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Natural and Artificial Radionuclide Concentrations for different Environmental Samples in Samara City

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Abstract **- Gamma-ray spectrometer based on NaI(Tl) detector has been used to determine the specific activities of naturally occurring radioactive material (NORM) includes ^{238}U, ^{232}Th and ^{40}K in soil samples collected from samara city of Salahdin Governorate-Iraq at 2016. The specific activity of ^{226}Ra in the soil ranged from MDA to 246.96 Bq/kg with an average value 80.44Bq/kg, ^{228}Ac from MDA to 60.52 Bq/kg with an average value 24.90 Bq/kg, ^{40}K ranged from 4.4 to 226.36 Bq/kg with an average value 116.98 Bq/kg and ^{137}Cs ranged from MDA to 41.53 Bq/kg with an average value 10.17Bq/kg respectively. The specific activity of artificial radionuclide ^{137}Cs ranged from MDA to 41.53 Bq/kg with average value 10.17 Bq/kg. Radium equivalent, absorbed gamma dose rate, annual effective dose equivalent, the external hazard index, internal hazard index and activity concentration index were calculated and comparable with other global radioactivity measurements and found to be safe for public and environment.**

Keywords **- Natural Radioactivity, Specific Activity, Soil, Radiation hazards, NaI(Tl) Detector.**

I. INTRODUCTION

The exposure of human beings to natural ionizing radiation emitted by the primordial radionuclide ^{238}U, ^{232}Th and ^{40}K is a continuing and inescapable feature of life on earth; for most individuals, this exposure exceeds that from all man-made sourceswhich are important in radiological monitoring and radiation dose assessment for public [1]. These doses vary depending upon the concentrations of the natural radionuclides, ^{226}Ra, ^{232}Th, their daughter products and ^{40}K, present in the soils androcks, which in turn depend upon the local geology of each region in the world [2].

Natural radioactivity in soil and rocks measurementsare very important to determine the amount of change of the natural radioactivity background with time as a result of any radioactive release; monitoring of any release of radioactivity to the environments is important for environmental protection[3]. Uranium occurs as a natural constituent in soil, originating from rocks in the earth's mantle. Natural sources of uranium in ambient air include resuspension of soil and volcanic eruptions (ATSDR 1999), anthropogenic sources of airborne uranium include coal and fuel combustion [4]. The concentration of uranium in soil varies widely, but typically contains about 3 parts per million (ppm), or about 2 picocuries per gram (pCi/g) [1]. However, concentrations can vary considerably from place to place, depending on local geology and other factors. Estimation of the radiation dose distribution is important in assessing the health risk to a population and serve as reference in documenting changes to environmental radioactivity in soil due to anthropogenic activities [5]. The natural radioactivity of soil samples is usually determined from the ^{238}U, ^{226} Ra, ^{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, the contribution from the ^{238}U and the other ^{226}Ra, precursors are normally ignored. The measurement of natural radioactivity due to gamma rays from the dose rate is needed to implement precautionary measures whenever the dose is found to be above the recommended limits [6]. The aim of the present paper details of natural radioactivity in soil (^{226}Ra, ^{232}Th, and ^{40}K) and evaluate potential health hazards, the representative level index, $I\gamma$, the radium equivalent activity (Ra_{eq}), and the annual effective dose equivalent (AEDE) in air for all soil samples were estimated to

assess the contribution of this radionuclide to public exposure.

II. EXPERIMENTAL PROCEDURE

A. *Sample Collection and Preparation*

10 surface soil samples have been collected randomly from the studied area as shown in Fig. 1. After collection, samples were crushed into fine powder by using mortar and pestle. Fine quality of the sample was obtained using scientific sieve of 150 micron-mesh size. Before measurement, samples are dried. One kilogram of each sample was packed and sealed in a marinelli beaker and kept for about 4 week's period to allow radioactive equilibrium among the daughter products of radon (^{222}Ra), thoron (^{220}Ra) and their short lived decay products.

B. *Measurement of Natural Radioactivity*

The activity of samples is counted using NaI(Tl) detector 3"x 3" coupled to PC-MCA (4096 channel) model (Canberra, USA), based on high efficiency gamma spectrometry system. Spectral data from the detector was analyzed by using computer software (GINE-2000). The energy (FWHM) in the peak 1.33 keV for ^{60}Co was 7%. The detector was surrounded by a lead shielding to reduce the background radiation.Energy calibration and efficiency calibration of gamma spectrometer were carriedout using standard source (radionuclides mixed). One kilogram of each soil sample marinelli beaker and counted 1080 second.

