Natural Radioactivity in Some Building Materials Originating from a High Background Radiation Area

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Abstract

Twenty four samples of building materials, collected from utilized quarries dispersed randomly in a high natural background radiation area, were analyzed for ^{226}Ra, ^{232}Th and ^{40}K by γ-spectrometry. This area lies in Hail province, Saudi Arabia. The collected samples were fragmented granites, granite gravels with clays, sands and crushed black rocks (mafic metavolcanic rocks). The results showed that the highest activity concentrations were found in the fragmented granite materials and ranged from 144-207, 671-1058 and 964-1440 Bq/kg with average values of 194, 912 and 1320 Bq/kg for ^{226}Ra, ^{232}Th and ^{40}K, respectively. The lowest activity concentrations were found in the black rock materials which ranged from 19-39, 47-125 and 212-306 Bq/kg with average values of 24, 82 and 255 Bq/kg for ^{226}Ra, ^{232}Th and ^{40}K, respectively. The radioactivity levels in the other materials lie somewhere in between. Granites and clays exceeded the proposed hazard indices for the usage as building materials and should be restricted, whereas the sands and the crushed black rocks complied with these indices and can be used without restrictions.

1. Introduction

Natural environmental radioactivity arises mainly from the primordial radionuclides of ^{232}Th and ^{238}U decay series, in addition to ^{40}K, which occur in trace levels in almost all ground formation. These radionuclides are formed by the process of nucleosynthesis in stars and are of half-lives comparable to the age of the cooled planet. The γ-radiation emitted from such radionuclides, also called terrestrial background γ-radiation, and represents the main source of radiation exposure to human body. The activity levels in different regions on the Earth's crust depend primarily on the local rock content of radionuclides, which varies widely with the geological composition of the rock formation in each region [1, 2]. Usually, igneous rocks of granitic composition are strongly enriched in thorium and uranium, compared to rocks of other composition [3-7]. On average 15 ppm of Th and 5 ppm of U in igneous rocks, compared to rocks of basaltic or ultramafic composition (<1 ppm) ([8, 9]. These radionuclides could pose potential health risk, especially if assisted by natural processes such as weathering deposition and wend erosion [10].

Accordingly, Airborne radiometric surveys on western Saudi Arabia, to estimate the γ-ray exposure rate arising from the surface geology have indicated higher γ-radiation on the granitic rocks of Aja heights of Hail province. An inhabited area, includes Hail city and some other scattered towns and villages, adjacent to this granitic massif, where the soil contains significant fractions of fragmented granites. Compared to the global average normal radiation levels of 0.46 mSv/y, the ground surveys in the inhabited and utilized zones in the region indicated higher terrestrial γ-radiation arising from the surface geology of the granitic rocks and the adjacent soils ranged from 0.7 to 9.82 mSv/y with average value of 1.81 mSv/y [11]. Several quarries are dispersed in this region and used to provide building materials as sands, fragmented granites, granite gravels and crushed metavolcanic rocks (black rocks). It was expected that these building materials may contribute to radiation dose to the inhabitants of homes and buildings that used these materials for construction.

This article presents radiological data on the natural radioactivity of some building materials provided from the utilized quarries dispersed in this province. Based on the obtained results, the contribution of these building materials to the radiation dose to the occupants of the building that used these materials has been assessed.

2. Experimental

2.1. Nature of the high radiation area

2.1.1. Location and Topography

The high radiation area includes Aja massif and the surroundings. It is bounded by lat 27° 00’ and 28° 05’, and long 41° 00’ and 42° 15’ E, and occupies an area of approximately 10500 km^2, in the northern part of the Arabian Peninsula (Fig. 1). It represents almost the major part of Hail quadrangle (Sheet 27E; international index NG-37-4) that bounded by lat 27° 00’ and 28° 00’ N., and long 40° 30’ and 42° 00’E.

Hail city lies at the foot of the Aja massif, in the central part of the area, at an elevation of about 980 m above sea level. It is one of the largest cities in north-central Saudi Arabia.

