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Gamma Spectroscopic of Soil Samples from Kufa in Najaf Governorate, Iraq

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Abstract: Natural radioactivity measurements and assessment of radiological hazards in soil samples obtained from Najaf governorate, Iraq were carried out using the NaI (Tl) detector system. The activity concentrations of ^{238}U, ^{232}Th and ^{40}K were found to be 78±18, 78±20 and 286±65 Bq kg^{-1}, respectively and the external gamma dose rate is 112±24 nGy h^{-1}. The activity concentrations for ^{238}U, ^{232}Th and ^{40}K in soil samples in the present study were within the activity values for other regions around the world. To assess the radiological hazard of radioactivity in the samples, the radium equivalent activity, annual effective dose, external hazard and internal indices were calculated. The Ra_{eq} values of soil samples were lower than 370 Bq kg^{-1}.

Key words: Environmental radioactivity · Scintillation detector · Soil · Gamma dose rate

INTRODUCTION

Natural environmental radioactivity emitted by gamma-ray depends on the geological, soil types and geographical conditions [1]. Natural radioactivity originates from extraterrestrial sources and radioactive elements in the Earth's crust. The natural radioactivity may vary considerably from one type of soil to another. Radionuclides are found naturally in air, water and soil. Radionuclides can be subdivided into primordial, cosmogenic and manmade [2]. Considering the radiation exposure of man, only some of these natural radionuclides are important. The external radiation exposure is mainly caused by ^{238}U, ^{232}Th and ^{40}K in soil on the Earth's surface. When a radionuclide comes in contact by soil, it can adsorb to reactive coatings on particles, undergo ion exchange, precipitate as an oxide-hydroxide or sulfide and be complexed with organic compounds, or remain in ionic form [3]. The radionuclides to be measured and sample types to be analyzed can differ depending on the field of science-a radiopharmaceutical to be administered of a cancer patient, radionuclides in air, water and soil samples taken from the environment or radioactive waste from a nuclear power plant serve as example. Gamma-ray spectrometer method for the determination of naturally occurring radioactive materials (NORM) is very interested in Earth's sciences [4, 5]. There are a number of possible applications spanning from ore exploration to environmental radiation monitoring problems, most of them involving the determination of the U, Th and K amount in soil [1].

These elements may be used as tracers and also in non-radioactive processes producing Para genesis associated with NORMs [6]. Radioactivity in the materials are released to and dispersed in the atmosphere enters the terrestrial environment due to dry and wet deposition on soil and vegetation. Internal exposures to humans occur from the use of contaminated plants as food and as feed for domestic animal. Direct contamination of foliage occurs over the long term-uptake of radionuclides from soil and deposition of resuspended contaminated soil particles [7]. The global average dose rate is 0.057 $mGyh^{-1}$. Maximum values have been measured in Guarapari, Brazil (50 $mGyh^{-1}$) and in Kerala, India (2 $mGyh^{-1}$) and on rocks with a high radium concentration in Ramsar, Iran (10 $mGyh^{-1}$) [8]. The study area is Najaf governorate in Iraq about 160 km south of Baghdad. Its estimated population in 2008 is 900,600 people. The Najaf area is located 30 km south to the ancient city of Babylon and 400 km north to the ancient Biblical city of Ur. Kufa is a city in Iraq, about 170 km south of Baghdad and 10 km northeast of Najaf. It is located on the banks of the Euphrates River. The estimated population in 2003 was 110,000. During the 2003 invasion

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of Iraq, Najaf was a key target of the invading United States forces. The city was encircled during heavy fighting on March 26, 2003 and was captured on April 3, 2003 by 1st, 2nd, 3rd Battalions, 327th Infantry Regiment, units of the 101st Airborne Division. Battle of Najaf in 2003 was fierce battle with USA army, where U.S. airborne and armored units with British air support fought an intense battle with Iraqi Regulars, Republican Guard units and paramilitary forces. AH-64 Apache helicopter gunships and with support from F/A-18 Hornet jets, setting out on a mission to attack Republican Guard armored units; while flying low the Apaches came under heavy anti-aircraft, arms and RPG fire which heavily damaged many helicopters and shot one down, frustrating the attack [9, 10]. The 1st Brigade Combat Team's air defense battery moved in and after heavy fighting with entrenched Iraqi Fedayeen seized a strategic bridge in Najaf [11]. The 101st Airborne Division, supported by a battalion from the 1st Armored Division, attacked Iraqi forces in the southern part of the city, near the Imam Ali Mosque and captured Najaf's airfield [12].

