Qassr Ibn Okial	0.28	33.7	2.36	10.0	15.0	12.0
Al Rass	0.50	60.7	4.25	20.0	24.0	21.0
Alkhabra	0.20	24.9	1.76	8.5	9.0	8.7
Al Badaya	0.07	9.0	0.63	3.0	3.2	3.1
Al Batin	0.27	32.9	2.31	11.0	12.0	12.0
Average	0.22	27.03	1.89	3.0	24.0	9.5
Max	0.55	60.72	4.25			
Min	0.07	8.96	0.63			
(El-Taher and Al-Zahrani, 2014)	0.46	43	3.02	5	18	9
world	0.46	0.08	5.67			29

(Al-Rass) with a mean value of 0.03 mSvy^{-1}. Also, for indoor exposure, the annual effective dose equivalent ranged from 0.04 mSvy^{-1} (Al Badaya) to 0.24 mSvy^{-1} (Al-Rass) with a mean value of 0.11 mSvy^{-1}. The corresponding world average value was 0.41 mSvy^{-1} of which 0.07 mSvy^{-1} was from outdoor and 0.34 mSvy^{-1} from indoor exposure.

Excess lifetime cancer risk calculated was $0.20x10^{-3}$. When life expectancy was taken as 70 years, the average lifetime outdoor gamma radiation was fount to be 1.89 mSv which yielded an average lifetime cancer risk of 9.5×10^{-5}. Potential carcinogenic effects were characterized by estimating the probability of cancer incidence in a population of individuals for specific life time from projected intake (and exposures) and chemical-specific dose-response data (*i.e.*, slope factors).

The measured dose values were lower than the international dose limit in the soil. The mean value of total absorbed dose rate was 22.03 nGy h^{-1} and the population-weight (world average) value is 65 nGy h^{-1}. In addition, the annual effective gamma doses and the lifetime hazards of cancer were lower than the international limit. The value of radium equivalent (Ra_{eq}) was less than 370 Bq kg^{-1}, the external hazard index (H_{ex}), internal hazard index (H_{in}) and representative level index ($I_{\gamma r}$) were found to be less than the acceptable limit of unity.

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