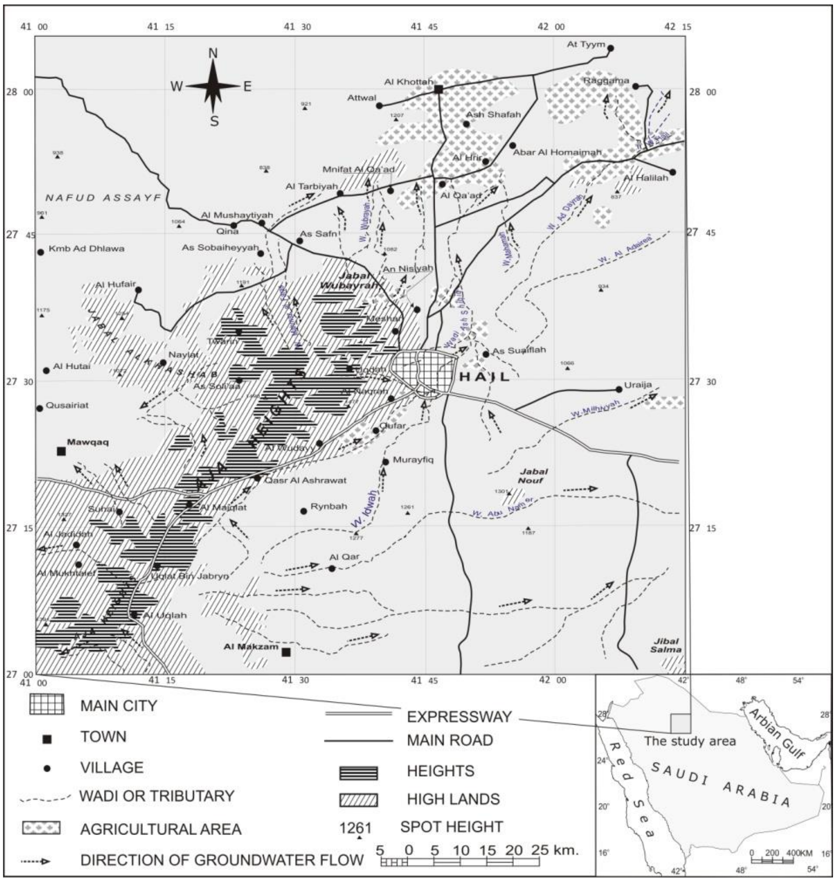

Fig. 1: Topographic map of the study area showing the groundwater flow directions (*Source: Adapted from topographic maps of Hai'l, Baq'a', Jubbah, and At Taysiyah quadrangles, sheets 27E, 27F, 27B, 27C,*

successively, scale 1 : 250000, Ministry of Petroleum and Mineral Resources, Kingdom of Saudi Arabia, 1987).

2.1.2. Geology

The high radiation area is dominated by Quaternary surficial deposits overlie most of the Phanerozoic bedrocks and parts of the Proterozoic basement of the Hail quadrangle. The deposits predominantly consist of eolian sand, and small occurrences of gravel, alluvium, and sabkhah. The study area is underlain by late Proterozoic volcanosedimentary and intrusive rocks of complex geology, and a Cambrian to early Silurian succession of essentially sedimentary rocks [12].

The Proterozoic rocks crop out in the south western part of the area, and predominantly consist of relatively young granitic intrusions that include monzogranite and more evolved alkali-feldspar granites of the Abanat suite; the latter-named occur chiefly as large batholiths in the center of the Hail quadrangle, where they form the topographically conspicuous Aja massif (Fig. 1).

2.2. Sampling

The samples were collected from the used building materials originating from the dispersed quarries in the region and classified by naked eye according to their lithologies. The samples were selected depending upon some factors as accessibility and utilization.

2.3. Measurement of ^{226}Ra, ^{232}Th and ^{40}K

In the laboratory, the samples were powdered, homogenized and packed into a 0.5-L Marinelli beaker and sealed. The sealed samples were stored for at least 1 month to attain secular equilibrium between the parent radionuclides (^{226}Ra and ^{232}Th) and their daughters before measurements by γ-spectrometry.

Instead of the absolute method, the relative method has been preferred in the measurement of natural radioactivity in the soil and rock samples. However, control samples (blank and standards) were prepared in an identical way. The used standards were the soil equivalent standards RGU-1, RGTh-1 and RGK-1 for measuring U-series, Th-series and ^{40}K radionuclides, respectively. These standards were supplied by the International Atomic Energy Agency (IAEA). The blank sample was 0.5 L- Marinelli beaker containing 0.5 L of distilled water.

