



Measurement and Analysis of Natural Radioactive Elements in Salty Sediments from Central and Southern Iraq

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Abstract

Fifteen sediment samples from different areas in the middle and south of Iraq were prepared and their structural characteristics were studied by powder X-ray diffraction analysis at $\lambda=1.542\text{\AA}$. The present work considered the specific activities of naturally occurring radioactive materials in salt samples, which were measured and analyzed using high resolution HPGe system. Also, the radiological parameters were considered in this work. The average specific concentration of radionuclides (in Bq.kg^{-1}) in the salt samples was found to be 16.86 ± 4.92 for ^{226}Ra , 5.97 ± 2.05 for ^{232}Th , and 9.85 ± 2.8 for ^{40}K , which are below the UNSCEAR globally permitted levels of 40, 30, and 400Bq.kg^{-1} , respectively. Similarly, the absorption dose rates were in the range of 3.71 to 12.591 nGy.h^{-1} . The measured radiation hazard indices showed that the concentrations of these radionuclides in all samples are within the allowable limits.

Keywords: X-ray diffraction, Intensity, HPGe-system, Specific concentration of Radionuclides, Effective dose, crystal structure analysis, domestic Salts.

قياسات وتحليل العناصر المشعة الطبيعية في الرواسب المالحة لوسط وجنوب العراق

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الخلاصة:

تم تحضير خمسة عشر عينة من الأملاح الطبيعية ومن رواسب مختلفة في وسط وجنوب العراق. درست الخصائص الهيكلية للعينات عن طريق تحليل حيود المسحوق بالأشعة السينية عند طول موجي (1.542) انكوستروم. أخذ العمل الحالي في الاعتبار النشاط المحدد للمواد المشعة التي تحدث بشكل طبيعي في عينات الملح والتي يتم قياسها وتحليلها باستخدام منظومة كاشف إشعاع جرمانيوم عالية النقاء. أيضا، تم النظر في المعلمات الإشعاعية لهذا العمل وتحديد متوسط التركيز النوعي (بوحدة البكرل/كغم) لعينات الملح وكانت، نظير الراديوم-226 (16.86±4.92)، نظير الثوريوم-232 (5.97±2.05) ونظير البوتاسيوم-40 (9.85±2.8) على التوالي، وهي أقل من المستويات الدولية (40، 30 و 400 بكرل/كغم) على التوالي. وبالمثل فإن معدلات جرعة الامتصاص تقع في المدى من 3.71 نانوكري/ساعة إلى 12.59

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نانوكري/ساعة . توضح مؤشرات مخاطر الإشعاع المقاسة أن تراكيز هذه النويدات المشعة في جميع العينات تقع ضمن الحدود المسموح بها .

1. Introduction

The spread of naturally occurring radionuclides and environmental radioactivity have become an important issue in many countries [1-7]. The measurement of these naturally occurring radionuclides in natural salt materials in a specific environment allows determining the exposure and the absorbed doses in the population or subjects working in close contact to these materials . The industrial activities that involve natural salts, such as extraction and remediation from soil components, may cause the incorporation of natural radionuclides with salt molecules, which varies from region to region. The daily consumption of natural salts that contain high amounts of natural radionuclides may lead to an increase in radiation toxicity inside the human body. The presence of radionuclides in natural salts leads to their decay and the emission of alpha particles with energy (in ^{238}U and ^{232}Th series) and beta particles with energy of 1.46MeV (in ^{40}K series). Usually, the momentum and activity of these particles vary from soil to another and from region to region depending on the abundance of natural radionuclides. However, necessary monitoring is required for the level of radiation in any area relative to the radiation background. Most of the surveys conducted in different areas of the world are used to aid in estimating the reference dose for public and workers and evaluate the environmental radioactivity according to the nuclear activities [8]. The natural radioactive materials have originated from the terrestrial left on the earth [9-11]. The main primordial radionuclides arise from ^{238}U and ^{232}Th series beside the decay of ^{40}K radionuclide. The amount of radioactivity is widely varied in the soil, vegetables, and other materials used in foods consumed by humans, including local salt [12-14]. The specific concentration measurements of radionuclides in salts have become an interesting subject to indicate radioactivity accumulation in the environment and its effects on different organisms [15-18].

