Assessment natural radioactivity of marl as raw material at Kufa Cement Quarry in Najaf Governorate

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Abstract
This Research involves radiological study to assess the marl layer in the Euphrates Formation (Early Miocene) as a raw material for Portland cement industry. Nine wells are drilled penetrating the marl layer to the limestone beneath it. Nine samples were collected from core wells. Each sample represents one well. The concentration of the natural radionuclides ^{226}Ra, ^{232}Th and ^{40}K are 14.91, 5.16 and 223.98 Bq/kg^{-1}, respectively. They are determined by using the technique of gamma-ray spectroscopy with HPGe detector. The radiation doesn't exceed the globally permissible limits. Then the results were compared with the reported data of other countries and with the world average activity of cement raw materials. The radium equivalent (Ra_{eq}) activities values of marl samples are lower than the limit of 370 Bq kg^{-1}. Gamma index (Iɣ) below 1 mSv y^{-1}. The values of the external hazard index (H_{ex}) and the internal radiation hazard index (H_{in}) are less than unity. For estimating the radiological hazards on human health, these parameters are used. The results are indicated that there is no negative effect of radioactive radionuclides on workers' health.

تقييم الاشعاع الطبيعي للمارل كمادة اولية ضمن مقلع سمنت الكوفة في محافظة النجف

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الخلاصة:
يتضمن هذا البحث دراسة اشعاعية لتقييم طبقة المارل في تكوين الفرات (المايوسين المبكر) كمادة اولية لصناعة السمنت . حفرت تسع ابار لاختراق طبقة المارل للوصول الى طبقة الحجر الجيري التي تحتها . وجمعت تسعة نماذج من اللباب المستخرج، حيث ان كل نموذج يمثل بئر واحد.
تركيز النويدات المشعة للعناصر ^{226}Ra, ^{232}Th and ^{40}K كان 14.91, 5.16 and 223.98 Bq/kg^{-1} على التوالي. تم الفحص بواسطة جهاز اشعة كاما مع كاشف الجرمانوم (HPGe). وكانت نسب الاشعاع ضمن الحدود المسموح بها عالميا . ومن ثم تم مقارنة النتائج مع التقارير للبلدان الاخرى ومع المعدلات العالمية للمواد الاولية لصناعة السمنت . كان مكافئ الراديوم (Ra_{eq}) لنماذج المارل اقل من 370 Bq kg^{-1}. وعامل اشعة كاما (Iɣ) اقل من 1 mSv y^{-1} . كانت قمة عامل المخاطر الداخلية (H_{in}) وعامل المخاطر الخارجية (H_{ex}) اقل من وحدة واحدة . تم استعمال هذه العوامل لغرض تخمين المخاطر الاشعاعية على صحة الانسان . دلت النتائج المستحصلة بان ه لا توجد تأثيرات سلبية للنويدات المشعة على صحة العاملين.

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Introduction

Cement raw materials can cause significant gamma dose indoors, due to their natural radionuclide content depending on the naturally occurring radioactivity of its raw materials. Radionuclides are heavy isotopes in the environment that are not stable. To be stable, these nuclides emit radiations or particles [1]. Natural radioactive materials under certain conditions can reach hazardous radiological levels. It is necessary to study the natural radioactivity levels in cement raw materials to assess the dose for the population in order to know the health risks. Humans are exposed to ambient ionizing radiation from ^{40}K, ^{238}U series and ^{232}Th series and their decay products which widely spread in the earth's environment [2]. Workers exposed to cement or its raw materials for a long time especially in mines and at manufacturing sites as well as individual typically spend 80% of their time indoors [3]. Knowledge of the natural radioactivity levels in building materials is an important issue in the assessment of overall human exposure to natural radiation associated with ^{226}Ra and ^{232}Th (and their decay progeny) and the primordial radionuclide ^{40}K [4]. The aim of the study is assessed the radiological to discover the radiation level of the marl layer as raw materials for cement industry.