Therefore, Najaf governorate needs to more details about radioactivity in the air, water and soil because this war. Many studies have been performed to measure the concentrations of radionuclides (NORM) and gamma dose rate in soil samples from different cities in Iraq using different techniques [13-18]. This study aims to assess the natural radioactivity levels and radiological hazards from soil samples. The gamma radiation levels measurements in different areas of Najaf governorate are useful for establishing base line data for assessment of radiation exposures to the population.

MATERIALS AND METHODS

Five soil samples were collected from different areas in Kufa in Najaf governorate on May-Jun 2009 as shown in Fig. 1.

The samples were dried and pulverized. Each sample was then weighed and sealed in Marinelli beaker. The gamma spectroscopy system was used for the quantitative and qualitative determination of radionuclides. Gamma spectroscopic measurement was performed using a NaI (Tl) detector (Spectrum Techniques, INC., USA) with diameter 1.76" and thickness 1.56". The system has efficiency of about 50% and a energy resolution (FWHM) of about 7.5% at energy at 662 keV (^{137}Cs) which is considered adequate to distinguish the gamma-ray energies of interest in this study. The detector is surrounded by a lead shielding in 5 cm thickness. A constant counting time for calibration sources (^{60}Co, ^{137}Cs, ^{22}Na, ^{241}Am and ^{226}Ra) from the International Atomic Energy Agency, for the background spectrum and for measuring soil of 3600 s was adopted. Instrument calibration was done at multiple energies from 25 keV to 2500 keV. The natural radioactivity of soil samples is determined from the ^{238}U, ^{232}Th and ^{40}K contents. Since 98.5% of the radiological effects of the uranium series are produced by radium and its daughter products and the contribution from the ^{238}U and the daughter ^{226}Ra precursors are normally ignored. The relevance of naturally occurring radionuclides with the present work is mainly gamma ray emitting nuclei of ^{238}U, ^{232}Th and a single occurring ^{40}K. The concentration of ^{40}K can be measured directly by its own gamma ray at

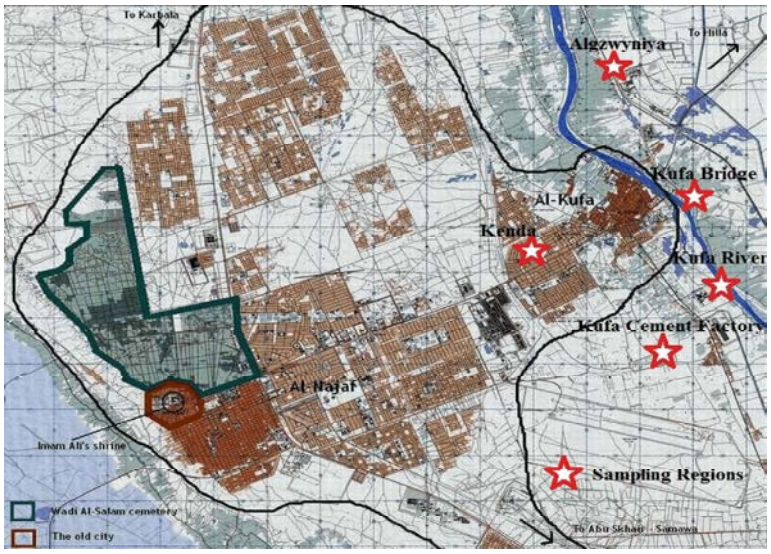

Fig 1: The administrative Najaf map

Table 1: The γ-transitions used to measure the activity concentrations of ^{238}U, ^{232}Th and ^{40}k.

^{238}U-series	Energy (kev)	Iγ	^{232}Th-series	Energy (kev)	Iγ
^{234}Th $T_{1/2}$=24.1 d	63	0.039	^{228}Ac $T_{1/2}$=6.15 h	338.32	0.113
^{226}Ra $T_{1/2}$=1602 y	186.20	0.035		463.01	0.044
^{214}Pb $T_{1/2}$=26.8 m	295.21	0.185		911.21	0.266
	351.72	0.358		968.96	0.213
^{214}Bi $T_{1/2}$=19.9 m	609.31	0.448	^{212}Pb $T_{1/2}$=10.64 h	238.63	0.433
	768.36	0.048	^{212}Bi $T_{1/2}$=60.55 m	727.33	0.065
	934.06	0.030			
	1120.29	0.148	^{208}Ti $T_{1/2}$=3.053 m	583.19	0.487
	1238.11	0.058	^{40}K	1460.8	0.107

1461 keV, while activity of ^{238}U and ^{232}Th were calculated based on the average activities of their decay products as shown in Table 1 [19, 20].

The specific activity is defined as follows [21]:

$$\text{Specific activity } (Bq\ kg^{-1}) = \frac{Net\ \ Area - B.G}{t\varepsilon P_{\gamma} M}, \quad (1)$$

where Net Area= Net area under energy peak (count).

