NATURAL RADIOACTIVITY AND RADIOLOGICAL RISK PARAMETERS IN LOCAL AND IMPORTED BUILDING MATERIALS USED IN SUDAN

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Abstract. Natural radioactivity levels in selected types of building materials used in Sudan were measured using gamma spectrometry. Radioactivity concentrations were measured in 52 samples including cements, porcelain ware, and ceramic tiles, cement blocks, and red clay bricks. Representative samples were powdered and stored in polyethylene Marinelli containers for four weeks to attain equilibrium between ^{226}Ra and ^{232}Th and their daughters before measurements. The average radioactivity (Bq.kg^{-1}) of ^{226}Ra, ^{232}Th, and ^{40}K, ranged between 12–40, 10–70, and 28–94, respectively in cement; 10–35, 12–28, and 87-143, respectively in cement blocks; 32–132, 26–87, and 285–1070, respectively in red bricks; and 8-527, 18–118, and 129–812, respectively in ceramics and porcelain tiles. The air absorbed dose rates ranged between 12.0±3.0 to 40.5±23.0 nGyh^{-1} in materials used in a superficial amount; 34.4±8.9 to 173.3±52.0 nGyh^{-1} in materials used in bulk. The annual effective doses were varied from 0.06 to 0.85 mSv. Excluding porcelain ware samples. The activity concentration indexes describing external and internal radiation hazards were within the acceptable limits. The study provides important baseline data for setting national regulatory control limits for protection against radiation.

Keywords: *gamma spectrometry, cements and bricks, gamma index, indoor radiation hazards*

Introduction

Building materials contain naturally occurring radioactive materials (NORM) due to the natural radionuclide content in their raw materials or due to the additives used in the manufacturing process, such as zircon sand which contains traces of uranium and thorium and constitutes the radioactive content in ceramic and tiles (IAEA, 2003). The radiological risk from external radiation exposure due to natural radioactivity in building materials is caused by terrestrial gamma radiation from ^{226}Ra, ^{232}Th, and ^{40}K NORM as well as the internal exposure from radon (EC, 1999; UNSCEAR, 2000; Righi and Bruzzi, 2006). Radon in building materials is the major cause of the incidence of fatal cancer in the global population (ICRP, 2019).

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Bearing in mind that certain levels of radiation exposures due to radioactive contents in building materials is inevitable, international radiation protection advisory bodies assert radiation exposure be kept as low as reasonably possible. As people spend nearly 80% of their time indoors, the main motive of radiological studies of building materials has been to limit public radiation exposure due to the natural radioactivity of the constituents of these materials.

Sudan is one of the largest countries in Africa where houses are built using stones and other local materials that are characterized by the local geological sands. This diversity of materials makes it difficult to study samples of materials used all over the country. We found only two studies on this subject. One is by Sam and Abbas (2001), who reported radioactivity in local and imported cement types used in Sudan. The other is by Salih et al. (2014), who studied the radiation exposure of workers in storage areas for building materials. Due to the dynamic nature of the construction industry, the issue of radioactivity in building materials remains an open question.

In this study, we aim to provide a comprehensive survey of radioactivity present in local and imported building materials in Sudan and its impact on the exposure of the public to radiation. Further, we aim to provide up-to-date information on this subject to assist institutions in formulating regulations on the national level to control radiation.

Materials and Methods

Sampling

In total, 52 samples of local and imported building materials of widely distributed origins were collected from stores in the Sudanese capital, Khartoum. A radioactivity measurement in building materials is important for radiation protection purposes. Representative samples of building materials were powdered and stored in polyethylene Marinelli containers for four weeks to allow equilibrium between ^{226}Ra and ^{232}Th and their daughters to be reached.

