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Study of Natural Radioactivity in Building Materials, and their Radiological Impact Assessment, in Marsa Matrouh Governorate, Egypt

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ABSTRACT

Fifty samples of ten types of building materials were collected from different locations of MarsaMatrouh Governorate, Egypt, and were analyzed using a gamma ray spectroscopy system. From the results, the observed values in the specific activities of226-Ra,232-Th andK-40 were within the national and international levels (10 Bq/kg for 226-Ra and 30 Bq/kg for 232-Th and 370 Bq/kg for K-40. The radiological hazards were assessed by calculating the radium equivalent activity (Ra_{eq}), the indoor absorbed gamma dose rate (DR), annual effective dose rate (HR), alpha index (Iα), gamma index (I_{γ}), and external hazard (H_{ex}) and internal hazard (H_{in}) indices. The estimated mean value of the absorbed dose rate of 117.63nGy h^{-1}isslightly higher than the world average value of 84 nGy h^{-1}, and the annual effective dose in the studied samples is 0. 144mSv y^{-1},was lower than the recommended limits. Multivariate statistical methods was applied to determine the existing relationship between radionuclides and radiological health hazard parameters and to identify the maximum contribution of radionuclide in radioactivity. The values of hazard indices were below the recommended levels; therefore, it is concluded that the buildings constructed from such materials are safe for the inhabitants.

KEYWORDS

Gamma Ray Spectroscopy; Building Materials; Hazard Indices,MarsaMatrouh Governorate.

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INTRODUCTION

Humans are continuously exposed to ionizing radiation from naturally occurring radioactive materials (NORM). Although the origin of these materials is the Earth's crust, they find their way into building materials, air, water, food, and the human body itself. Measuring the activity concentrations of radionuclides in building materials is important for the assessment of population exposures, as most individuals spend 80% oftheir time indoors;**El-Taher*et al.*, 2010 and Agbalagba*et al.*, 2014.**

Knowledge of the natural radioactivity in humans and the human environment is importantbecause naturally occurring radionuclides are the majorsource of radiation exposure to humans. It is an established fact that all of the construction materials containtrace amounts of natural radioactivity.Natural radioactivity is a major source of externaland internal radiation exposure to the occupants of the dwelling. The natural radioactivity in soil and building materials mainly comes from uranium series (Ra-226), thorium series (Th-232) and a radioactive isotope of potassium (K-40);**Ali Safdar*et al.*, 1996; Merle*et al.*, 2012 andZalewski*et al.*, 2001.**

All of these natural sources of radioactivity can be sources of both internal and external radiation exposure. Internal exposure occurs through the inhalation of radon gas, and external exposure occurs through the emission of penetrating gamma rays from radioactive sources. Therefore, it is important to measure the radioactivity levels in the built-up areas to assess the radiological consequences;**Quindos*et al.*, 1994 andViruthagiri*et al.*, 2011.**

Even more important is the knowledge of the amount of natural activity present in the materials that are used in the construction of dwellings. The amount of activity present in building materials will determine their use in the construction of dwellings. The knowledge of the natural radioactivity level is useful to set the standards and national guidelines used for providing recommendations. Due to increasing social concerns, a large number of research groups are engaged in the measurement of natural radioactivity in building materials at the national and worldwide levels;**Prasong*et al.*, 2008; Ravisankar*et al.*, 2012 andRavisankar*et al.*, 2014.**

The knowledge of the radioactivity present in construction materials helps to (i) assess the possible radiological hazards to human health and(ii) develop the standards and guidelines for the use and management of these materials. In this study, the concentrations of natural radionuclides were measured in 50 samples of ten types of building materials that are commonly used inMarsaMatrouh Governorate, Egypt; by means of gamma-ray spectrometry, with the aim of assessing the radiological hazards due to external gamma ray exposure in dwellings. The potential radiological hazards associated with these materials were assessed by calculating the radium equivalent activity (Ra_{eq}), absorbed gamma dose rate (DR), annual effective dose rate (HR), alpha index (I_{α}), gamma index (I_{γ}), and external hazard (H_{ex}) and internal hazard (H_{in}) indices. The obtained results were recommended to assess the radiation hazards to humans resulting from building materials.

Site description

MarsaMatrouh Governorate occupies a wide sector in the northwest of Egypt and is stretched from km 61 in the west of Alexandria up to the Egyptian- Libyan borders on the northern coast of Egypt Fig.(1);the area ofMarsaMatrouh governorate is about 212,112 km^2 (about 21% of Egypt's area) of which 3921.40 km^2 only is the total populated area. This area ranked the second largest Governorate in Egypt. The Governorate is characterized by a very low population density (population density of about 1.6 people per km^2).

