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Estimation of radiation hazard indices from natural radioactivity of some rocks

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Abstract Different samples of igneous and metamorphic rocks from Egypt and Germany have been considered to measure γ-ray activity concentrations due to naturally occurring, potentially hazardous radonuclides ^{226}Ra, ^{232}Th and ^{40}K. The radiation hazard parameters including radiation equivalent activity, gamma-absorbed dose rate, and external and internal hazard indices have been estimated. The gamma-absorbed dose rates in air of rocks in Egypt range from 4.2 to 128.5 nGy·h^{-1} with a mean value of 55.3 nGy·h^{-1}. For igneous and metamorphic rocks from Germany, the values of absorbed dose rates fluctuate from 5.1 to 148.6 nGy·h^{-1}, with a mean value of 60.9 nGy·h^{-1}. Generally, it is found that the radiation hazard indices in common igneous rocks are distinctly higher in acidic than in ultrabasic rocks. The results are discussed and compared with the corresponding published data.

Key words Natural radioactivity, Rocks, Hazard indices

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1 Introduction

Naturally occurring radionuclides ^{226}Ra, ^{232}Th and ^{40}K are widespread in the earth's environment and they exist in various geological formation such as soil, rock, water and plants [1]. These radionuclides pose exposure risks externally due to their γ-ray emissions and internally due to radon and its progeny, which emit alpha particles. Even though these radionuclides are widely distributed, their concentrations have been found to depend on the local geological condition and as such they vary from one place to another[2,3].

A significant report of data on the radioactivity contents of specific building materials had been stated by Hamilton [4]. He observed that the mean concentrations of the naturally occurring radionuclides in building materials used in the United Kingdom varied considerably. This variation is mainly attributed to the fact that the radioactivity concentration depends on geochemical characteristics of the raw materials. Negligible concentrations of ^{226}Ra, ^{232}Th and ^{40}K for thermal insulation materials such as rock wool and silica wood have also been reported.

In this article, the concentrations of ^{226}Ra, ^{232}Th and ^{40}K content of igneous and metamorphic rock samples as well as some building materials are estimated using γ-ray spectroscopy. The samples were collected from two different locations in Egypt and another in Germany. This article presents the results for the measured activities, the radium equivalent concentrations and the values of both internal and external hazard indices associated with the usage of some rocks and building materials.

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2 Experimental procedure

The concentration of natural radionuclides ^{226}Ra, ^{232}Th and ^{40}K in the material samples was determined using a low-background (HpGe) gamma spectrometry system. The measurement was carried out in ZSR, Hannover University. Using the liquid multi-nuclide standard solution 98-QCY48 (PTB, Germany), calibration was performed. The radionuclide mixtures are recommended by the National Institute of Standards and Technology (NIST, USA). The counting time for each sample was 54000 s to ahieve statistically smaller error levels. With appropriate corrections for laboratory background, the activity of ^{226}Ra was evaluated in all cases, from the 0.609 MeV and 1.76 MeV gamma rays of ^{214}Bi, while the ^{232}Th activity was determined from 0.238 MeV gamma rays of ^{212}Pb and 0.911 MeV gamma rays of ^{228}Ac, and the ^{40}K peak was at 1.46 MeV [5].

2.1 Radiation hazard indices for building materials

The distribution of ^{226}Ra, ^{232}Th and ^{40}K in soil is not uniform. Uniformity with respect to exposure to radiation has been defined in terms of radium equivalent activity (Ra_{eq}, in $Bq \cdot kg^{-1}$) to compare the specific activity of materials containing different amounts of ^{226}Ra, ^{232}Th and ^{40}K [6]. The radium equivalent activity is a weight sum of activity of the above three radionuclides based on the estimation that 370 $Bq.kg^{-1}$ of ^{226}Ra, 259 $Bq.kg^{-1}$ of ^{232}Th and 4810 $Bq.kg^{-1}$ of ^{40}K produce the same γ-ray dose rates. Ra_{eq} is given by

$$Ra_{eq} = (A_{Th} \times 1.43) + A_{Ra} + (A_K \times 0.077) \quad (1)$$

Another radiation hazard index called the representative level index, $I_{\gamma r}$, is defined as follows [7,8]

$$I_{\gamma r} = (1/150)\, A_{Ra} + (1/100)\, A_{Th} + (1/1500)\, A_K \quad (2)$$

where A_{Ra}, A_{Th} and A_K are the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K (in $Bq \cdot kg^{-1}$) respectively.