Location of the study area

The study area is a part of the limestone quarry of the Kufa Cement Plant. It is situated to the west of the Najaf city with a distance of about 26 km. The limestone layer at quarry belongs to the Euphrates Formation in the Khreba valley [5]. It is accurately determined by the latitude and longitude coordinates E 427122.7-N 3524181.7 and E 426305.1-N 3521444.6. Nine boreholes are drilled in the study area for collecting marl samples. The thickness and UTM location of boreholes show in Table1. The study area is located within the Salman Subzone which belongs to the Stable Shelf Zone [6]. The detailed location of the study area is illustrated in Figure 1.

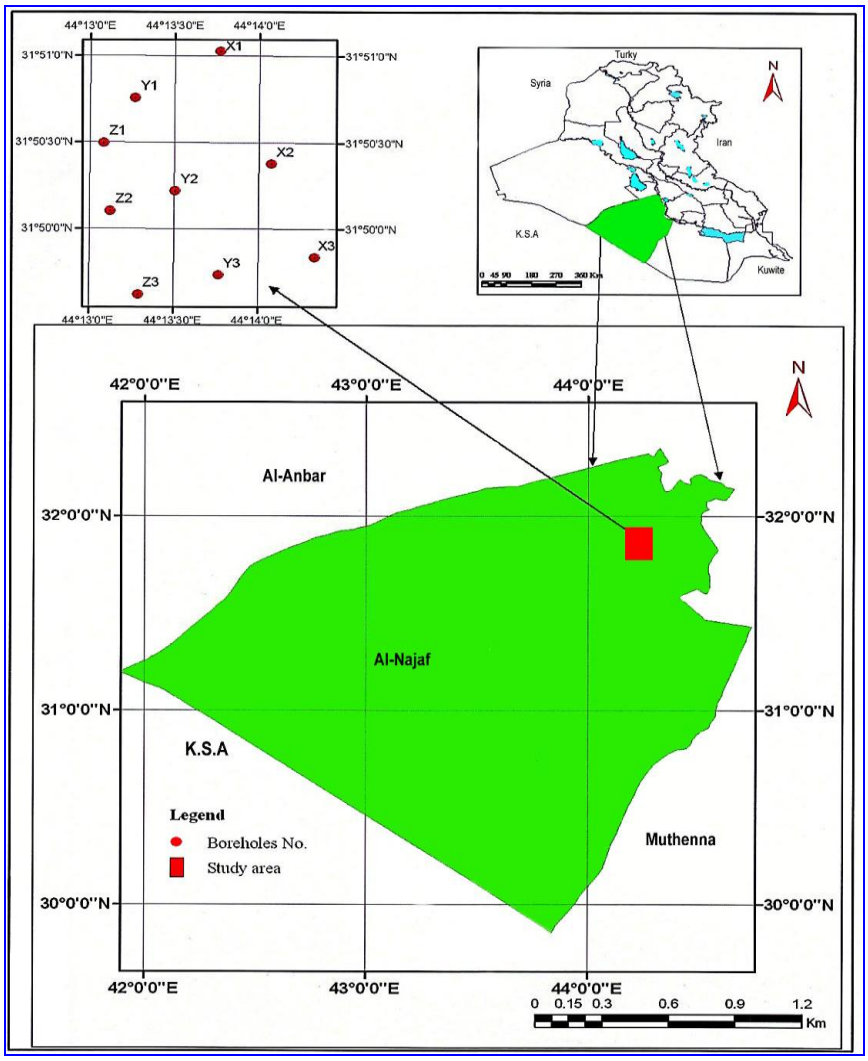

Figure 1- Location map of the study area.

Materials and method

Radioactive elements ^{226}Ra, ^{232}Th and ^{40}K concentrations are measured in the marl samples of the study area. Isorad map of the marl layer is drawn. Nine samples are used for radiation measurement.