MarsaMatrouh is the western gate of Egypt, a transit for travelers from East to West and the connective point between the countries of northern Africa and Asia continent. The north – western coast itself was used as a farm during the Greek and Roman eras where the cistern and Roman wells were used for irrigation.**WWW.nre@ idsc. Net. eg**

Fig. (1): Map of Marsa Matrouh governorate.

Geology: The geology ofMarsaMatrouh area is covered by sedimentary rocks that vary from limestone to lime-sandstone and marl. The country rocks belong to the Miocene, Pliocene and Pleistocene age.

Climate: The study area has a semi-arid Mediterranean climate, characterized by a brief, mild, rainy winter and long warm summer months (May to September), the temperature does not exceed 28.5° C in the summer and does not go below 10°C in winter) of clear sky, high radiation, and no rain, and the average humidity percentage is around 61.3% and 75.6% during the year. The picture changes in October when a windy and relatively rainy winter begins; the amount of rainfall is approximately 140 mm/yr. **Egyptian General Authority for Meteorology, 1999-2009.**

Natural resources

Water: The area depends mainly on winter rain for irrigation and most of the drinking water.In general the underground water is found at the sediments layers. Water generated by shallow well is limited in quantity. Underground water can also be found in the limestone layers available under the Fuka basin. It can give a rate of 20 m3/ hour for some wells with an average quality, which gives approximately 2 million m3/year, only 72 000 m^3/year are used which gives a special importance for the basin. Also, some additional potable water is provided, mainly to the population on the coast, through the water pipeline from the Nile through Alexandria, which extends MarsaMatrouh desalination units.**WWW.nre@ idsc. Net. eg**

Flora: The area has a heavy flora that begins at the coastal zone and extends to the rocky plateau. There are two kinds of flora in this area; the first kind: arks planted with olive, palm tree, wheat depending on rainfall and wells that are randomly distributed;the second kind: parks of coastal plants and herbs.

Soil: All the beaches are composed of white, loose carbonate sands, well-polished and round moving towards island. The loose carbonate sand gradually changes to fairly consolidate limestone-forming ridges that skirt to coast. The ridges are of marine origin and represent bars and depressions, which separate ridges from lagoons in which alluvial loam deposits are present, mixed with calcareous sand. Generally; the soil in the beaches – that is affected by salt – are unsuitable for cultivation; opposite to the solid in the wadies and around highways.**WWW. nre@ idsc. Net. eg.**

Economic activities

Tourism;When domestic tourists think of a summer destination, he thinks of MarsaMatrouh city, as a quiet coastal city, with many beaches, despite congestion in the summer months. **Agriculture,** According to governmental estimation of the agriculture state in MarsaMatrouh area the land use foragriculture is about 86 543 feddan, the main crops are wheat and barley. The main problem that faces agriculture is water, as local farmers depend on rainfall, so crops productivity varies according to rainfall. **Grazing;**Grazing of sheep; goats; camels in addition to some cattle and donkey breedingare the main activity for Bedouin.**Handicrafts and agro-Industry,** Women of the region mainly undertake handicrafts and agri-products. To a large extent they depend on agriculture and animal raw materials. The most important of these activities are:− Producing handmade carpets, blankets and tents, spinning wool, leather curing, embroidery andsewing; picking olives, drying peppermint, and producing olive oil; Breeding poultry and rabbits.**WWW.nre@ idsc. Net. eg.**

MATERIALS AND METHODS:

Sampling**:** A total of 50 samples of common building materials, i.e., brick, Marble, granite, ceramic, dolomite,mixture of fine dolomite with soil (MFDS), sand, gypsum, portalandcementand white cement, werecollected from eighteen different locations of MarsaMatrouh Governorate, Egypt. As shown in Fig. 1. The sampling locations were chosen according to the amount ofbuilding material used in the study area;the locations in the district were chosen for this investigation,the building material samples were directly takenfrom the Stores in these provinces, and the materials were studied in their natural form. Each samplewerelabeled and transported to the lab. The samples were crushed,dried in an oven at approximately 105°C,the sampleswere sealed in plastic containers,then stored for 30 daysto bring222-Rn and its short-lived daughter products intoequilibrium with226-Ra;**Ravisankar*et al.,* 2012**.