2. Materials and methods

Fifteen natural salt samples were collected from different Iraqi areas in Falluja town, Samawah salted plant in Muthanna governorate, Diwaniyah salt pond town in Qadisiyah governorate, Babil governorate, and Dhi Qar governorate. The areas of natural salt presence were selected according to their popularity with production and its spread in the local markets; see Figures 1A and 1B. These samples were purified through re-crystallization after removing the imperfections and dust. Then, the samples were dried, grinded, and sieved individually. Samples were analyzed using the XRD system at the College of Science, University of Baghdad. The structural characteristics were determined by using the powder X-ray diffraction analysis at $\lambda=1.542\text{\AA}$. Typical spectra for samples S1 and S4 are shown in Figures 2a and 2b.

The concentrations of ^{226}Ra , ^{232}Th , and ^{40}K natural radionuclides found in NaCl samples were measured using high-purity Germanium detector (HPGe) gamma spectroscopy, with the standard setup, at the radiation protection center (RPC)-Ministry of Health and Environment, Baghdad, Iraq. The samples were crushed, milled, and grinded. All samples were placed in an oven for drying at $t 110^{\circ}\text{C}$ for one hour until complete removal of any residual moisture and ensuring that a constant weight was achieved [19, 20]. The dried samples were pulverized into a fine powder and passed through a standard 1 mm mesh size sieve. A Marinelli beaker (1 L) was used to measure the radioactivity concentration by HPGe system, with 40 % efficiency. As shown in Figure 3, the distribution of natural radionuclides in local natural salt samples is not uniform.