For radioactivity test, the samples are crushed into homogenized powder of particle size 1 mm using a grinder machine. The powder of was dried at 105°C for a period of 24 h in the oven to remove moisture [7]. Then it was sieved through a 100 mesh [8]. One kg of each sample is put into standard Marinelli beaker and the activity of ^{226}Ra, ^{232}Th and ^{40}k is determined using the technique of gamma spectrometry. This technique depends on the high-purity germanium detector with an efficiency of 40% and resolution of 2 keV at 1332 keV of a standard source of radioactive isotope ^{60}Co. The measurements are done directly without any chemical treatment and the background and sample counting was carried out for a time of 3600 second. The measurements of gamma ray for samples were done in the laboratories of the Radiation Research Department of Radiation Protection Center in the Ministry of Environment.

Result and Discussion

Radioactivity Element Specification

The measurement of the natural radioactivity in the cement raw material is very important to determine the environmental hazards. Generally all of the building materials contain natural radionuclides representing ^{238}U decay series, ^{232}Th series and ^{40}K [9]. In the ^{238}U series, the decay chain segment starting from Radium ^{226}Ra which is radiologically the most important in addition to its decay products which produce 98.5% of the radiological effects of uranium series [10]. Therefore, reference is often made to ^{226}Ra instead of ^{238}U [11].

The specific activities are averaged from gamma-ray photo peak at several energies. The gamma-ray lines at 295.2 and 351.9 keV from ^{214}Pb and at 609.3 and 1764.5 keV from ^{214}Bi were used to determine the specific activity of ^{226}Ra. The gamma-ray lines of 338.4 and 911.2 keV from ^{228}Ac, the 727.3 keV from ^{212}Bi and 583.2 and 2614.5 keV from ^{208}Tl were used to determine the specific activity of ^{232}Th. The specific activity of ^{40}K was measured directly by its own gamma-ray line at 1460.8 keV.

Radioactivity of marl layer

Radiological analyses of had been carried out to determine ^{226}R, ^{232}Th and ^{40}K in the representative nine samples of the marl layer. Table 1 summarizes the measured concentrations of the naturally occurring radioactive elements ^{226}Ra, ^{232}Th and ^{40}K in samples of marl layer and Figures 2 and 3 displays the gamma-ray spectrum for boreholes X3 and Y3.

Al-Bassam [12] clarified the origin of U in the Euphrates Formation (Early Miocene) from source rocks, lying several kilometers underneath. The late Early Miocene tectonic unrest triggered fracturing and faulting that allowed for uranium−rich groundwater, trapped in the Paleozoic aquifers, to ascend to surface in the shallow parts of the late Early Miocene Sea, together with bitumen and H_2S seepages. Uranium was precipitated below sediment−water interface within the Euphrates Formation sediment. Because of the marl contains high percent of clay minerals, the radioactive nucleoids are strongly absorbed by them especially montmorillonite and palygorskite [20].

Table 1- Activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in marl layer.

Sample No.	Well No.	UTM-Coordination		Thickness of marl layer (m)	Activity concentration(Bq kg^{-1})		
		Easting	Northing		^{226}Ra	^{232}Th	^{40}K
1	X1	427122.7	3524181.7	9.00	15.4	4.5	235.4
2	X2	427637.6	3522915.6	8.70	12.4	6.2	222.05
3	X3	428062.3	3521863.2	8.50	13.4	5.1	185.8
4	Y1	426282.1	3523630.0	8.75	11.9	4.4	244.1
5	Y2	426683.1	3522616.9	8.50	16.9	5.7	255.4
6	Y3	427104.4	3521636.9	8.40	12.7	4.9	174.4
7	Z1	425947.6	3523163.2	8.75	10.6	5.2	242.2
8	Z2	426026.8	3522402.3	7.75	12.8	5.06	211.1
9	Z3	426305.1	3521444.6	7.80	28.1	5.4	245.4
minimum				7.80	10.6	4.4	174.4
maximum				9.00	28.1	6.2	255.4
Mean				8.46	14.91	5.16	223.98