2.2 Calculation of absorbed dose rates

The outdoor air-absorbed dose rates ($nGy \cdot h^{-1}$) due to gamma rays at 1 m above the ground were calculated from ^{226}Ra, ^{232}Th and ^{40}K concentration values in some rocks and building materials. The conversion factors used to calculate the absorbed dose rate is given as [9]

$$D = 0.427 A_{Ra} + 0.662\, A_{Th} + 0.043 A_K \quad (3)$$

In the above conversion, it is assumed that all the decay products of ^{226}Ra and ^{232}Th are in radioactive equilibrium with their precursors.

3 Results and discussion

The results for the average absorbed dose rate, the radium equivalent activity and the representative level index are reported in Table 1. The maximum values are shown in acidic dykes samples while the minimum values are found in basalt. The average values of Ra_{eq} for Bir El-Sid, Wadi El-Gemal and Germany samples are 106.8, 93.9 and 146.4 $Bq \cdot kg^{-1}$ respectively. They are below the internationally accepted value (370 $Bq \cdot kg^{-1}$).

From the result it is evident that there are considerable variations in Ra_{eq} values of different materials and also within the same type of material originating from different areas. This fact is important from the point of view of selecting suitable materials for use in building and construction especially concerning the materials having large variation in their activities. For example, in granite the Ra_{eq} varies from 22.9 to 599.5 $Bq \cdot kg^{-1}$, i.e., from a low to very high level when compared with most of the other natural materials. Large variation in radium-equivalent activities may suggest that it is advisable to monitor the radioactivity levels of materials from a new source before adopting it for use as a building material.

The average values of gamma-absorbed dose rates in air of Bir El-Sid is 53.1 $nGy.h^{-1}$, which is comparable to the average terrestrial radiation dose rates (55 $nGy \cdot h^{-1}$). For Wadi El-Gemal area the value is lower than that of the average global value. For igneous and metamorphic rocks from Germany the values of absorbed dose rates fluctuate from 5.1 to 148.6 $nGy \cdot h^{-1}$ with an average value 72.4 $nGy \cdot h^{-1}$, which are comparable with the world range (28—120 $nGy \cdot h^{-1}$) and the average value (59 $nGy \cdot h^{-1}$) reported from normal background areas. [10].

Table 1 Average values of radiation hazard indices of different rocks from areas under study.

Type of rock	Absorbed dose rate / nGy·h^{-1}			Radium equivalent Ra_{eq} / Bq·kg^{-1}			Representative level $I_{\gamma r}$ / Bq·kg^{-1}		
	B.S	W.G.	GR	B.S.	W.G.	GR	B.S.	W.G.	GR
Granite	104.8	92.9	142.1	213.9	186.8	289	1.6	1.43	2.12
Serpentine	41.8	10.3	5.1	84.2	21.4	11.4	0.64	0.16	0.08
Metagabbro	43	18.1	22.6	84.7	35.9	47.9	0.66	0.3	0.35
Acidic dykes	72.2	88.9	148.6	144.3	176.1	291.2	1.1	1.37	2.29
Diorite	27.0	56.4	16.4	53.1	113.8	33.3	0.4	0.87	0.3
Lamprophyre	67.6		73.0	102		134.9	1.04		1.01
Dolerite	20.9	12.5	81.5	42.9	24	167.5	0.3	0.19	1.25
Basalt	3.40	...	....	7.3	....	...	0.05	...	....
Melange		26.9	….	...	52.8	...	...	0.41	....
Amphibolite	...	16.1	5.2	...	32.4	11.5	...	0.25	0.08
Granodiorite	94.50	....	....	189.7	....	....	1.45	....	....
Q-diorite	78.20	....	....	157.2	....	....	1.20	....	....