Fig. 1. Locations of the samples in region under study

The background spectra distribution due to naturally occurring radionuclides in the environment around the detector, an empty marinelli beaker was counted in the same manner as the samples.

Gamma transitions of 1461 keV for ^{40}K, 186 keV and 609keV of ^{214}Bi for ^{226}Ra, 338, 463, 911, 968 keV for ^{228}Ac, 727 keV for ^{212}Bi, 238 keV for ^{212}Pb were used for the laboratory measurement of activity concentration potassium, radium and thorium.

The activity of the radionuclide is calculated from the background subtracted area prominent gamma ray energies. The specific activity (A) of radionuclides are calculated using the following equation [7]:

$$A\left(\frac{Bq}{kg}\right) = \frac{\text{net photopeak area}}{\mathrm{T} \times \mathrm{Eff\,(E)} \times \mathrm{P\gamma\,(E)} \times \mathrm{M}} \quad \ldots . \quad (1)$$

Where, T is the counting time, Eff (E) is the full-energy peak efficiency at photon energy Eγ, Pγ(E) is the emissionprobability of gamma-ray photons of energy Eγ, and M isthe mass of the sample (kg).

The minimum detectable activity (MDA) was calculated for each radionuclide according to equation [8], [9].

$$\mathrm{MDA} = \frac{\mathrm{LD}}{\mathrm{T} \times \mathrm{Eff\,(E)} \times \mathrm{P\gamma\,(E)} \times \mathrm{M}} \quad \ldots . \quad (2)$$

Where, LD is the detection limit calculated using thefollowing equation [9]:

$$LD = LC + K\,\sigma D \quad \ldots . \quad (3)$$

Where, LC is the critical level below which no signal can be detected, σDis the standard deviation, and K is the error probability.

C. *Radium Equivalent Activity Ra_{eq}*

Uniformity with respect to exposure to radiation has been defined in terms of radium equivalent activity (Ra_{eq}) in Bq/kg to compare the specific activity of materials containing different amounts of ^{238}U, ^{232}Th and ^{40}K. It is calculated through the following relation [10]:

$$Ra_{eq} = A_U + 1.43A_{Th} + 0.077A_K \quad \ldots . \quad (4)$$

Where: A_U, A_{Th} and A_K are the specific activity of ^{238}U,^{232}Th and ^{40}K in (Bq/kg) respectively. In the definition of Ra_{eq}, it is assumed that 10 Bq/kg of ^{226}Ra, 7 Bq/kg of ^{232}Th and 130 Bq/kg of ^{40}K produce equal gamma-ray dose rate[11].

D. Air-Absorbed Dose Rate

Absorbed gamma dose rate (D_γ) in (nGy/h) due to terrestrial gamma rays at 1 meter above the ground surface was calculated by using the conversion factor (by $nGy.h^{-1}/Bq.kg^{-1}$) of 0.0414 for ^{40}K, 0.461 for ^{226}Ra, and 0.623 for ^{232}Th [10], [12]:

$$D_\gamma = 0.461A_U + 0.623A_{Th} + 0.0414A_K \quad . \ . \ . \ . \quad (5)$$

E. Annual Effective Dose Equivalent

The estimated annual effective dose equivalent received by a member was determined using a conversion factor of (0.7Sv/Gy), which was used to convert the absorbed rate to human effective dose equivalent with an outdoor occupancy of 20% and 80% for indoors using thefollowing relations [10], [12]:

$$(AEDE)_{in} = D_\gamma \times 10^{-6} \times 8760 \times 0.8 \times 0.7 \quad . \ . \ (6)$$

$$(AEDE)_{out} = D_\gamma \times 10^{-4} \times 8760 \times 0.2 \times 0.7 \quad . \ . (7)$$

F. External Hazard Index (H_{ex}) and Internal Hazard Index (H_{in})

The external hazard index (H_{ex}) can be calculated by the following equation [10], [12]:

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad . \ . \ . \ . \quad (8)$$

The internal hazard index (H_{in}) can be calculated by the following equation [9] , [12]:

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad . \ . \ . \ . \quad (9)$$

G. Activity Concentration Index (Iɣ)

The activity concentration index (I_γ)forsoil samples was determined using the following relation [13]:

$$I_\gamma = \frac{C_{Ra}}{300} + \frac{C_{Th}}{200} + \frac{C_K}{3000} \quad . \ . \ . \ . \quad (10)$$