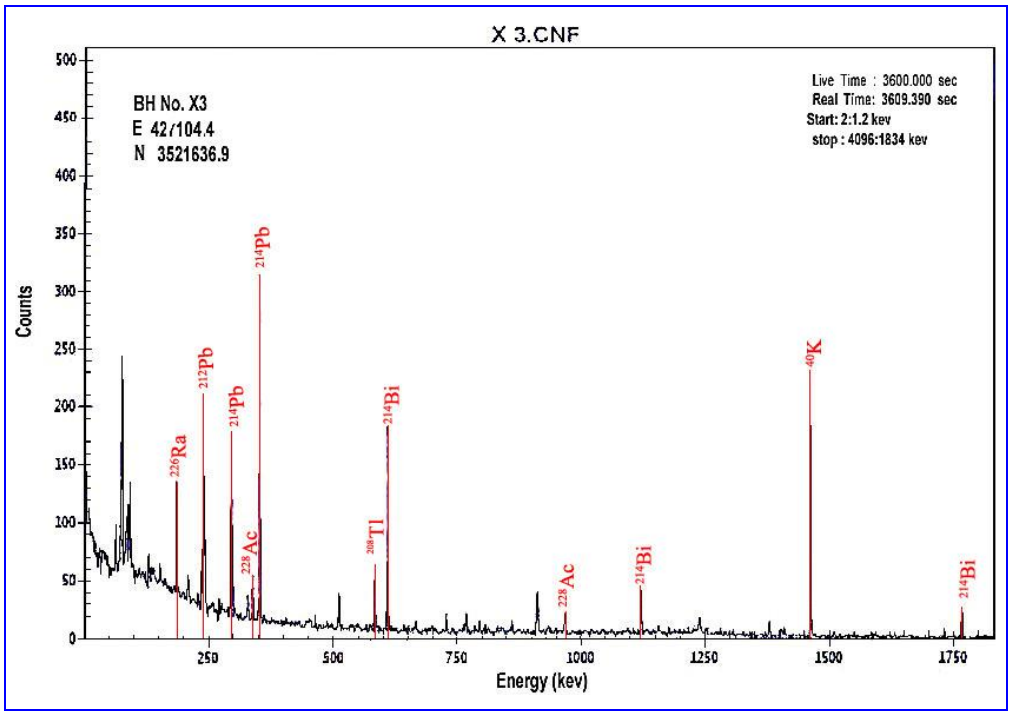

Figure 2- Gamma-ray spectrum of marl in BH X3.