Note: B.S., W.G. and GR denote samples collected from Bir El-Sid area, Wadi El-Gemal area, Egypt and from Germany respectively.

The calculated values of $I_{\gamma r}$ for the samples studied in this work range from 0.05 to 2.29 Bq·kg^{-1}. Since most values are lower than unity, therefore, according to the radiation protection 112 report (European Commission, 1999) [11], samples from these regions are safe and can be used as a construction material without posing any significant radiological threat to population.

3.1 Calculation of annual effective dose

The value of the quotient of effective dose equivalent rate to absorbed dose rate in air is taken as in the UNSCEAR 2000 Report, to be 0.7 Sv per Gy for environmental exposure to gamma rays of moderate energy. This value is assumed to apply equally to males and females and to the indoor and outdoor environments. Taking the outdoor occupancy factor to be 0.2 and the average dose rate value (59 nGy·h^{-1}) reported for normal background areas [10], the annual effective dose equivalent from outdoor terrestrial gamma radiation is found to be

$$59\ (\mathrm{nGy \cdot h^{-1}}) \times 8.760\ (\mathrm{h}) \times 0.2 \times 0.7\ (\mathrm{Sv \cdot Gy^{-1}}) \\ = 70 \mu\mathrm{Sv} \tag{4}$$

The results showed that the calculated annual effective doses for diorite, dolerite, serpentine, metagabbro and basalt in Bir El-Sid area are lower compared to the world average of 70 µSv·a^{-1} [10].

Also, the absorbed dose rates due to natural radioactivity of 12.6, 23.2, 33, 19.7, 69.2 and 15.3 µSv·a^{-1} for serpentine, metagabbro, mélange, amphibolite, intermediate dykes and basic dykes, respectively, in Wadi El-Gemal are lower compared with the world average, which indicates that the level of the radioactivity concentrations in rocks around these areas is low. This, therefore, shows that the radiation burdens do not pose as a source of radiation contamination or hazard to the environment. But the values for Fawakher granitoide (113.4 µSv·a^{-1}), dyke rocks (88.5 µSv·a^{-1}) in Bir El-Sid and for granite (113.9 µSv·a^{-1}), acidic dykes (109 µSv·a^{-1}) in Wadi El-Gemal are higher than the world average. With respect to the different samples from Germany, the effective dose equivalent arising from the average absorbed doses in air are 174.3, 182.2, 100 and 80 µSv·a^{-1} for granite, acidic dyke, dolerite and lamprophyre which are higher than the world average. But for serpentine, gabbro, dolerite and amphipolite, the values are 6.3, 27.7, 20.1 and 6.4 µSv·a^{-1}, respectively.

3.2 External and internal hazard index

For limiting the radiation dose from building materials in Germany to 1.5 mGy·a^{-1} Krieger[12] proposed the following conservative model based on infinitely thick walls without windows and doors to serve as a criterion for calculating the external hazard index H_{ex}:

$$H_{\mathrm{ex}} = A_{\mathrm{Ra}}/370 + A_{\mathrm{Th}}/259 + A_{\mathrm{K}}/4810 \tag{5}$$

where A_{Ra} , A_{Th} and A_K are the activity concentrations of ^{226}Ra ,^{232}Th and ^{40}K (in Bq·kg^{-1}) respectively in building materials. This criterion considers only the external exposure risk due to γ-rays and corresponds to a maximum Ra_{eq} of 370 $Bq \cdot kg^{-1}$ for the material. The model was also accepted by the former Soviet Union[13] and Norway[14]. These very conservative assumptions were later corrected by Hewamanna[15] after considering a finite thickness of walls and the existence of windows and doors through the application of a weighing factor of 0.7 in each case. Therefore, the maximum permissible concentrations were increased by a factor 2, this means:

$$H_{ex} = A_{Ra}/740 + A_{Th}/520 + A_K/9620 \qquad (6)$$

The value of this index must be less than unity for the radiation hazard to be negligible, i.e. the radiation exposure due to radioactivity in construction material must be limited to 1.5 $mGy \cdot a^{-1}$. For the maximum value of H_{ex} to be less than unity the maximum value of Ra_{eq} must be less than 370 $Bq \cdot kg^{-1}$ [2] .