Radiometric analysis

Measurements of the activity concentrations ofRa-226, Th-232 and K-40in Bq/kg dry weight of the collected samples were performed with a counting time of 80,000 sec using gamma-ray spectrometry. Ge-detector was employed with adequate lead shielding, which reduced the background by a factor of approximately 95%. The concentrations of various radionuclides of interest were determined in Bq/kg using the count spectra. The gamma-ray photo peakscorresponding to 1.46 MeV (K-40), 1.76 MeV (Bi-214)and 2.614 MeV (Tl-208) were considered to correspondto the activities ofK-40, Ra-226 andTh-232, respectively,in the samples. The detection limit of the Ge - detector system forK-40, U-238 andTh-232 is 8.50, 2.21and 2.11 Bq/kg, respectively, for a counting time of80, 000 sec.

Measurements of the radiological parameters:

Radium equivalent activity (Ra_{eq})

To assess the radiation hazard associated with thebuilding materials used, the $Ra_{eq}$was evaluated, whereit is assumed that all of the decay products ofRa-226 andTh-232are in radioactive equilibrium with their pre-cursors. $Ra_{eq}$is calculated according to the followingformula; **Beretka*et al.,* 1985 andMalanca*et al.,* 1993**.

$$Ra_{eq} = 7A_{Ra} + 1.43A_{Th} + 0.07A_K$$

Where A_{Ra}, A_{Th}, and $A_K$are the specific activityconcentrations ofRa-226,Th-232, andK-40 in Bq/kg,respectively. This formula is based on the estimation that1 Bq/kgofU-238, 0.7 Bq/kgofTh-232 and 13 Bq/kgofK-40 produce the same gamma-ray dose rates. Thisindex (Ra_{eq}) is related to both internal doses due to theradon and external gamma doses;should havethe highest value of 370 Bq/kgfor safe use in buildingmaterials**El-Taher, 2010.**

Absorbed dose rate (DR)

The absorbed dose rate (nGy/h) in air fromterrestrial gamma radiation at 1 m above the groundis calculated after applying the conversion factors (nGy/h per Bq/kg) to transform the specific activitiesA_{Ra}, $A_{Th}$and $A_K$into the absorbed dose rate accordingto the formula provided by **UNSCEAR,2000; EC, 1999**.

$$DR\ (nGy/h) = 0.92 \times A_{Ra} + 1.1 \times A_{Th} + 0.080 \times A_K$$

Where A_{Ra}, A_{Th}, and A_K are the activity concentration of226-Ra,Th-232 andK-40 in (Bk/kg), respectively.

Annual effective dose rate (HR)

To estimate the annual effective dose rate, it is necessary to use the conversion coefficient from the absorbeddose in air to the effective dose (0.7 Sv/Gy) and theoutdoor occupancy factor (0.2 Sv/Gy) proposed by**UNSCEAR, 2000**; Therefore, the effective dose rate isdetermined as follows:

$$HR\ (mSv/y) = DR\ (nGy/h) \times 8760h \times 0.2 \times 0.7\ Sv/Gy \times 10^{-6}$$

$$OR\ HR\ (mSv/y) = DR\ (nGy/h) \times 0.00123$$

Where,*D*Ris the dose rate in nGy/h.

Hazard indices

Hazard indices for external gamma radiation (H_{ex}&I*γ*)

For the assessment of excess gamma radiation fromthe building materials to ensure of the safety of thebuilding materials, two indices were used in this paper.**Beretkaand Mathew,1985**introduced a hazard index for theexternal gamma radiation dose from building materialsas given below:

$$H_{ex} = A_{Ra} /370\ Bq/kg + A_{Th} /259\ Bq/kg + A_K/ 4810\ Bk/kg$$

The value of *H*exshould be below 1 to ensure the safeuse of building materials, which corresponds to the upperlimit of Ra_{eq}(370 Bk/kg). The European Commission(EC) proposed an index called the gamma index (Iγ) toverify whether the guidelines of EC for building material usage are met. Iγis calculated using the followingformula;**EC, 1999**.

$$I\gamma = A_{Ra}/ 300 + A_{Th} /200 + A_K/ 3000 \leq 1$$

The European Commission (EC) introduced a twodose criteria for the gamma dose of building materials: an exemption criterion of 0.3 mSv/yand an upperlimit of 1 mSv/y. Most of the countries apply theircontrol on the upper limit (1 mSv/y). If the exemptionlevel of 0.3 mSv/yis considered, then the values of Iγshould be below 0.5 for materials used in bulk (i.e., brickand cement); however, if the upper level of 1 mSv/yisconsidered, then the values of Iγshould be below 1 forsuch materials. For superficial building materials withrestricted use (i.e., tiles and board), Iγshould be below2 and 6, supposing control values of 0.3 and 1 mSv/y,respectively.