$B.G$ = the number of counts for the background spectrum, ε = the absolute efficiency of the detector and M = the weight of the dried sample (kg).

Radiological Hazard Index

Radium Equivalent Activity (Ra_{eq}): The significance of ^{226}Ra, ^{232}Th and ^{40}K concentrations was defined in terms of radium equivalent activity in Bq kg^{-1}. Ra_{eq} was calculated from equation [22]:

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_{K} \quad (2)$$

where C_{ra}, C_{th} and C_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq kg^{-1}, respectively. This equation is based on the estimate that 1 Bq kg^{-1} of ^{226}Ra, 0.7 Bq kg^{-1} of ^{232}Th and 13 Bq kg^{-1} of ^{40}K generate the same gamma-ray dose rate [23].

The maximum value of Ra_{eq} must be less than 370 Bq kg^{-1} for safe use as recommended by the Organization for Economic Cooperation and Development [24].

Air-Absorbed Dose Rates: The absorbed dose rates in outdoor air (D_R), at about 1 m above the ground surface were calculated. The conversion factors used to compute absorbed gamma-ray dose rate in air corresponds to 0.46 nGy h^{-1} for ^{226}Ra, 0.62 Gy h^{-1} for ^{232}Th and 0.042 nGy h^{-1} for ^{40}K. Therefore, D can be calculated according to [1] using equation

$$D_R\,(nGy\ h^{-1}) = 0.46\ C_{Ra} + 0.62\ C_{Th} + 0.042\ C_K \quad (3)$$

Annual Outdoor Effective Dose Equivalent: To estimate the annual outdoor effective doses (ED), the conversion coefficient from absorbed dose rate in air to effective dose (0.7 Sv Gy^{-1}) and the outdoor occupancy factor (0.2) are used [1]. The effective dose equivalent rate is calculated from equation [22]:

$$ED\ (mSv\ y^{-1}) = D_R \text{ x } 8766\ h\ y^{-1} \text{ x } 0.7\ (Sv\ Gy^{-1)} \text{ x } 0.2 \text{ x } 10^{-3} \quad (4)$$

External Hazard Index (H_{ex}): Radiation exposure due to ^{226}Ra, ^{232}Th and ^{40}K may be external. This hazard, defined in terms of external or outdoor radiation hazard index and denoted by H_{ex}, can be calculated using equation [22]:

$$H_{ex} = C_{Ra}/370 + C_{Th}/259 + C_K/4810 > 1 \quad (5)$$

Internal Hazard Index (H_{in}): Internal hazard index (H_{in}) is given by equation [22]:

$$H_{in} = C_{Ra}/185 + C_{Th}/259 + C_K/4810 \quad (6)$$

H_{in} must be less than one for safe use of samples and for the radiation hazard to be negligible.

RESULTS AND DISCUSSION

Activity levels of U, Th and K of the various soil samples were presented in Table 2.

Soil ^{238}U, ^{232}Th and ^{40}K in the study area were found to be 78±18, 78±20 and 286±65 Bq kg^{-1}, respectively. From Table 2, the higher ^{238}U and ^{232}Th concentrations in soil samples are noted in site K_C in Kufa Cement Factory and site K_R in Kufa River, respectively and the higher ^{40}K concentration noted in site K_E in Kenda, whereas the lower ^{238}U, ^{232}Th concentrations are noted in site K_E in Kenda and the lower ^{40}K concentration noted in site K_B in Kufa Bridge. The world average concentrations are

Table 2: Average activity level (BqKg^{-1}) in the soil samples

SC	Site	^{238}U	^{232}Th	^{40}K
K_C	Kufa Cement Factory	102±17	80±19	261±92
K_R	Kufa River	80±18	92±19	288±56
K_B	Kufa Bridge	81±17	76±21	243±61
K_E	Kenda	55±15	64±19	369±50
A_L	Algzwyniya	70±23	78±22	270±68
Avg.		78±18	78±20	286±65

SC= Site code

Table 3: Radiation hazard indices of soil samples

Sc	Ra_{eq} (Bq kg^{-1})	D_R (nGy h^{-1})	ED (mSv y^{-1})	H_{ex}	H_{in}
K_C	266	121	0.15	0.72	1.08
K_R	283	129	0.16	0.76	1.11
K_B	244	111	0.14	0.66	0.97
K_E	213	98	0.12	0.58	0.83
A_L	222	102	0.13	0.60	0.84
Avg.	246±51	112±24	0.14±0.03	0.66±0.14	0.97±0.19

Table 4: Comparison of natural radioactivity levels in soil (Bq kg^{-1}) at different sites with those in other countries