Measurements of radioactivity

Sample specific activities were determined using the p-type high purity germanium (HPGe) gamma spectrometry with 30% relative efficiency to (NaI)Tl gamma spectroscopy for 1.33 MeV ^{60}Co radiation of 122 and 1332 keV gamma line of 0.875 and 1.850 keV, respectively (Baltic Instrument, Riga, Latvia). Energy and efficiency calibrations were performed using a mixed radionuclide gamma calibration standard (Amersham Buchler B1575). For the radioactivity quantifications, ^{226}Ra activities were determined from ^{214}Pb (351.92 keV) and ^{214}Bi (609.31 keV). The gamma energy lines of ^{212}Bi, ^{212}Pb, and ^{228}Ac were used to determine the ^{232}Th activity. The radioactivity of ^{40}K was determined from the 1460.81 keV gamma line (IAEA, 2003). The activity concentration (A) in Bqkg^{-1} in samples was calculated using the following expression:

$$A = \frac{N}{PE \times \mathcal{E} \times Tc \times M} \quad \text{(Eq.1)}$$

where, M denotes the mass of sample in kg, N the sample net area in the peak, PE the gamma emission probability, Tc the counting time, and $\mathcal{E}$ denotes the photopeak efficiency. Uncertainty in the activity concentration was determined at a 95% confidence level (σ =2) using *eq. (2)*:

$$\frac{u(C_{sp})}{C_{sp}} = \sqrt{\left(\frac{u(N)}{N}\right)^2 + \left(\frac{u(P_E)}{P_E}\right)^2 + \left(\frac{u(\mathcal{E})}{\mathcal{E}}\right)^2 + \left(\frac{u(T_C)}{T_C}\right)^2 + \left(\frac{\Delta M}{M}\right)^2} \qquad \text{(Eq.2)}$$

where, $\frac{u(N)}{N}$, $\frac{u(P_E)}{P_E}$, $\frac{u(\mathcal{E})}{\mathcal{E}}$, $\frac{u(T_C)}{T_C}$, and $\frac{\Delta M}{M}$ are the relative uncertainties in the counting rate, gamma emission probability, photopeak efficiency, counting time, and sample mass, respectively. Uncertainty in the activity concentration was determined by the SpectraLineGP software at 1 σ (Baltic Instrument, Riga, Latvia). The overall uncertainty in the determined activity was the square root of the quadratic sum of different activity parameters at a 95% confidence level (σ =2) according to the ISO standard (ISO, 1995).

Radium equivalent activity (Raeq)

In comparing radioactivity amounts in different samples, the radium equivalent activity (Ra_{eq}) is used (Beretka and Mathew, 1985). Ra_{eq} represents the combined specific activities of ^{226}Ra, ^{232}Th, and ^{40}K and determined according to the following formula:

$$Ra_{eq} = A_{Ra} + 1.43A_{th} + 0.077A_K \qquad \text{(Eq.3)}$$

where, A_{Ra}, A_{Th}, and A_K are the specific activity of ^{226}Ra, ^{232}Th, and ^{40}K, respectively.

Air absorbed dose rates

The indoor gamma absorbed dose rates in the air arising from radioactivity in building materials can be estimated using the radionuclide-specific dose rates conversion coefficients (nGy h^{-1} per Bq kg^{-1}). For materials of superficial use, such as ceramic and porcelain tiles, absorbed dose rates are determined as (EC, 1999):

$$D = 0.12.C_{Ra} + 0.14.C_{Th} + 0.0096.C_K \qquad \text{(Eq.4)}$$

For bulk use materials such as concrete and red brick, absorbed dose rates are determined as (EC, 1999):

$$D = 0.92.C_{Ra} + 1.1.C_{Th} + 0.08.C_K \qquad \text{(Eq.5)}$$

Effective doses

The effective dose is an important dosimetry quantity to estimate radiation risk. It facilitates comparison among different exposure categories. According to UNSCEAR (2000), a value of 0.7 Sv/Gy absorbed dose to effective dose conversion coefficient and 0.8 as an indoor occupancy factor. Therefore, an effective dose can be determined from air absorbed dose rates as follows:

$$E(Svy^{-1}) = D(nGyh^{-1}) \cdot 8760hy^{-1} \cdot 0.8 \cdot 0.7SvGy^{-1} \cdot 10^{-3} \qquad \text{(Eq.6)}$$