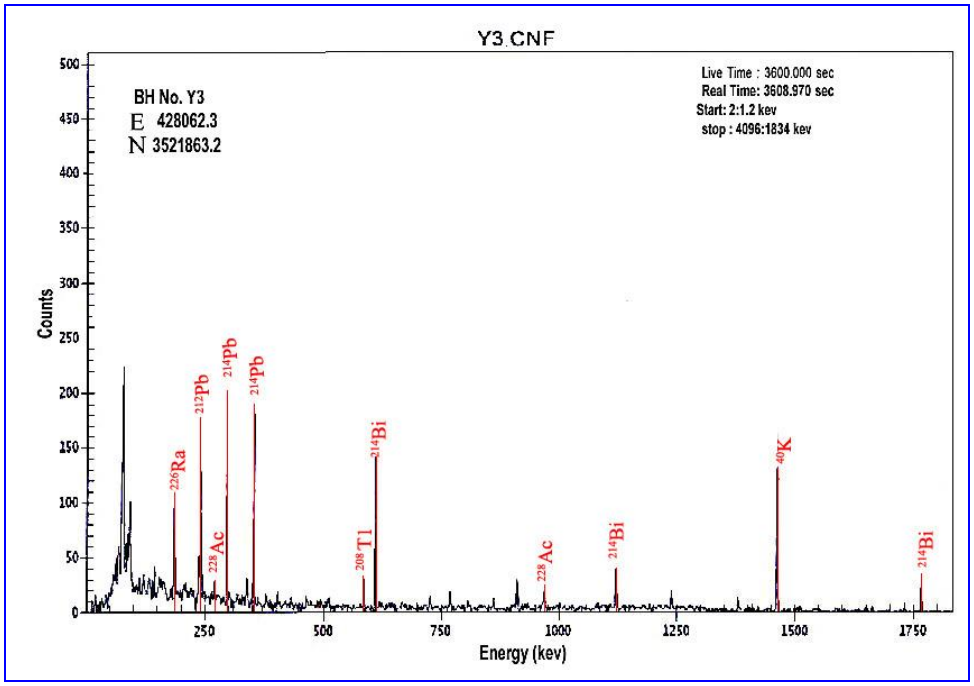

Figure 3- Gamma-ray spectrum of marl in BH Y3.

Radium-226

Radium is the heaviest in alkaline earth elements. It is well known that strontium and radium are chemically similar to Ca and radium exists in the environment typically as a divalent cation. Ra, which behaves similarly to Ca, may form aqueous complexes with sulfate ions or carbonate ions [13]. Zhang [14] suggests that Ra can form both inner- and outer-surface complexes on the clay surface. In addition Ra has higher affinity for Mn hydrated oxides [15]. The elements ^{214}Pb and ^{214}Bi are represented ^{226}Ra [16]. Activity concentration of ^{226}Ra of marl layer is ranging between 10.6 Bq kg^{-1} and 28.1 Bq kg^{-1} with mean 14.91 Bq kg^{-1}. The distribution of ^{226} Ra in the study area is shown in Figure 4.

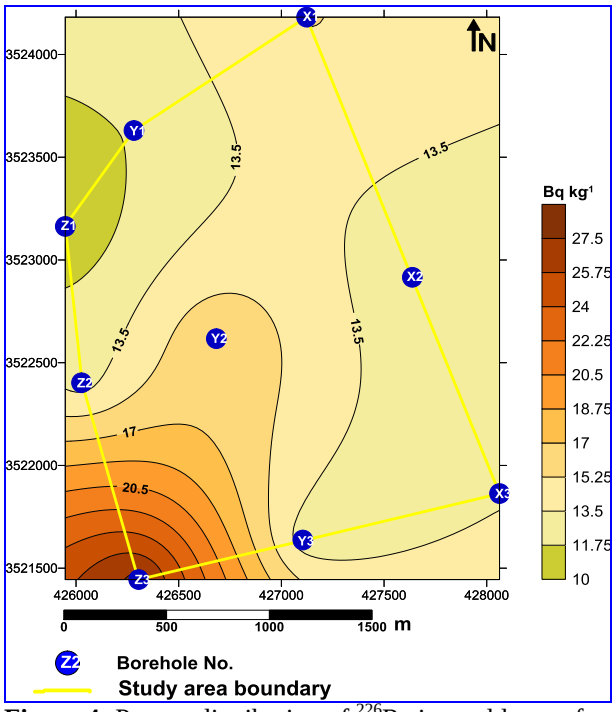

Figure 4- Pattern distribution of ^{226}Ra in marl layer of study area.

Thorium-232

Naturally occurring thorium is mainly the single isotope ^{232}Th [17] which has a half-life of 1.4 x 10^{10} years. Hydroxides of thorium are the dominant species in soil and aquatic systems, although carbonate complexes also form [13]. The elements ^{208}Ti and ^{228}Ac are represented ^{232}Th [16]. Activity concentration of ^{232}Th of marl layer in is ranging from 4.4 Bq kg^{-1} to 6.2 Bq kg^{-1} with mean 5.16 Bq kg^{-1}. Any Th released by weathering has a transient existence in solution as it is strongly absorbed by clay minerals. Limestone is normally very low in Th, since Th^{4+} cannot form a stable carbonate similarly and almost completely absent from evaporate deposits. Long-term exposure to Th increases the chances of developing lung diseases and lung, pancreas and bone cancer.