Hazard indices for internal alpha radiation (H_{in}& I_α)

The assessment of the excess radiation due to radongas from the building materials can be calculated bysome indices. Two indices, called the internal hazardindex (*H*in) and the alpha index (Iα), were studied in thispaper. Hincan be used for considering the excess internal radiation due to the inhalation of222Rn and its shortlived decay products from building materials, which isdefined as**El-Taher e*t al.*, 2012**: This quantity should be less than unity for the safe useof materials in the construction of buildings.

$$H_{in} = A_{Ra}/185\ Bq/kg + A_{Th}/259\ Bq/kg + A_K/4810\ Bk/kg$$

The quantityIαhas been proposed by **Krieger, 1981; and Stoulos, *et al.*, 2003**;and is given below:

$$I_\alpha = A_{Ra} /200$$

The recommended values of Iαand Iγare below 0.5and 1, respectively as **EC, 1999.**

RESULTS AND DISCUSSIONS

In this study the activity concentrations ofRa-226,Th-232 andK-40 for the collected samples were analyzed and representedin table (1);And from the obtained data, the concentration of Ra-226, Th-232 and K-40 in the analyzed samples are within the International level of the same type samples

Table (1) *Thenatural radioactivity in all samples.*

Materials	Ra-226	Th-232	K-40
Brick	28.63	26.91	240.0
Marble	10.67	6.19	19.83
Granite	44.07	65.04	1392.0
Ceramic	67.48	53.16	664.10
Dolomite	24.70	13.70	175.0
(MFDS)	14.60	5.25	77.20
Natural sand	<DL	<DL	<DL
Gypsum	10.40	9.31	161.0
Portland cement	8.67	16.30	2.35
White cement	21.80	15.60	76.90

DFMS = a mixture of fine dolomite with soil, <DL= under detection limits

Radium equivalent activity (Raeq)

According to the calculated mean values of the radium equivalent activities for all samples; are presented Table 2;the mean value of radium equivalent activities were85.59Bq/kg for brick, and 21.05Bq/kg for marble, and 244.26Bq/kg for granite and 194.63for ceramic and 57.77Bq/kg for dolomite and 28.05Bq/kg for (MFDS)and 0.3Bq/kg for natural sand and 36.11Bq/kg for gypsum and 32.16 Bq/kg for Portland cement and 50.03Bq/kg for white cement respectively.

Absorbed gamma dose rate (DR) and annual effective dose rate (HR)

According to the calculated mean values of the absorbed gamma dose rate (DR)nGy/h and annual effective dose rate (HR)mSv/y for all samples; are presented in Table 2, and fig 3. the mean value of the absorbed gamma dose rate (DR)nGy/h and annual effective dose rate (HR)mSv/y were 39.31nGy/h, 48.21mSv/yfor brick, and 9.42nGy/h, 11.56mSv/yfor marble, and 117.63nGy/h, 144.26mSv/y for granite and 90.63nGy/h, 111.14mSv/yfor ceramic

Table (2) *Radiation hazard indices in all samples.*

Materials	Dose (nGy/h)	Hex (Bq/kg)	Hin (Bq/kg)	Ra eq (Bq/kg)	AEDE (mSv/y)	Ic (Bq/kg)	I□ (Bq/kg)
Brick	39.31	0.232	0.309	85.59	48.21	0.31	0.62
Marble	9.42	0.057	0.086	21.05	11.56	0.073	0.146
Granite	117.63	0.661	0.779	244.26	144.26	0.936	1.872
Ceramic	90.63	0.526	0.708	194.63	111.14	0.712	1.424
Dolomite	26.86	0.156	0.223	57.77	32.94	0.209	0.418
MFDS	13.06	0.076	0.115	28.05	16.02	0.101	0.201
Natural sand	0.15	0.001	0.001	0.3	0.19	0.001	0.002
Gypsum	17.1	0.098	0.126	36.11	20.97	0.135	0.27
Portland cement	13.84	0.087	0.11	32.16	16.97	0.111	0.222
White cement	22.55	0.135	0.194	50.03	27.66	0.176	0.353

and 26.86nGy/h, 32.94mSv/y for dolomite and 13.06nGy/h, 16.02 mSv/y for (MFDS) and 0.15nGy/h, 0.19 mSv/y for natural sand and 17.1nGy/h, 20.97 mSv/y for gypsum and 13.84nGy/h, 16.97mSv/y for Portland cement and 22.55nGy/h, 27.66mSv/y for white cement respectively.

