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Research Note

NaI (Tl) Spectrometry to Natural Radioactivity Measurements of Soil Samples in Najaf City

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A B S T R A C T

This study conducted using a NaI (Tl) gamma-ray spectrometer for the assessment of naturally radioactive materials. This apparatus is devoted to the quantitative and qualitative determination of U, Th, and K in soil samples collected from the city of Najaf, Iraq. The average of concentrations in the surveyed soil samples were ranged from 55 to 102 Bq kg^{-1}, ND to 448 Bq kg^{-1} and 79 to 1887 Bq kg^{-1} for ^{238}U, ^{232}Th, and ^{40}K, respectively. To assess the radiological hazard of radioactivity in the soil 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} recommended maximum levels of radium equivalents in soil.

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INTRODUCTION

The Gamma-ray spectrometer method for the determination of naturally radioactive materials is big interest in environmental and Earth's sciences [1-13]. 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 and rocks [14]. These elements may be used as tracers also in non-radioactive processes producing Para genesis associated with naturally occurring radioactive materials (NORMs) [15]. The main objective of this study was to identify and determine natural radionuclide concentrations in soil samples collected from Najaf city.

MATERIALS AND METHODS

Eleven soil samples are collected from Najaf city as shown in Fig. 1. These regions are Alansar-Najaf (7 samples), Alfateh-Hurya (3 samples), and Alrashadia-Kufa (1 sample).

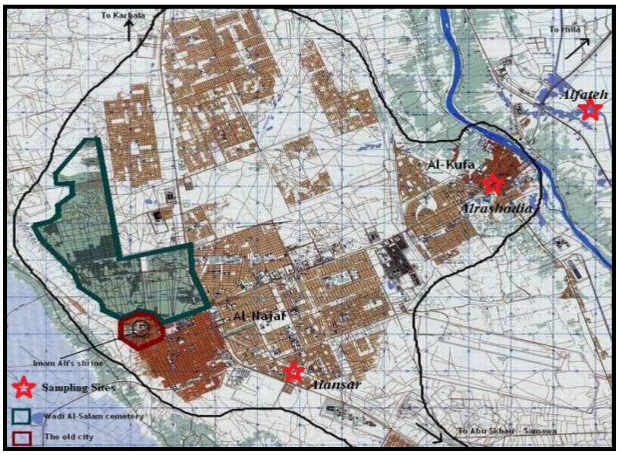

Figure. 1. The administrative Najaf city map with sampling sites

The samples were dried, homogenized, and weighted. Each sample (1 kg) was sealed in Marinelli beaker. Gamma spectroscopic measurements using a NaI (Tl) scintillation detector (1.76" × 1.56") and a leybold cassy lab multichannel analyzer (Pocket-CASSY 524058) were performed. The detector is surrounded by a lead shielding in 5 cm thickness. A

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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 to 2500 keV. The natural radioactivity of soil samples is usually determined from the ^{238}U, ^{232}Th, and ^{40}K contents. It is worth to mention that about 98.5% of the radiological effects of ^{238}U are produced by radium and its daughter products. The contribution from the ^{238}U and the daughter ^{226}Ra precursors are ignored. The naturally occurring radionuclides of relevance for the present work are mainly gamma ray emitting nuclei of ^{238}U, ^{232}Th, and ^{40}K. Activity concentration of ^{40}K can be measured directly by its own gamma ray at 1461 keV, whereas, activity of ^{238}U and ^{232}Th were calculated based on the average activities of their respective decay products (Table 1) [10, 16, 17].

The specific activity is defined as follows [18, 19]:

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

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, P_{γ} is the gamma-ray emission probability, 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 the following equation [20]:

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

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 [21]. 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 [22].

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 literature [14] using the following equation

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

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 [14]. The effective dose equivalent rate is calculated from the following equation [20].

$$ED\,(mSv\ y^{-1}) = D_R \times 8766\ h\ y^{-1} \times 0.7\ (Sv\ Gy^{-1)} \times 0.2 \times 10^{-3} \qquad (4)$$

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 the following equation [20]:

$$H_{ex} = C_{Ra}/370 + C_{Th}/259 + C_{K}/4810 \leq 1 \qquad (5)$$

Internal hazard index (H_{in})

Internal hazard index (H_{in}) is given by the following equation [20]:

$$H_{in} = C_{Ra}/185 + C_{Th}/259 + C_{K}/4810 \leq 1 \qquad (6)$$

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 ^{238}U, ^{232}Th, and ^{40}K of the various soil samples were determined as shown in Table (2).