	^{40}K		^{238}U		^{232}Th	
Country	Mean	Range	Mean	Range	Mean	Range
United States [1]	370	100-700	35	4-140	35	4-130
Armenia [1]	360	310-420	46	20-78	30	29-60
Bulgaria [1]	400	40-800	40	8-190	30	7-160
Croatia [1]	490	140-710	110	83-180	45	12-65
India [1]	400	38-760	29	7-81	64	14-160
Japan [1]	310	15-990	29	2-59	28	2-88
Greece [1]	360	12-1570	25	1-240	20	1-190
Portugal [1]	840	220-1230	49	26-82	51	22-100
Russia [1]	520	100-1400	19	0-67	30	2-79
Spain [1]	470	25-1650	---	---	33	2-210
Norway [1]	---	114-643	---	17-134	---	10-52
France [26]	---	348-802	---	28-53	---	22-42
Hungary [27]	---	176-567	---	0-1346	---	15-41
Argentina [28]	---	568-817	---	---	---	35-48
Malaysia [29-33]	615	87-1827	133	2-799	133	6-667
Iraq (Present Study)	286±65	243-369	78±18	55-102	78±20	64-92

Table 5: Radiation Hazard indices of soil samples compared with the values reported from other countries

Country	Ra_{eq} (Bq kg^{-1})	D_R (nGy h^{-1})	ED (mSv y^{-1})	H_{ex}	H_{in}
Nigeria [34]	50-110	23-52	0.06-0.02	0.29-0.14	0.18-0.37
Serbia [35]	---	92-316	---	…..	---
Jordan [36]	12-702	45-71	0.05-0.08	0.87-4	---
Yemen [37]	191	89	---	0.52	---
Bangladesh [38]	77-151	74-35	---	---	---
Egypt [39]	152	82	---	---	---
Thailand [40]	---	81-90	0.10-0.11	---	---
China [41]	230-676	86-237	0.10-0.29	0.60-1.80	…..
Malaysia [29-33]	127-1103	125-496	0.07-0.60	0.34-2.90	0.48-4.01
Iraq (Present Study)	213-283	98-129	0.12-0.16	0.58-0.76	0.83-1.11

35 and 45 Bq kg^{-1} for ^{238}U and ^{232}Th, respectively. The typical ranges are 16 Bq kg^{-1} to 116 Bq kg^{-1} for ^{238}U and 7 Bq kg^{-1} to 50 Bq kg^{-1} for ^{232}Th. The world average concentration is 420 Bq kg^{-1} for ^{40}K and the typical range is 100 Bq kg^{-1} to 700 Bq kg^{-1} for ^{40}K [1]. The average value of Ra_{eq} in the study area is 246±51 Bq kg^{-1} as shown in Table 3, which are less than the 370 Bq kg^{-1} recommended maximum levels of radium equivalents

in soil [24]. Therefore, the soil is suitable for use for agriculture and building materials. The average absorbed dose rate is 112±24 nGy h^{-1} for soil samples. This value is about two times higher than the world average dose rate of 55 nGy h^{-1} [1]. The outdoor annual effective doses ranged from 0.12 mSv y^{-1} to 0.16 mSv y^{-1} with a mean value of 0.14±0.03 mSv y^{-1} in soil, while the worldwide average annual effective dose is 0.5 mSv y^{-1} and the results for individual countries being generally within the ranges from 0.3 mSv y^{-1} to 0.6 mSv y^{-1} [1]. The calculated external hazard values are between 0.58 H_{ex} to 0.76 H_{ex} (mean = 0.66 H_{ex}). The value of H_{in} ranged from 0.83 H_{in} to 1.11 H_{in} (mean = 0.97 H_{in}) for soil samples. The values of H_{ex} in sampling sites K_C and K_R are higher than unity, which may cause harm to people in this region.

Tables 4 and 5 summarize the natural radioactivity levels and radiation hazard indices in soils obtained in some world regions as well as this study. The activity levels of ^{238}U, ^{232}Th and ^{40}K in the present study were within the activity range of radionuclides in other listed regions. The values of Ra_{eq}, D_R, ED, H_{ex} and H_{in} are also within the values reported in other listed regions.

CONCLUSIONS

It is concluded that the activity concentrations for ^{238}U, ^{232}Th and ^{40}K in soil samples in the present study were within the activity values for other regions around the world. The low concentrations of ^{238}U, ^{232}Th and ^{40}K measured in soil samples suggest their suitability for use as building materials. The levels of natural radioactivity in the study areas were within normal values (H_{ex} and H_{in}< 1), except those in Kufa Cement Factory and Kufa River samples. The average absorbed dose rate calculated from the soil samples was 112 nGy h^{-1}. This value is about two times higher than the world average dose rate of 55 nGy h^{-1}.

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