Identical Marinelli beakers have been maintained for all measurements for a fixed 23 h counting time. Each peak area under the photo-peak, corresponding to the respective gamma line used, was subjected to background correction. The 1460.8 keV photo-peak was used to measure ^{40}K, whereas the high energy photo-peaks 1764 keV (from ^{214}Bi) and 2620 keV (from ^{208}Tl) were used to measure ^{226}Ra and ^{232}Th, respectively, to reduce the self-attenuation effect.

Assuming identical measurement conditions, the activity concentration (A), has been calculated for the radionuclides of interest using the following equation:

$$A\ (\mathrm{Bq/kg}) = A_s\ (C/C_s$$

Where, A_s is the certified activity concentration of the standard (in Bq/kg); C is the net count rate (in counts/sec.) of the sample at specific γ-ray line; C_s is the net count rate (in counts/sec.) of the standard at the same specific γ-ray line. The spectral analysis was performed with the aid of computer software.

For quality control purposes, IAEA-375 and IAEA-312 standard reference materials (soil matrix) were analyzed and the obtained results were compared with the reference values. The results reflect good performance of the procedure and the counting system, and data is given elsewhere [13].

2.4. Assessment of radiation hazards

To evaluate the radiation impact due to utilization of building materials originating from the region, the following radiation hazard indices were proposed.

2.4.1. Radium equivalent activity

To assess the radiological risk of a used building materials, it is useful to represent the activities due to ^{226}Ra, ^{232}Th, and ^{40}K by a single quantity, which takes into account the associated radiation hazard. A common index called "radium equivalent activity index (Ra_{eq})" has been introduced by Beretka and Mathew [14]. It is defined as:

$$Ra_{eq} = A_{Ra} + (10/7)\ A_{Th} + (10/130)\ A_{K}$$

Where, A_{Ra}, A_{Th} and A_{K} are the activity concentrations (Bq/kg) of ^{226}Ra, ^{232}Th and ^{40}K, respectively, in the evaluated material. For radiation risk from building material to be negligible, the maximum value of Ra_{eq} must be less than 370 Bq/kg. ^{238}U has been replaced by ^{226}Ra, a decay product of ^{238}U, as they are supposed to be in equilibrium and the contribution of the radiation hazard are from ^{226}Ra sub-series radionuclides.

2.4.2. External hazard index

Assuming 370 Bq/kg of ^{226}Ra, 259 Bq kg^{-1} of ^{232}Th and 4810 Bq/kg of ^{40}K produce the same gamma-ray dose rate, and limiting the external γ-radiation dose up to 1.5 mSv/y, a proposed "external hazard index (H_{ex})" has been introduced [14]:

$$H_{ex} = (A_{Ra}/370) + (A_{Th}/259) + (A_{k}/4810)$$

To keep the radiation risk negligible, the limit of H_{ex} is suggested to be less than unity.

2.4.3. Internal hazard index

In addition to external hazard, Radon and its progenies are internally hazardous to the respiratory organs. The internal exposure to radon and its daughter products is quantified by "the internal hazard index (Hin)", which is defined as [14]:

$$H_{in} = (A_{Ra}/185) + (A_{Th}/259) + (A_{K}/4810)$$

If the maximum concentration of ^{226}Ra is half that of the normal acceptable limit, then H_{in} will be less than unity. However, H_{in} is supposed to be less than unity for the safe use of a material in the construction of dwellings. The internal hazard is much more pertinent to the dwellers.

2.4.4. Gamma-radiation hazard index

The γ-radiation hazard index ($I_{\gamma r}$) is a representative level index which is defined as [15]:

$$I_{\gamma r} = (A_{Ra}/150) + (A_{Th}/100) + (A_{k}/1500)$$

This index can be used to estimate the level of γ-radiation hazard associated with the natural radionuclides in specific materials. Values of index $I \leq 1$ correspond to ≤ 0.3 mSv/y, while $I \leq 3$ correspond to ≤ 1 mSv/y.

According to this dose criterion, materials with $I \geq 3$ should be avoided, since these values correspond to dose rates exceed the limit 1 mSv y^{-1} of dose rate in air recommended for population [16].
Therefore, where the non-dimensional value of the activity index does not exceed unity, the material can be used without restriction [17].