Hazard indices

The computed values of the alpha and gamma indices(Iα and Iγ) and the external and internal hazards ($H_{ex}$and H_{in}) are also given inTable (2). The obtained mean values of Iα and Iγinthe present study are lower than the recommended valuesof 1 and 0.5, respectively;EC,1999.The mean values of $H_{ex}$and $H_{in}$are belowthe recommended level of 2,therefore, the materialsincluded in this study can safely be used in the construction of buildings.

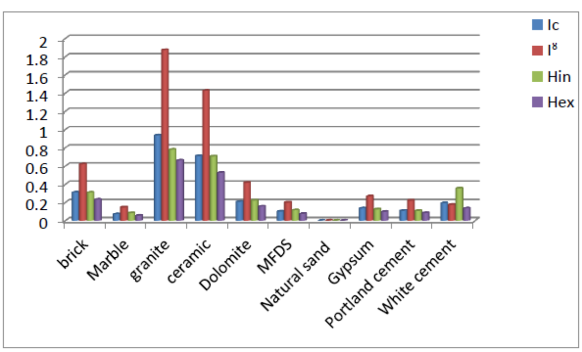

Fig. (2): Mean values of Hazard indices for samples.

CONCLUSION

Gamma ray spectrometry was used to determinethe radioactivity concentrationsRa-226,Th-232 andK-40in the studied samples of various building materialsfrom MarsaMatrouh Governorate, Egypt. Theradium equivalent activity, absorbed gamma dose rate inindoor air and the corresponding annual effective doserate, activity utilization index, and external and internal hazard indices of the radiological parameters werecalculated to qualify and quantify the radiological hazardassociated with the studied building materials. Hence, itis concluded that the radiological parameters obtainedare normal and within the recommended limits. Therefore, using of these building material samples underinvestigation in the construction of dwellings is considered to be safe for inhabitants. The results may be used as reference data forradiation assessment in building materials.

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مجلة
التقنيات النووية
فى العلوم التطبيقية
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الجمعية المصرية للعلوم الإشعاعية وتطبيقاتها
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دراسة النشاط الإشعاعي الطبيعي في مواد البناء، وتقييم أثرها الإشعاعي، في محافظة مرسى مطروح، مصر

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الهدف من هذه البحثهودراسة النشاط الإشعاعي الطبيعي لمواد البناء المستخدمة في محافظة مرسى مطروح، وتقييم الآثار الإشعاعية الناتجة عنها.ولذلك تم تجميع عدد خمسون عينه من المواد المستخدمة في أعمال البناء (٥ عينات من كل نوع من أغلب عشرة أنواع للمواد المستخدمة في أعمال البناء(مثل الطوب ، الرخام، الجرانيت، السراميك، الدلوميتناتج تكسير الكسارات،(MFDS) وهو عبارة عن مخلوط من التربةمع الدولوميت الناعم ناتج تكسير الكسارات،والرمل الطبيعي، الجبس، الأسمنت البورتلاندي، والأسمنت الأبيض.

وتم استخدام جهاز الجيرمانيوم عالي النقاوة لقياس النويدات المشعة الطبيعية لهذه العيناتوكانتالتركيزات لعناصر سلسلتي اليورانيوم-٢٣٨والثوريوم - ٢٣٢ ولنظيرالبوتاسيوم-٤٠ بيكريل/كجم داخل الحدود الدولية والمحلية للنويدات التي تم قياسها.وتم تقييم المخاطر الإشعاعية المحتملة من خلال حساب نشاط مكافئ الراديوم بيكريل/كجم ومعدل الجرعه الممتصة(نانوسيفرت/ساعة)ومعدل الجرعه الممتصة السنوية (نانو جراي/عام)ومعامل التعرض الخارجي ومعامل التعرض الداخليومؤشرألفا ومؤشر جاما.وخلصت هذه الدراسةإلي أن المباني التي شيدت من المواد محل الدراسة آمنة للسكان حيث أن الجرعة الفعالة سنويا في العينات بمنطقة الدراسة كانت أدنى من المستويات الموصى بها دوليا.

1. قسم المواقع والبيئة- هيئة الرقابة النووية والإشعاعية - ص. ب: 1557- مدينة نصر: 26711- القاهرة - مصر.