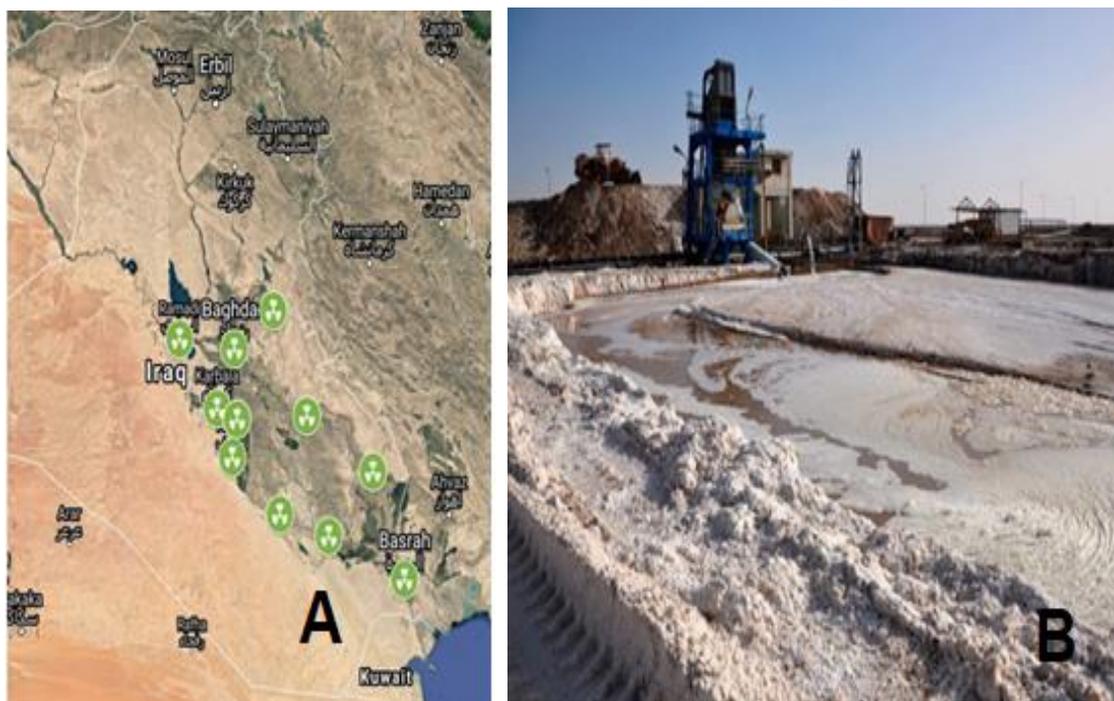


Figure 1-(A) The sites of sampling areas in the middle and south of Iraq, (B) the Samawah salt plant.

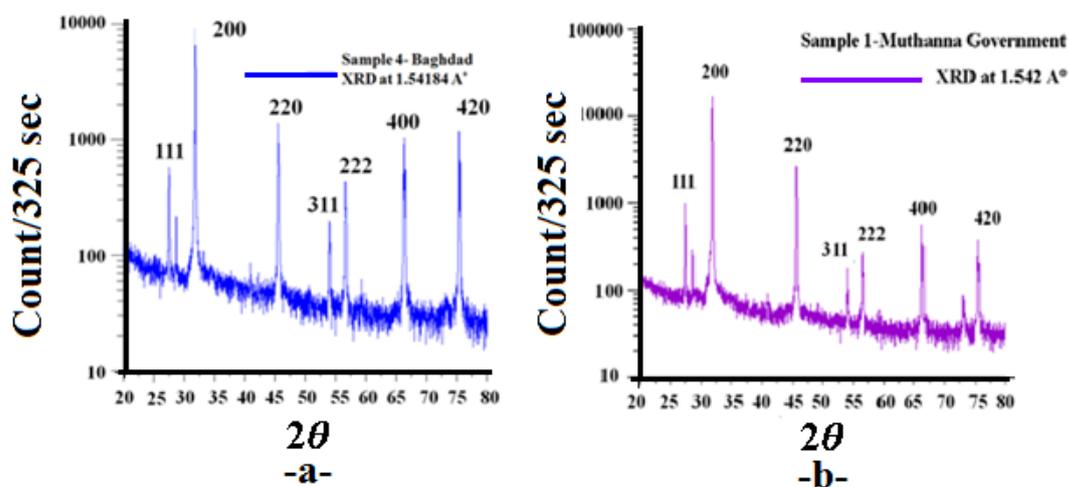


Figure 2-The intensity distribution of the XRD as a function of two theta for **a-** S4-Baghdad-domestic salt (Reference sample), **b-** S1-Muthanna-domestic salt. Purification of natural salt samples was achieved in the Department of Chemical Industry, Institute of Technology, Baghdad Middle Technical University).

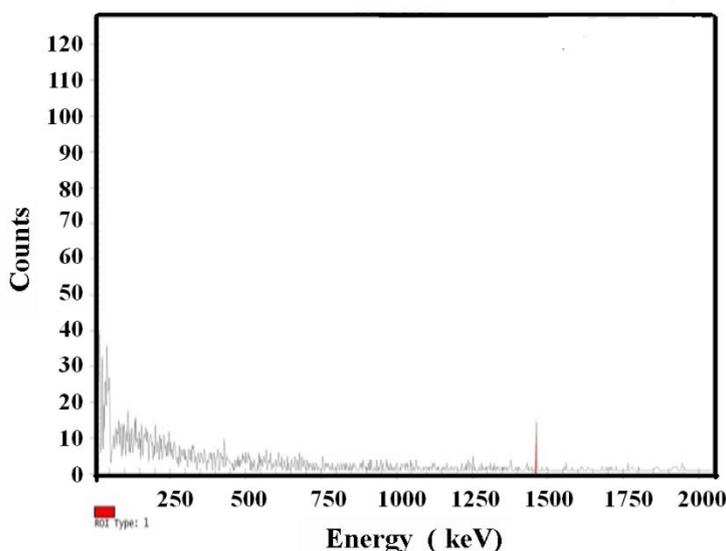


Figure 3-Gamma ray spectrum for sample 1 of natural salts, with a Peak of 1480 keV for ⁴⁰K

3. Results and discussion

Different gamma ray parameters were considered in the present investigation to evaluate the concentrations of radioactive elements in natural salt samples.