Soil ^{238}U, ^{232}Th, and ^{40}K in the study area were found to be 69.78±0.53, 125.63±0.47, and 1165.29±0.45 Bq kg^{-1}, respectively. From Table 2, the higher ^{238}U and ^{232}Th concentrations in soil samples are noted in site ANS2 and site FAT3, respectively. The high ^{40}K concentration was noted in site ANS6. Whereas, the low ^{238}U, ^{232}Th the low ^{40}K concentration is noted in site FAT2. The

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

Radionuclides of interest	Measured radionuclides	Photon intensity %	Energy (keV)
^{232}Th	^{212}Pb $T_{1/2}$=10.64 h	43	238.63
	^{228}Ac $T_{1/2}$=6.15 h	11	338.32
^{238}U	^{226}Ra $T_{1/2}$=1602 y	3.5	186.20
	^{234}Th $T_{1/2}$=24.1 d	3.5	63
	^{214}Pb $T_{1/2}$=26.8 m	19, 36	295.21, 351.72
^{40}K	^{214}Bi $T_{1/2}$=19.9 m	45	609.31
	^{40}K	11	1461

world average concentrations are 35 and 45 Bq kg^{-1} for ^{238}U and ^{232}Th, respectively. The typical ranges are 16 to 116 Bq kg^{-1} for ^{238}U and 7 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 to 700 Bq kg^{-1} for ^{40}K [14]. The average value of Ra_{eq} in the study area is 339.16±1.25 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 [22]. Therefore, the soil is suitable for use for agriculture and building materials. The average absorbed dose rate is 163.57±0.56 nGy h^{-1} for soil samples. This value is about three times higher than the world average dose rate of 55 nGy h^{-1} [14]. The outdoor annual effective doses ranged from 0.06 to 0.50 mSv y^{-1} with a mean value of 0.2007±0.0006 mSv y^{-1} in soil; while the worldwide average annual effective dose is 0.5 mSv y^{-1}. The results for individual countries are being generally within the ranges from 0.3 to 0.6 mSv y^{-1}[14]. The calculated external hazard values are between 0.28 to 2.33 (mean = 0.92). The value of H_{in} ranged from 0.34 to 2.61 (mean = 1.10) for soil samples. The values of H_{ex} and H_{in} in some sampling sites are higher than unity, which may cause harm to people in these regions.

Tables 4 and 5 summarize the natural radioactivity levels and radiation hazard indices in soil 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.

TABLE 2. Average activity concentration in the soil samples

Location name	SC	Activity concentration Bq kg^{-1}		
		^{238}U	^{232}Th	^{40}K
Alansar	ANS1	27.96±0.65	44.44±0.42	1258.25±0.48
Alansar	ANS2	137.67±0.70	144.44±0.45	1022.23±0.45
Alansar	ANS3	45.54±0.48	21.51±0.36	1179.6±0.47
Alansar	ANS4	70.28±0.72	22.22±0.24	865.05±0.36
Alansar	ANS5	90.26±0.57	133.33±0.46	1494.18±0.59
Alansar	ANS6	34.72±0.58	ND	1887.30±0.54
Alansar	ANS7	25.51±0.59	11.11±0.22	786.41±0.44
Alfateh	FAT1	93.55±0.49	31.07±0.42	1022.33±0.33
Alfateh	FAT2	96.08±0.42	271.40±0.90	78.64±0.26
Alfateh	FAT3	102.46±0.41	447.71±0.97	1572.82±0.56
Alrashadia	RASH	43.55±0.32	254.78±0.77	1651.46±0.50
	Average	**69.78±0.53**	**125.63±0.47**	**1165.29±0.45**

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}
ANS1	188	96	0.1173	0.5087	0.5843
ANS2	423	199	0.2441	1.1422	1.5143
ANS3	167	84	0.1036	0.4513	0.5744
ANS4	169	82	0.1006	0.4555	0.6455
ANS5	396	192	0.2351	1.0693	1.3133
ANS6	180	96	0.1177	0.4862	0.5800
ANS7	102	52	0.0639	0.2753	0.3442
FAT1	217	105	0.1283	0.5853	0.8381
FAT2	490	226	0.2763	1.323	1.5835
FAT3	864	410	0.5026	2.3325	2.6094
RASH	535	259	0.3182	1.4447	1.5624
Average	**339.16±1.25**	**163.57±0.56**	**0.2007±0.0006**	**0.915±0.003**	**1.104±0.004**