Potassium-40

^{40}K is a radioactive isotope of potassium which has a very long half-life of 1.248×10^{9} years. Activity concentration of ^{40}K in marl layer is in minimum 174.4 Bq kg^{-1} and in maximum 255.4 Bq kg^{-1} with mean 223.98 Bq kg^{-1}. The percent of ^{40}K from total potassium percent was 0.72% because each 310 Bq kg^{-1} equal to 1% of potassium depending on [18]. The results obtained from marl layer are compared with data from other countries. The activity concentration means of ^{226}Ra and ^{232}Th is lower than most of them but the activity concentration means of ^{40}K is higher than most of them as shown in Table 2.

The obtained results indicate that the distribution of natural radionuclides in the marl samples is semi uniform. The overall values of ^{226}Ra, ^{232}Th, and ^{40K} are also much lower than the activity concentration of 50, 50, and 500 Bq kg^{-}1 for ^{226}Ra, ^{232}Th, and ^{40}K, respectively, in typical masonry [19]. The concentration of ^{40}K and ^{232}Th in marl layer depends upon the relative amounts of the clay minerals [20].

Table 2- Comparison specific gamma activities (Bq kg $^{-1}$) of the marl layer Euphrates Formation as raw materials with other countries.

Country	Specific activity (Bq kg $^{-}$1)			Reference
	^{40}K	^{226}Ra	^{232}Th	
Iraq/ marl	223.98	14.91	5.16	Present study
Pakistan	13.80	14.32	2.05	[20]
China	113.20	24.20	20.20	[21]
Greece	-	16.00	1.60	[22]
India	64.60	73.09	-	[23]

Country	Specific activity (Bq kg ⁻1)			Reference
	^{40}K	^{226}Ra	^{232}Th	
Italy	13.50	13.10	6.00	[24]
Syria	22.00	29.00	2.00	[25]
Algeria	36	16	13	[26]
Turkey	55.00	20.20	5.00	[27]
Egypt	19.30	20.40	4.40	[28]
Bangladesh	928.00	60.20	60.80	[29]
Hong Kong	30.00	3.00	5.00	[30]
Malaysia	222.00	40.70	25.90	[31]

Assessment of radiation hazard

The knowledge of radioactivity in these materials is important to estimate the radiological hazards on human health. Conversion factors to transform specific activities A_K, A_{Ra} and A_{Th} of K, Ra and Th, respectively, in the absorbed dose rate at 1m above the ground (in nGy h^{-1} by Bq kg^{-1}) are calculated by many equations. There are many hazard indices commonly used to measure the exposures to gamma rays in building materials.

Radium equivalent activity

Radium equivalent activity (Ra_{eq}) is used to assess the hazards associated with materials that contain ^{226}Ra,^{232}Th and ^{40}K in Bq kg^{-1} [32] because the not uniform of their distribution in the raw materials [33]. The radium equivalent activity is a weighted sum of activities of the ^{226}Ra, ^{232}Th and ^{40}K radionuclides based on the assumption that 370 Bqkg^{-1} of ^{226}Ra, 259 Bq kg^{-1} of ^{232}Th and 4810 Bq kg^{-1} of ^{40}K produce the same gamma ray dose rate [34]. Radium equivalent activity can be calculated formula 4-1 [35]. The published maximal admissible Ra_{eq} is 370 Bq kg^{-1} to keep the external dose below 1.5 mSv y^{-1} [32].

$$Ra_{eq} = 1.43A_{Th} + A_{Ra} + 0.077A_{k} \quad \text{..... (1)}$$

Where: A_{Th}, A_{Ra} and A_K are the activity concentration of ^{232}Th, ^{226}Ra and ^{40}K in Bqkg^{-1} respectively.
The Ra_{eq} in the marl layer is 37.97 Bq kg^{-1}. It appears acceptable value and no hazard because it is lower than the standard guideline.