3.1 Radium equivalent activity

The specific activity level for ²²⁶Ra, ²³²Th, and ⁴⁰K radionuclides was measured in terms of radiological index (Ra_{eq}), which is called radium equivalent activity. This parameter is used to assess the radiation hazards associated with salt samples that contain ²²⁶Ra, ²³²Th, and ⁴⁰K radionuclides, measured in $Bq.kg^{-1}$ [21]. It can be calculated by assuming that 370 $Bq.kg^{-1}$ of ²²⁶Ra, 260 $Bq.kg^{-1}$ of ²³²Th, and 4810 $Bq.kg^{-1}$ of ⁴⁰K produce the same γ -dose rate [22]. The following relation is used to calculate Ra_{eq} [23]:

$$Ra_{eq} = A_{Ra} + 1.43 A_{Th} + 0.077 A_K \tag{1}$$

3.2 External and internal radiation hazard indices

The external and internal radiation hazard indices (H_{ext} and H_{int}) were obtained to evaluate the γ -ray level associated with natural radioactivity in the investigated salt samples using the following equations [22, 23]:

$$H_{ext} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \tag{2}$$

$$H_{int} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1$$

3.3 The absorbed dose rate in air

The absorbed dose rate (D_γ) in air at one meter above the ground surface, where the natural salt samples are collected, is represented by the relation between concentrations of natural radionuclides and their exposure values. The relation can be described as [21, 23]:

$$DG = 0.462 A_{Ra} + 0.604 A_{Th} + 0.0417 A_K \tag{3}$$

1- Outdoor external exposure

$$DG_{out}(nGyh^{-1}) = 0.436A_{Ra} + 0.599A_{Th} + 0.0417 A_K \tag{4}$$

2- Indoor external exposure to gamma

$$DG_{in}(nGyh^{-1}) = 0.92A_{Ra} + 1.1A_{Th} + 0.081 A_K \tag{5}$$

where the factors 0.462, 0.604, and 0.0417 are the conversion factors for naturally occurring radionuclides ²²⁶Ra, ²³²Th, and ⁴⁰K, respectively [21].

3.4 The annual effective dose equivalent

Another radiological parameter considered in the present study is the annual effective dose equivalent (AEDE) at the outdoor (in air). It is calculated by using the absorbed dose and taking into account two factors; the conversion factor of 0.7 Sv.Gy^{-1} and the occupancy factor for outdoor, which is 0.2. The AEDE-out calculation in $\mu\text{Sv.yr}^{-1}$ follows the following formula [21,22]:

$$\text{AEDE}_{\text{OUT}}(\mu\text{Sv.yr}^{-1}) = D(\text{nGy.h}^{-1}) \times 8760\text{h} \times 0.7(\text{Sv.Gy}^{-1}) \times 0.2 \times 10^{-3} \quad (6)$$

3.5 The gamma-ray hazard

To estimate the gamma-ray hazard (GI) associated with the natural radionuclides in different salt samples, is the following formula was applied:

$$\text{GI} = \frac{A_{\text{Ra}}}{150} + \frac{A_{\text{Th}}}{100} + \frac{A_{\text{K}}}{1500} \quad (7)$$

This parameter correlates the annual dose rates occurring due to excess external gamma radiation that is caused by superficial materials. A value of $\text{GI} \leq 1$ corresponds to an annual effective dose of less than 1mSv, while $\text{GI} \leq 0.5$ corresponds to an annual effective dose of less than or equal to 0.3mSv.