III. RESULTS AND DISCUSSIONS

In the ^{238}U decay series, both ^{214}Pb and ^{214}Bi are considered as ^{226}Ra indicators corresponding with radium mobility in geological times because of geochemical reasons [14]. The specific activities of natural radionuclides of ^{214}Pb, ^{214}Bi,^{226}Ra,^{228}Ac, ^{40}K and artificial radionuclide, ^{137}Cs in the surface soil samples were determined using equation 1 as shown in Table I. The specific activities of ^{214}Pb ranged from MDA to 66.87 Bq/kg with an average value of 19.86 Bq/kg, ^{214}Bi ranged from MDA to 170.25with an average value 38.61 Bq/kg, ^{226}Ra ranged from MDA to 246.96 Bq/kg with an average value 80.44Bq/kg, ^{228}Ac from MDA to 60.52 Bq/kg with an average value 24.90 Bq/kg, ^{40}K ranged from 4.4 to 226.36 Bq/kg with an average value 116.98 Bq/kg and ^{137}Cs ranged from MDA to 41.53 Bq/kg with an average value 10.17Bq/kg are calculated respectively.

Table I

Specific Activities (Bq/kg) of ^{226}Ra, ^{214}Pb and ^{214}Bi in Soil Samples from samara city of Salahdin Governorate-Iraq

Locations	Specific Activities (Bq/kg)					
	^{214}Pb	^{214}Bi	^{226}Ra	^{228}Ac	^{137}Cs	^{40}K
S_1	MDA	25.23	MDA	44.12	14.46	4.40
S_2	14.06	170.25	66.67	7.32	3.11	81.82
S_3	MDA	MDA	146.10	25.84	4.13	178.06
S_4	1.16	8.60	246.96	20.10	3.16	139.23
S_5	66.87	MDA	MDA	7.79	2.52	56.41
S_6	5.32	24.70	87.94	MDA	6.18	194.31
S_7	MDA	MDA	37.49	27.81	16.07	66.43
S_8	54.49	72.46	MDA	54.95	41.53	226.36
S_9	M DA	18.45	MDA	60.52	10.57	37.36
S_{10}	56.66	66.39	219.22	0.57	MDA	185.43
Average	19.86	38.61	80.44	24.90	10.17	116.98

In this study, the highest activities of ^{226}Ra, ^{228}Ac and ^{40}K was found in S_4,S_9 and S_8 respectively while the lowest activities was found in S_1S_5 S_8S_9, S_6 and S_1respectively as shown in fig. 2.

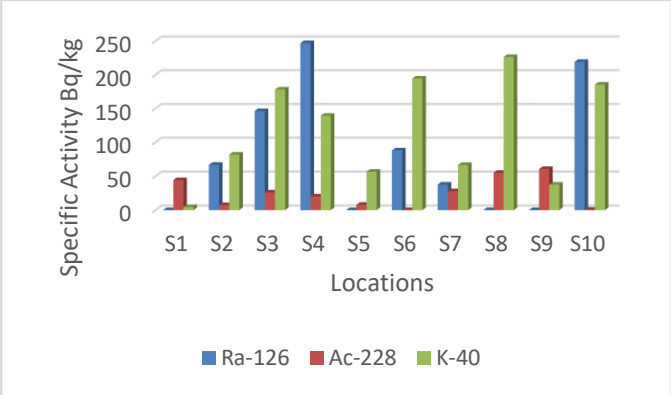

Fig.2: The specific activities of ^{226}Ra, ^{228}Ac and ^{40}K in surface soil samples

The mean values of the specific activities of ^{226}Ra 80.44 Bq/kg was higher than the world mean values 35 Bq/kg, ^{232}Th and ^{40}K in surface soil samples of this study are higher (24.90, 116.98)Bq/kg respectively,lower than the corresponding world mean values which are (30 and 400) Bq/kg (UNSCEAR, 2000), respectively. The specific activity of artificial radionuclide ^{137}Cs ranged from MDA to 41.53 Bq/kg with average value 10.17 Bq/kg. The maximum was higher than the world average value of 14.8 Bq/kg and the mean value lower than the world average [12], [14]. Radium equivalent (Ra_{eq}) activity have been calculated using equation 4, the results are summarized in Table II. The calculated Ra_{eq} values range from 63.42 to 286.39 Bq/kg with average of 131.78 Bq/kg for all soil samples. The mean Ra_{eq}value is lower than the recommended maximum value of 370 Bq/kg. Thus, these samples are within the recommended safety limit. In this study, the highest Ra_{eq} activity was found in S4, while the lowest Ra_{eq}activity was found in S1. Absorbed gamma dose rate (D_γ) in (nGy/h)and the annual effective dose rate were calculated using equations 5, 6 and 7, the estimated results are given in Table II. The absorbed gamma dose rate raged from 18.85 to 44.12 nGy/h with an average value 35.58 nGy/h as shown in fig. 3. The maximum and the mean values of absorbed dose rate is lower than the world average value of 55 nGy/h [9], [10]. The annual effective dose indoor ranged from 0.13 to 0.64 mSv/y with an average 0.29 mSv/y and outdoor ranged from 0.04 to 0.54mSv/y with an average 0.24 mSv/y shown in fig. 4. The maximum and the mean values are lower than the world average value of 1 mSv/y [12], [15].