The gamma indexes

The gamma index (I_γ) is an important single quantity used in determining whether the external absorbed dose originating from radioactive content in building materials are within the recommended annual dose limit for the public (EC, 1999): I_γ is determined from the respective radioactivity of the three natural radionuclides as follows:

$$I_\gamma = \frac{C_{Th}}{200\ Bq/kg} + \frac{C_{Ra}}{300\ Bq/kg} + \frac{C_K}{3000\ Bq/kg} \quad \text{(Eq.7)}$$

where, C_{Ra}, C_{Th}, and C_K are the radioactivity of ^{226}Ra, ^{232}Th, and ^{40}K, respectively.

Alpha index (I_α)

This is another parameter used in estimating the internal radiation exposure attributed to radon from uranium isotopes in building materials. With A_{Ra} being radioactive (Bq.kg^{-1}) of ^{226}Ra, I_α, is defined as follows (Righi and Bruzzi, 2006):

$$I_\alpha = \frac{A_{Ra}}{200\ Bq{\cdot}kg^{-1}} \quad \text{(Eq.8)}$$

Building materials with $I_\alpha < 1$ corresponding to radium radioactivity of 200 (Bq.kg^{-1}) are considered safe for building constructions (NORDIC, 2000). The gamma index (I_γ) and alpha index (I_α) represent the levels to which the annual dose must be limited. The limits are summarised in *Table 1*.

Table 1. *The gamma index (I_γ) and alpha index (I_α) limits to achieve the recommended annual dose limits*

Dose criterion	**0.3 mSv/y**	**1 mSv/y**
Activity concentration index (I_γ)		
Materials used in bulk amount: Concrete, red bricks Materials used in ssuperficial amount: porcelain and ceramic tiles	I ≤ 0.5 I ≤ 2	I ≤ 1 I ≤ 6
Alpha index (I_α)		
All building materials		I ≤ 1

Statistical analysis

The statistical analysis has been carried out using Pearson correlation and was considered significant at P-value <0.05, which indicates strong evidence against the null hypothesis.

Results and Discussion

Natural radioactivity and radiological risk parameters were measured on 52 types of local and imported building materials used in Sudan. *Table 2* shows the type of building materials, their uses, and their countries of origin.

Radioactivity and radium equivalent activity (Raeq)

Table 3 presents the statistical summary of radioactivity concentrations in selected building materials used in Sudan. The boxplot distribution of activity concentrations ^{226}Ra, ^{232}Th, and ^{40}K natural radionuclides is shown in *Fig. 1*. As shown, mean ^{226}Ra activity concentrations ranged from 31.6–131.7 Bq.kg^{-1} in red bricks to 228.4±172.6 Bq.kg^{-1} in Italian porcelain (IP), whereas the mean ^{232}Th activity concentrations of ^{232}Th ranged from 6.2±2.0 (SC) to 82.8±20.3 Bq.kg^{-1} in (CP). For ^{40}K, average activity concentration ranged from 62.2±27.1 (SC) to 721.6±319.5 in red bricks from Sudan. The considerable variation in activity concentrations may be ascribed to the raw materials and industrial by-products of present building materials, as these replicate the geological characteristics of the sites of their origin.