Table 1 and Figure 4 show the activity concentrations for natural radioactive ^{226}Ra , ^{232}Th , and ^{40}K , in different salt samples collected from different area in Iraq.

3.6 The Excess Life Cancer Risk

Most researchers in this field agreed on the possibility of cancer or developing a genetic defect in the human progeny in the future due to taking low-effective ionized radiation doses over a long period of time [23-27]. Therefore, to evaluate this possibility, the 3.6 The Excess Life Cancer Risk (ELCR) value is calculated using:

$$\text{ELCR}_{\text{out}} = E_{\text{out}} \times D_L \times f_L \quad (8)$$

$$\text{ELCR}_{\text{in}} = E_{\text{in}} \times D_L \times f_L$$

$$\text{ELCR}_{\text{tot}} = \text{ELCR}_{\text{out}} + \text{ELCR}_{\text{in}}$$

where D_L is the life period of 66 years and R_f is the fatal risk factor per 0.05 Sv^{-1} for the public [4, 5].

The Annual Gonadal Equivalent Dose The Annual Gonadal Equivalent Dose (AGED) is measure of the threat and stomach from exposed to a particular level of radiation. It is calculated by using the following formula:

$$\text{AGED}(\mu\text{Sv.y}^{-1}) = 3.09A_{\text{Ra}} + 4.18A_{\text{Th}} + 0.314A_{\text{K}} \quad (9)$$

The results of the different radiation hazards indices mentioned above, which were calculated from the measurements of radiation levels in salt samples, are shown in Table 1 and Figures 4, 5, and 6.

Table 1-The values of radiation hazards indices in natural salts collected from different areas in the middle and south of Iraq.

Sample No.	^{226}Ra Bq/kg	^{232}Th Bq/kg	^{40}K Bq/kg	Raeq Bq/kg	DG _{TOT} nGy/h	GI nGy/h	AEDE _{TOT} μSv/y	AGED μSv/y	ELCR _T 10 ⁻³	Hex	Hin
S1	6.21 ± 2.0	4.11 ± 1.5	23 ± 7.4	13.86 ±0.34	18.41 ±2.88	0.087 ±0.01 4	22.575 ±2.31 2	43.591 ±6.805	0.074 ±0.012	0.062 ±0.01	0.054 ±0.008
S2	4.8 ± 1.4	10.6 ± 3.2	19.6 ± 4.0	21.47 ±1.88	27.10 ±4.33	0.142 ±0.02 3	33.237 ±3.46 4	65.294 ±10.42 6	0.11 ±0.018	0.077 ±0.01 3	0.071 ±0.01 1
S3	7.4 ± 2.7	BDL *	7 ± 3.2	7.94 ±0.72	11.09 ±1.81	0.051 ±0.00 8	13.598 ±1.47 3	25.068 ± 4.085	0.045 ±0.007	0.051 ±0.00 8	0.041 ±0.00 7
S4	5.5 ±	15.1 ±	22.3 ±	28.81 ±2.51	36.07 ±5.37	0.192 ±0.02 9	44.233 ±4.29 1	87.115 ± 12.95	0.146 ±0.022	0.100 ±0.01 5	0.093 ±0.01 4