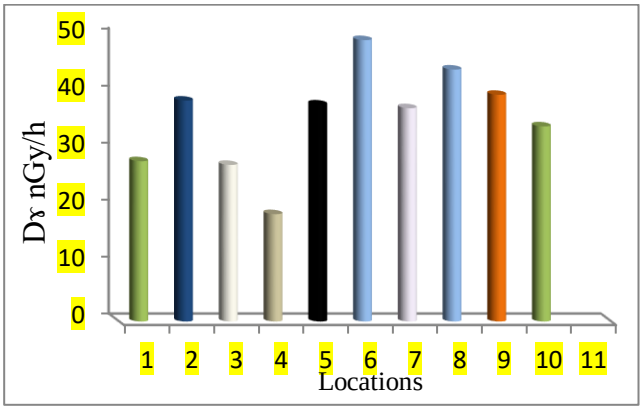

Fig.3. Absorbed Gamma Dose Rate (D_γ) in the samples for various Locations in the study area.

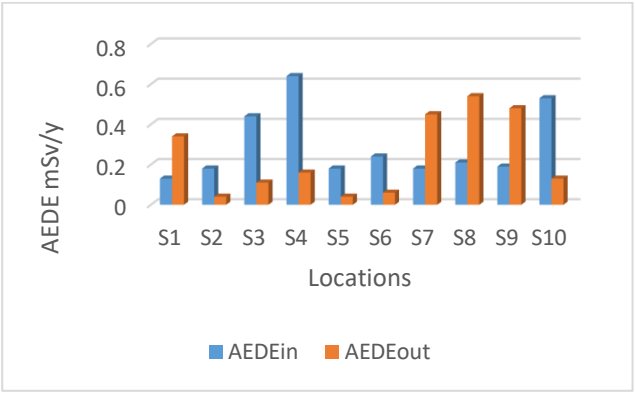

Fig.4. Annual Effective Dose Equivalent in the samples for various Locations in the study area.

Table II

Radiological hazards (Ra_{eq}, H_{ex}, H_{in}, I_γ), air-absorbed dose rates and annual effective doses

Location	Ra_{eq} (Bg/kg)	D_γ (nGy/h)	AEDE(mSv/y)		Hazard Index		I_γ
			Indoor	Outdoor	H_{ex}	H_{in}	
S_1	63.42	28.12	0.34	0.13	0.30	0.23	0.30
S_2	82.76	38.70	0.04	0.18	0.40	0.22	0.28
S_3	196.73	27.48	0.11	0.44	0.92	0.53	0.67
S_4	286.39	18.85	0.16	0.64	0.09	0.77	0.97
S_5	82.34	38.02	0.04	0.18	0.40	0.22	0.24
S_6	104.32	49.26	0.06	0.24	0.51	0.27	0.31
S_7	82.36	37.37	0.45	0.18	0.32	0.22	0.27
S_8	95.99	44.12	0.54	0.21	0.76	0.40	0.58
S_9	89.14	39.71	0.48	0.19	0.34	0.29	0.37
S_{10}	234.31	34.20	0.13	0.53	0.34	0.19	0.79
Average	131.78	35.58	0.24	0.29	0.44	0.33	0.48

The external and internal hazard index and the activity concentration index (I_γ) for soil samples were calculated using equations 8, 9 and 10, the estimated results are given in Table 2. The maximum, minimum and average values were lower than the world's average [15, 16].

IV. CONCLUSIONS

The results obtained showed that the distribution of natural radionuclides in the soil samples was not uniform and artificial radionuclide ^{137}Cs was detected in surface soil sample measured. The results of the mean specific activity of ^{238}U are higher than the world average and the mean specific activities of ^{232}Th and ^{40}K in the soil samples are lower than the world average. The mean value of radium equivalent was lower than the recommended maximum value. Absorbed gamma dose rate, the annual effective dose rate the external, internal hazard index and the activity concentration index were lower than the world's average.

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