Table 2. *Distribution of building materials, their uses, and the country of origin*

Code	Type	Uses	Sample size	Country of origin
PC	Tiles (Porcelain)	Superficial	5	China
CC	Tiles (Ceramic)	Superficial	8	China
PI	Tiles (Porcelain)	Superficial	7	Italy
CE	Tiles (Ceramic)	Superficial	4	Egypt
TS	Tiles (Ceramic)	Superficial	6	Sudan
CS	Cement	Superficial/Bulk	10	Sudan
BC	Concrete block	Bulk	6	Sudan
RB	Red brick	Bulk	6	Sudan

Table 3. *Statistical summary of the activity concentration and radium equivalent activity (Bq.kg^{-1}) for ^{226}Ra, ^{232}Th, ^{40}K in selected types of building materials used in Sudan*

Type	A_{Ra}		A_{Th}		A_K		Ra_{eq}	
	Mean	**Range**	**Mean**	**Range**	**Mean**	**Range**	**Mean**	**Range**
Materials used in superficial amount								
ST	22.9	7.9-29.8	28.2	17.5-38.6	662.1	438.5-812.5	114.2	83.1-128.3
CC	48.2	19.3-84.6	32.6	22.2-41.1	169.4	129.4-238.9	107.9	79.3-161.8
CP	158.5	87.6-215.2	82.8	59.0-113.4	489.8	378.4-607.3	314.6	23.8-41.9
IP	228.4	78.3-526.8	69.9	41.5-101.6	346.5	238.9-428.1	355.1	159.2-646.0
EC	58.0	47.2-69.2	53.7	33.1-80.4	398.3	262.4-515.5	165.4	129.8-209.0
Materials used in bulk amount								
SC	24.6	12.0-39.6	6.2	1.4-8.7	62.8	28.1-93.7	38.2	21.3-51.6
BC	23.1	9.8-35.0	21.9	11.7-28.0	108.8	87.0-143.0	62.7	34.4-86.1
RB	63.9	31.6-131.7	51.6	25.8-86.9	721.6	284.5-1070.0	193.3	120.2-290.8

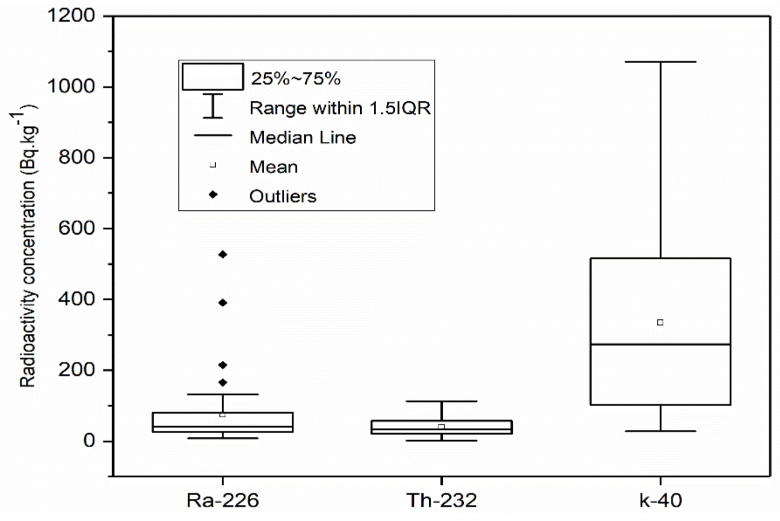

Figure 1. *Boxplot distribution of activity concentrations ^{226}Ra, ^{232}Th and ^{40}K natural radionuclides showing the interquartile range (IQR)(25th to 75th percentile), the range within 1.5IQR, the median and the mean lines and the outliers*

Of particular interest is the high radioactivity content of the porcelain samples (CP & IP), which show radioactivity approximately one order of magnitude higher than other subjects. Despite their high radioactivity content, as porcelain is used superficially and in small amounts, their relevant radiological risk is expected to be low as compared to other materials that are used in bulk amount. Elevated radioactivity of porcelain samples is related to their zirconium silicate content known for its high natural radionuclide content (IAEA, 2003).

In conformity with the previous observations, the average activity concentrations in building materials obtained in this study and reported in the literature are higher than those found in the earth's crust, being 35, 30, and 400 Bq.kg^{-1} of ^{226}Ra, ^{232}Th, and ^{40}K, respectively (UNSCEAR, 2000).