	2.1	4.1	6.4								
S5	BDL * ±	9.6 ± 2.4	12.4 ± 3.6	14.68 ±1.29	17.88 ±2.75	0.099 ±0.01 5	21.93 ±2.18 3	44.025 ± 6.772	0.072 ±0.011	0.040 ±0.00 6	0.040 ±0.00 6
S6	6.1 ± 2.8	9.1 ± 2.6	14.1 ± 3.4	20.20 ±1.97	25.67 ±3.94	0.135 ±0.02 1	31.478 ±3.15 8	61.314 ±9.416	0.104 ±0.016	0.079 ±0.01 2	0.071 ±0.01 1
S7	7.7 ± 3.0	11.3 ± 3.6	8.0 ± 3.6	24.48 ±3.33	30.88 ±5.07	0.166 ±0.02 7	37.869 ±4.06 4	73.539 ±12.07 5	0.125 ±0.021	0.097 ±0.01 6	0.087 ±0.01 4
S8	5.9 ± 2.4	10.4 5± 3.7	12.7 ± 4.7	21.82 ±2.51	27.51 9 ±4.42	0.146 ±0.02 4	33.749 ±3.53 1	65.9 ± 10.624	0.111 ±0.018	0.082 ±0.01 3	0.075 ±0.01 2
S9	6.1 ± 2.7	12.6 6± 3.8	30.1 ± 7.9	26.52 ±1.54	33.7 ±5.0	0.174 ±0.02 6	41.326 ±4.01 6	81.222 ±12.10 5	0.136 ±0.02	0.096 ±0.01 4	0.088 ±0.01 3
S10	8.1 ± 3.1	BDL * ±	22.9 ± 6.1	9.86 ±0.38	14.01 ±2.04	0.059 ±0.00 9	17.177 ±1.66 4	32.224 ± 4.688	0.057 ±0.008	0.059 ±0.00 9	0.049 ±0.00 7
S11	5.6 ± 2.1	9.55 ± 3.2	10.3 2 ± 4.1	20.05 ±2.26	25.28 ±3.92	0.1/35 ±0.02 1	31.002 ±3.13 7	60.463 ± 9.4	0.102 ±0.016	0.076 ±0.01 2	0.069 ±0.01 1
S12	6.23 ± 2.3	9 ±3	17.7 6 ± 5.1	20.45 ±1.69	26.10 8 ±3.98	0.135 ±0.02 1	32.019 ±3.18 4	62.406 ±9.512	0.106 ±0.016	0.08 ±0.01 2	0.072 ±0.01 1
S13	7.1 ± 2.2	11.7 ± 3.8	13.3 ± 3.9	24.87 ±2.81	31.40 ±4.91	0.167 ±0.02 6	38.511 ±3.93 9	75.072 ±11.68 8	0.127 ±0.02	0.095 ±0.01 6	0.086 ±0.01 4
S14	BDL * ±	12.0 ± 3.7	21.2 ± 4.2	18.79 ±0.68	23.05 ±2.92	0.124 ±0.01 6	28.272 ±2.32 2	56.826 ±7.212	0.093 ±0.012	0.051 ±0.00 6	0.051 ±0.00 6
S15	6.89 ± 2.5	9.56 ± 2.9	14.6 ± 5. 9	21.69 ±2.53	27.61 ±4.71	0.145 ±0.02 5	33.855 ±3.78 2	65.838 ±11.16 1	0.112 ±0.019	0.086 ±0.01 5	0.077 ±0.01 4
S_{ave}	5.6 ± 2.1	9.0 ± 2.8	16.6 ± 5.2	19.7 ± 1.7	25.05 ± 3.87	0.13 ± 0.02	30.722 ± 4.74 9	59.993 ± 9.261	0.101 ± 0.160	0.075 ± 0.01 2	0.068 ± 0.01 1

*BDL: Below Detectable Limit.