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

Country	^{40}K		^{238}U		^{232}Th	
	Mean	Range	Mean	Range	Mean	Range
United States [14]	370	100-700	35	4-140	35	4-130
Armenia [14]	360	310-420	46	20-78	30	29-60
Bulgaria [14]	400	40-800	40	8-190	30	7-160
Croatia [14]	490	140-710	110	83-180	45	12-65
India [14]	400	38-760	29	7-81	64	14-160
Japan [14]	310	15-990	29	2-59	28	2-88
Greece [14]	360	12-1570	25	1-240	20	1-190
Portugal [14]	840	220-1230	49	26-82	51	22-100
Russia [14]	520	100-1400	19	0-67	30	2-79
Spain [14]	470	25-1650	---	---	33	2-210
Norway [14]	---	114-643	---	17-134	---	10-52
France [23]	---	348-802	---	28-53	---	22-42
Hungary [24]	---	176-567	---	0-1346	---	15-41
Argentina [25]	---	568-817	---	---	---	35-48
Malaysia [3, 6-10]	615	87-1827	133	2-799	133	6-667
Iraq [3]	286±65	243-369	78±18	55-102	78±20	64-92
Iraq (Present Study)	**1165.29±0.45**	**79-1887**	**69.78±0.53**	**26-137**	**125.63±0.47**	**ND-448**

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 [26]	50-110	23-52	0.06-0.02	0.29-0.14	0.18-0.37
Serbia [27]	---	92-316	---	---	---
Jordan [28]	12-702	45-71	0.05-0.08	0.87-4	---
Yemen [29]	191	89	---	0.52	---
Bangladesh [30]	77-151	74-35	---	---	---
Egypt [31]	152	82	---	---	---
Thailand [32]	---	81-90	0.10-0.11	---	---
China [33]	230-676	86-237	0.10-0.29	0.60-1.80	---
Malaysia [3, 6-10]	127-1103	125-496	0.07-0.60	0.34-2.90	0.48-4.01
Iraq [3]	213-283	98-129	0.12-0.16	0.58-0.76	0.83-1.11
Iraq (Present Study)	**102-864**	**52-410**	**0.06-0.50**	**0.28-2.33**	**0.34-2.61**

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 ANS2, ANS5, FAT2, FAT3, and ALRASHADIYA samples. The average absorbed dose rate calculated from the soil samples was 164 nGy h^{-1}. This value is about three times higher than the world average dose rate of 55 nGy h^{-1}. The high concentration of radium found to be in FAT3 and thus to consider the main reason for the presence of radon gas, which of role causes a direct radiation exposure by inhalation and thus the high concentration of thorium. The excessively use of chemical fertilizers and insecticides in FAT3 site, that contain high levels of radioactive isotopes.

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Persian Abstract

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چکیده

این مطالعه با استفاده از اسپکترومتر اشعه گاما NaI(T1) برای ارزیابی مواد رادیواکتیو طبیعی انجام شده است. این دستگاه برای تعیین کمی و کیفی اورانیوم، توریوم و پتاسیم در نمونه های خاک جمع آوری شده از شهر نجف عراق به کار گرفته شد. میانگین غلظت ها در نمونه های خاک بررسی شده در محدوده ی ۵۵ تا ۱۰۲ $Bq\ kg^{-1}$، ND تا ۴۴۸ $Bq\ kg^{-1}$ و ۷۹ تا ۱۸۸۷ $Bq\ kg^{-1}$ برای ^{238}U, ^{232}Th و ^{40}K بود. برای ارزیابی خطر رادیولوژیکی رادیواکتیویه در نمونه های خاک، فعالیت معادل رادیم، دوز موثر سالانه، خطر خارجی و شاخص های داخلی محاسبه شد. مقادیر Ra_{eq} نمونه های خاک کمتر از ۳۷۰ $Bq\ kg^{-1}$ معرفی شده سطح ماکزیمم مقادیر رادیم در خاک بود.