2.5. Absorbed dose and annual effective dose

Outdoor
Absorbed dose rate at 1 m above the ground has been calculated from the respective specific activities A_{Ra}, A_{Th} and A_K of ^{226}Ra, ^{232}Th and ^{40}K, respectively, using the conversion factors, as mentioned in the expression below by Monte Carlo method [17]:

$$D\ (nGy/h) = 0.462\ A_{Ra} + 0.604\ A_{Th} + 0.0417\ A_k$$

In the calculation of annual effective dose, conversion factor of 0.7 Sv/Gy and the outdoor occupancy factor of 0.2 have been used in the expression below by Monte Carlo method [17]. However, the annual external effective dose rate ($D_{an,\ ext}$) is given by the following equation:

$$D_{an,\ ext}\ (mSv/y) = D\ (nGy/h) \times 8760\ (hy^{-1}) \times 0.2 \times 0.7\ (SvG/y) \times 10^{-6}$$
$$= 0.0012264\ D\ (nGy/h)$$

Indoor
To calculate the annual effective dose for indoor occupied area ($D_{an,\ in}$) of dimensions $4\,m \times 5\,m \times 2.8\,m$, assuming that the material is used in floor, ceiling and walls, the following expression has been used:

$$D_{an,\ in}\ (mSv/y) = (0.92\ A_{Ra} + 1.1\ A_{Th} + 0.08\ A_k)\ x\ (10^{-6}\ Gy/h)\ x\ (0.7\ Sv/Gy)\ x\ (24\ x\ 365\ x\ 0.8\ h/y)$$

Or

$$D_{an,\ in}\ (mSv/y) = 0.0049056\ (0.92\ A_{Ra} + 1.1\ A_{Th} + 0.08\ A_k)$$

Where, 0.7 is the conversion from Gray to Sievert for adults and 0.8 indoor occupancy conversion factor, as mentioned by Monte Carlo method [17].

2.6. Equipment

An ultra-low level γ-spectrometry system (from ORTEC) having 40% relative efficiency and resolution of 1.9 keV at 1332 keV was used in measuring all the samples. The system is interfaced with data acquisition system ORTEC DSPEC Jr. 2.0 coupled with MCA Emulation software MAESTRO-32, and gamma ray spectra analysis and MCA Emulation software Gamma Vision – 32. The detector is housed in a cavity of conventional ultra-low background shielding supplied by ORTEC. The detector has been energy-calibrated with a set of standard reference gamma sources such as ^{22}Na, ^{57}Co, ^{133}Ba, ^{137}Cs, ^{241}Am and ^{252} Eu point sources. The minimum detection limit of the HPGe detector system used in the present measurement for ^{226}Ra and ^{228}Ra were 1.3 and 1.1 Bq/kg, respectively. The minimum detection limit for ^{40}K was about 1 Bq/kg.

The minimum detectable activity (MDA) at 95% confidence level by a counting system for a sample of a definite size and definite count time was calculated using Currie equation [18].

3. Results and Discussion

Twenty four samples of building materials, used for instruction of dwellings, were collected from the utilized quarries and analyzed for ^{226}Ra, ^{232}Th and ^{40}K. The results are summarized and given in Table 1. The common used materials were fragmented weathered granites, granite gravels and clays, red-yellow and yellow sands and crushed mafic metavolcanic rocks (known as black rocks).

Table 1: Activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in the common building materials collected from the area.

Sample	No. of samples	Activity concentration, Bq/kg Average (min. max.)		
		^{226}Ra	^{232}Th (^{228}Ra)	^{40}K
Fragmented weathered granite	6	194 (144-207)	912 (671-1058)	1320 (964-1440)
Granite gravel + high clay %	5	180 (126-225)	843 (544-1010)	1344 (869-1519)
Granite gravel + less clay %	3	92 (75-140)	391 (190-672)	845 (708-945)
Fine reddish-yellow sand	6	29 (19-37)	158 (107-204)	139 (135-180)
Crushed black rocks	4	24 (19-39)	82 (47-125)	255 (212-306)

The results in Table 1 showed that the highest activity concentrations were found in the fragmented granite materials and ranged from 144-207, 671-1058 and 964-1440 Bq/kg with average values of 194, 912 and 1320 Bq/kg for ^{226}Ra, ^{232}Th and ^{40}K, respectively. The lowest activity concentrations were found in the crushed black rocks which ranged from 19-39, 47-125 and 212-306 Bq/kg with average values of 24, 82 and 255 Bq/kg for ^{226}Ra, ^{232}Th and ^{40}K, respectively. The radioactivity levels in the other materials lie in between.