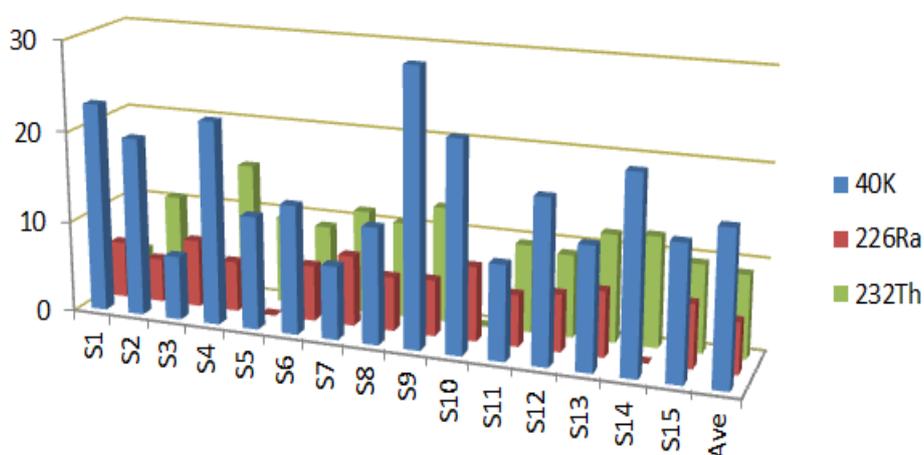


Figure 4 -The Activity concentrations for radioactive ²²⁶Ra, ²³²Th, and ⁴⁰K in natural salt samples collected from different areas in Iraq. The values were shown to be below the national levels of 40, 30, and 400Bq.kg⁻¹, respectively [1].

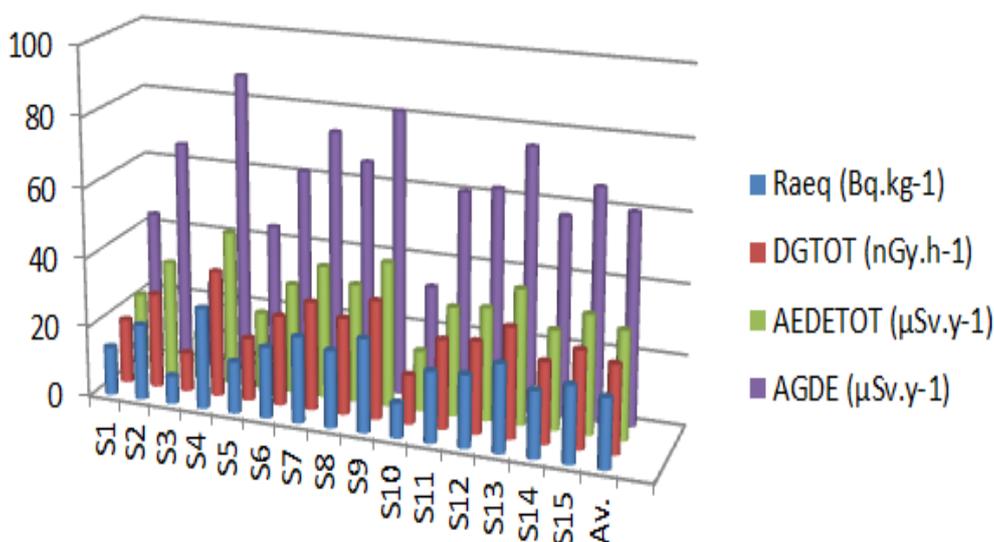


Figure 5-The values of radiation hazard indices calculated in natural salt samples collected from different areas in Iraq.