Gamma index

In order to assess whether the safety requirements for building materials are being fulfilled, a gamma index proposed by the European Commission [36] was used formula 4-2 [37]:

$$I\gamma = \frac{A_{Ra}}{300} + \frac{A_{Th}}{200} + \frac{A_k}{3000} \quad \text{..... (2)}$$

The gamma dose rate due to building materials should be in the range of 0.3 – 1 mSv y^{-1} [36]. The value of Iɣ of marl layer is 0.0755 mSv y^{-1} which below the EC index. Consequently, it appears acceptable value and no hazard.

External hazard index

The external hazard index (H_{ex}) resulting from the exposure to gamma rays of naturally occurring radioactive materials has been evaluated as an estimate of radiation risk. In order to evaluate this index, a model 4-3 was used in the current study [35]:

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_k}{4810} \leq 1 \quad \text{..... (3)}$$

Where: A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K respectively.
This index value must be less than unity in order to keep the radiation hazard in significant [38]. The total air absorbed dose rate (nGyh^{-1}) due to the mean activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K (Bq kg^{-1}) can be calculated using the formula 4-3 [39]. The value of H_{ex} in the marl layer is 0.1068 nGyh^{-1}. The results appear acceptable value within the guideline limits and no hazard.

Internal hazard index

In addition to the external hazard, Radon (^{222}Rn), a radioactive noble gas with a half-life of about 3.8 days and daughter product of ^{226}Ra, accounts for half of the radiation dose to the general population [40] and is currently considered as a major source of lung cancer [41]. The internal exposure to Radon and its daughter products are quantified by the internal hazard index (H_{in}) which has been calculated by formula 4-4 relationship [35]:

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_k}{4810} \leq 1 \quad \text{(4)}$$

Where: A_{Ra}, A_{Th} and A_K are the mean activity concentrations of ^{238}U, ^{232}Th and ^{40}K, respectively, in (Bqkg^{-1}). H_{in} should be less than unity for a radiological safe material [42]. The H_{in} value in marl layer is 0.1471 nGyh^{-1}. The results appear acceptable value within the guideline limits and no hazard.

Discussion and Conclusion

The natural radioactive series notably ^{40}K, ^{226}Ra and ^{232}Th of marl samples have been measured by using the technique of gamma-ray spectroscopy with HPGe detector. The means concentration of radioactive elements ^{226}Ra, ^{232}Th and ^{40}K are 14.91, 5.16 and 223.98 Bq kg^{-1} respectively. The results of hazard parameters are: radium equivalent activity is 37.97 Bq kg^{-1}, the gamma index is 0.0755 mSv y^{-1}, the external hazard index is 0.1068 nGyh^{-1} and the internal hazard index is 0.1471 nGyh^{-1}. The high concentration of radiation in construction material is very harmful for all domestic uses including digestion and inhalation. Cancer is a major effect of low radiation doses expected from exposure to radioactive contamination especially cancers of the lungs, female breast, bones, thyroid and skin [43].

The marl layer is useful as a raw material because the concentrations of ^{232}Th, ^{226}Ra and ^{40}K are within the acceptable limits. The results analyses show that they are safe within the limits of the European Commission specifications [36] for building materials. The radiological indices are assessed and appeared safe for the workers in the quarry and cement plant because they are below the recommended limit of [32]. Therefore, the use of cement produced from marl in construction of dwellings is considered to be safe for inhabitants and posed no significant radiation exposure to occupants. There is no negative effect of radioactive radionuclides on health of workers in the quarry and cement plant.

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