The activity concentration of both ^{226}Ra and ^{232}Th follow the sequence: fragmented granites ≈ granitic gravel + high clay % (>30%) > granitic gravel + low clay % (<30%) > sand ≈ black rocks. This pattern of variation of natural radioactivity with lithology is consistent with the relative average abundance of uranium (and hence ^{226}Ra) and thorium in the common lithologic units [19].

3.1. Potential radiation hazards due to building materials

As mentioned earlier, about 98% of the external γ-dose rate from ^{238}U series is derived by ^{226}Ra subseries. So disequilibrium, if any, between ^{238}U and ^{226}Ra will not affect the dose estimation from the measurement of ^{226}Ra [20]. To assess the potential radiation hazards due to exposure to radiation release from a specific building material, potential radiation hazard indices based on the activity concentration were calculated. These indices were calculated based on the average activity concentration of each lithologic group of the collected samples. The results are given in Table 2.

Table 2: The average radiation hazard indices of the utilized materials of different lithologies.

Material	Hazard index			
	Ra_{eq}	H_{ex}	H_{in}	I_{yr}
Fragmented weathered granite	1598	4.3	4.8	11.3
Granite gravel + high clay %	1488	4.0	4.5	10.5

Granite gravel + less clay %	716	1.9	2.2	5.1
Reddish-yellow to yellow sand	265	0.7	0.8	1.9
Crushed black rocks	161	0.4	0.5	1.2

For any building material, it well be classified as safe material if it complied with the proposed values of the hazard indices, where the Ra_{eq} index should be < 370 and the other indices (H_{ex} and H_{in}) should be < unity [14] and $I_{\gamma r}$ Should be ≤ 3 [15]. Applying these regulation limits on each lithologic group of the analyzed materials, the average activity concentration of each group was used to calculate its hazard indices (Table 2).

The data in Table 2 showed that the fragmented granites and granite gravels with clays exceeded the proposed indices. However, it is recommended that these materials should be used under specific circumstances or excluded from utilization as safe building materials. In the other hand, the black rocks and the sands complied with these indices and do not represent significant radiological health risk and can be used for building construction without restrictions. It may be considered in general as safe materials.

3.2. Annual effective dose

The annual effective dose was also calculated, based on the average activity concentration values of the analyzed materials and the results are given in Table 3.

Table 3: The average dose rates (absorbed and annual effective) due to the use of the materials of the different lithologies in building construction.

Material	Average dose rates		
	Absorbed, nGy/h	Annual effective, mSv/y	
		indoor	out door
Fragmented weathered granite	695.5	6.31	0.85
Granite gravel + high clay %	648.4	5.89	0.80
Granite gravel + less clay %	313.9	2.86	0.38
Reddish-yellow to yellow sand	114.3	1.04	0.14
Crushed black rocks	71.3	0.65	0.09

The data in Table 3 showed that the fragmented granites and granite gravels with clays causing average absorbed dose several orders of magnitude higher than that of the global average of 55 nGy/h [21], and causing an average annual external effective dose rate higher than that of the global average of 0.46 mSv/y from the terrestrial radionuclides in the areas having normal background [22, 23]. The absorbed dose and annual indoor effective dose reported by Dziri et al [24] (<230 nGy/h and <1.1 mSv/y, respectively) are lower than that reported in Table 3. Generally, the average activity concentration and the accompanied calculated radiation hazard indices and doses in this study are high for the fragmented granites and the granite gravels with clays compared to reported values in other studies [24, 25].

3. Conclusions and recommendations

- For granites and clays, the average value of the health hazard indices were found greater than the proposed limits (exceeded the risk level) which shows potential risk due to radiation exposure if these materials are used in building construction.

- The average values of the health hazard indices were complied to the regulation limits in the black rocks and sands, indicating no need for controlling action due to the use of these building materials
- The options available to the competent authority to reduce the dose should be by considering these information, where any strategy considered in this regard should be justified (in the sense that it achieves a net benefit) and then optimized in accordance with the recommendations of ICRP (1989, 1991) in order to produce the maximum net benefit.

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