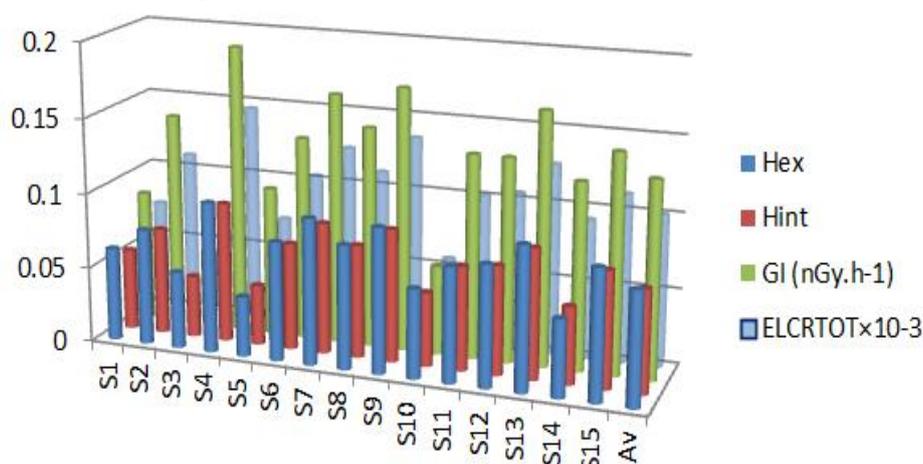


Figure 6 - The values of external and internal radiation hazard indices (H_{ext} and H_{int}) obtained to evaluate the γ -ray level associated with natural radioactivity, the gamma-ray hazard, GI, associated with the natural radionuclides

4. Statistical Analysis of the data

In previous research works, the univariate statistics were applied for the purpose of obtaining data with reasonable, scientific interpretation. However, this approach may be inaccurate [28]. Accordingly, for the purpose of achieving a correlation analysis between any pair of radiation indices, shown in Table 2, a linear Pearson correlation coefficient was implemented in present work, using a multivariate analysis [28]. This method treats the radio-environmental data with a successful justification in building the relationships among the variables of radiology indices.

As shown in Table 2, high, positive correlations are observed between ^{226}Ra (^{238}U -series), ^{232}Th -series, and ^{40}K -series, whereas a negative correlation was found between ^{226}Ra (^{238}U -series) and ^{232}Th -series. These results indicate that the radiation indices are existed due to the availability of radionuclides concentrations in the areas salt sampling.

Table 2-The Pearson correlation matrix for different radiation indices in natural salts samples collected from different areas in Iraq..

	226Ra	232Th	40K	Raeq	DG _{TOT}	GI	AEDEtot	AGDE	ELCRtot	Hex	Hin
226Ra	1										
232Th	-0.351	1									
40K	0.152	0.152	1								
Raeq	0.033	0.921	0.20	1							
DG _{TOT}	0.095	0.893	0.22	0.99	1						
GI	0.021	0.928	0.15	0.99	0.994	1					
AEDEtot	0.095	0.893	0.22	0.99	1.000	0.99	1				
AGDE	0.053	0.910	0.22	0.99	0.999	0.99	0.999	1			
ELCRtot	0.099	0.891	0.21	0.99	1.000	0.99	1.000	0.999	1		
Hex	0.537	0.599	0.11	0.86	0.891	0.85	0.891	0.870	0.893	1	
Hin	0.401	0.713	0.14	0.92	0.950	0.92	0.950	0.935	0.951	0.988	1

5. Conclusions

Different natural salt samples were collected and purified from different areas of Iraq. The structural characteristics of the prepared samples were analyzed using the X-ray diffraction. The power of X-ray diffraction was adjusted at $\lambda=1.542 \text{ \AA}$ to obtain a diffraction pattern, followed by a subsequent analysis of these patterns which produced the initial crystal of the salt (NaCl). The behavior of the measured intensities, diffracted from different lattice sites of NaCl, was similar to that calculated theoretically. Also, values of radiological parameters were obtained using the available high resolution HPGe spectrometer. The average specific activities of the natural radionuclides occurring in the salt samples were found to be 16.86 ± 4.92 for ^{226}Ra , 5.97 ± 2.05 for ^{232}Th , and 9.85 ± 2.8 for ^{40}K , all being below the nationally accepted levels of 40Bq.kg^{-1} , 30Bq.kg^{-1} , and 400Bq.kg^{-1} , respectively. The results of the radiation indices showed that the concentrations of these radionuclides in all samples are within the allowable limits, which are less than unity. Measurements of radiation hazard indices showed that the concentrations of these isotopes in all samples are within the allowable limits internationally. The absorbed dose rates were calculated, for five samples, from the specific concentrations of ^{226}Ra , ^{232}Th , and ^{40}K and showed a range of 3.71 to 12.591 nGy.h^{-1} .

6. References

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