The calculated values of H_{ex} for the granite samples studied in this work range from 0.03 to 0.81, while for bricks and sand samples the range is 0.09 to 0.28 $Bq \cdot kg^{-1}$ and 0.03 to 0.1 $Bq \cdot kg^{-1}$, respectively.

According to the criterion formula (Eq. (6)) for gamma activity, it is indicated that the commonly used building materials in Hanover examined in this work could be used in building construction without exceeding the proposed radioactivity criterion level.

In addition to the external irradiation, radon and its short-lived products are also hazardous to the respiratory organs. The internal exposure to radon and its daughter products is quantified by the internal hazard index (H_{in}) which is given by the following equation[9] :

$$H_{in} = A_{Ra}/185 + A_{Th}/259 + A_K/4810 \qquad (7)$$

If the maximum concentration of radium is half of that of the normal acceptable limit then H_{in} will be less than 1.0. [12] For the safe use of a material in the construction of dwellings, H_{in} should be less than unity. The average calculated values were less than unity in most samples except for granite samples. The results presented in Table 2 indicate that the building material samples used in Egypt could be used in building construction without exceeding the proposed radioactivity criterion level.

Table 2 Average annual gonadal dose equivalent values and calculated index of building materials in selected samples

Type	H_{ex}		H_{in}	
	Germany	Egypt	Germany	Egypt
Granite	0.39	0.25	0.98	0.63
Red brick	0.18	0.18	0.45	0.40
Sand	0.06	0.11	0.13	0.29
Cement	0.20	0.21	0.53	0.46
Gypsum	0.06	0.19	0.14	0.57

The ICRP-60 [16] recommended that any exposure above the natural background radiation should be kept as low as reasonably achievable-ALARA-but below the individual dose limits, which for radiation workers averaged over 5 years is 100 mSv and for members of the general public is 1 $mSv \cdot a^{-1}$. These dose limits have been established on the prudent approach by assuming that there is no threshold dose below which there would be no effect. This means that any additional dose will cause a proportional increase in the chance of a health effect. This relationship has not yet been established in the low-dose range where the dose limits have been set. A lower value by a factor of 3 to 10 would now be needed to satisfy most regulations[2]. Therefore, granite samples with a Ra_{eq} of more than 370 $Bq \cdot kg^{-1}$ should not be used in the construction of dwellings. Most of the samples analysed in this work have Ra_{eq} values below this limit.

4 Conclusion

From the results it can be seen that the values of Ra_{eq}, $I_{\gamma r}$ and gamma-absorbed dose rates of granite samples vary appreciably from sample to another due to the variation of radium, thorium and potassium contents besides the region of collection. The average Ra_{eq} values for granite rocks in the studied areas are below the internationally accepted values 370 $Bq \cdot kg^{-1}$.

The values of gamma-absorbed dose rates in air of rocks in Egypt are comparable to the average terrestrial radiation of 55 $nGy \cdot h^{-1}$. For igneous and metamorphic rocks from Germany the values of absorbed dose rates fluctuate from 5.1 to 148.6 $nGy \cdot h^{-1}$, with a mean value of 72.4 $nGy \cdot h^{-1}$. The dose rates are comparable with the world range (28—120 $nGy \cdot h^{-1}$).

The effective dose equivalent values of diorite, dolerite, serpentine, metagabbro and basalt in Bir El-Sid and serpentine, metagabbro, mélange, amphibolite, intermediate dykes and basic dykes of Wadi El-Gemal in Egypt are also lower. But for Fawakher granitoide, dyke rocks in Bir El-Sid and granite, acidic dykes in Wadi El-Gemal the values are higher. With respect to the samples of Germany granite, acidic dyke, dolerite and lamprophyre, the values are higher.

H_{ex} and H_{in} values for samples collected from Egypt and Germany are less than unity except some granite samples, which are higher than the acceptable value. So it could be used in building construction without exceeding the proposed radioactivity criterion level.

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