The activity concentrations of natural radionuclides ^{226}Ra, ^{232}Th, and ^{40}K are not uniform in a given sample, which necessitates setting of a single parameter to represent the total radioactivity, the radium equivalent activity, to be used for comparing the specific activity of materials containing different amounts of ^{226}Ra, ^{232}Th and ^{40}K. The human body treats radium in a similar way to calcium accumulating in human body bones resulting in internal exposure that could cause bone cancer considered to be among the most common radiation cacogenics (NRC, 1988).

As observed in *Table 3*, Ra_{eq} values ranged from 22.9±7.9 to 355.1±195.1 Bq.kg^{-1}. These values fall below the maximum permissible value of 370 Bq/kg (UNSCEAR, 2000).

Table 4 shows the comparison of activity concentrations (Bq.kg^{-1}) in building materials obtained in this study with those found in the literature. As seen, the activity concentrations in cement found in this study are comparable with or below those observed in previous studies performed in Jordan, Turkey by IAEA and EC (EC, 1999; IAEA, 2003; Mavi and Akkurt, 2010; Salih et al., 2014; Shayeb et al., 2017). The activity concentrations in porcelain and ceramic tiles used in small amounts are comparable with or higher than those reported in the literature.

As portrayed in *Table 4*, there exist wide differences between radioactivity in building materials in our study and those reported by Salih et al. (2014); this could be ascribed to differences in the origin of the studied building materials and this emphasises the importance of regular monitoring radioactivity levels in the materials used for building construction to ensure the safety of the occupants.

Radiation doses

The indoor dose rates from ^{226}Ra, ^{232}Th, and ^{40}K in building materials used in this study (excluding red brick) ranged from 12.0–173.0 nGyh^{-1} (*Table 5*). The determined absorbed doses are comparable to the world average for areas having normal background radiation (55 μGyh^{-1}) (UNSCEAR, 2000). The obtained average annual effective dose values ranged from 0.06 to 0.85 mSv and are, therefore, below the annual dose limit for the public (1.0 mSv).

Radiological risk parameters

The radiological risk to an individual from radioactivity content in building materials is better described using gamma index, I_γ for external exposure and the alpha index (I_α) for internal exposure to radon. I_γ and I_α index limits prescribed for achieving the recommended annual dose limits are summarised in *Table 1* (EC, 1999).

Table 4. *Comparison of activity concentrations (Bq.kg^{-1}) in building materials obtained in this study with the literature*

Country	A_{Ra}	A_{Th}	A_K	Reference
Soil, world average	**35**	**30**	**400**	**UNSCEAR (2000)**
Cement uses in bulk and superficial amount				
Turkey	26	10	130	Mavi and Akkurt (2010)
Sudan	15	33	230	Salih et al. (2014)
Jordan	37-121	54-142	255-621	Shayeb et al. (2017)
IAEA	7–180	7–240	24–850	IAEA (2003)
European Community	50(25–87)	35(10–70)	235(38–587)	Trevisi et al. (2018)
Sudan	25(12±40)	6.1(1.4–8.7)	63(28–94)	This study
Cement block uses in bulk				
Sudan	11.5±1.4	14.98±1.4	378.37±42.0	Salih et al. (2014)
European Community	59(14–272)	34(5–138)	340(17–685)	Trevisi et al. (2018)
IAEA	1–250	1–190	5–1570	IAEA (2003)
Sudan	23(10–35)	22(12–28)	109(87–143)	This study
Red brick uses in bulk				
Turkey	58.9	11.7	248.8	Mavi and Akkurt (2010)
Sudan	20.4 ± 2.1	58.1 ± 3.1	459.6 ± 116.8	Salih et al. (2014)
Jordan	13-19	55-63	160-202	Shayeb et al. (2017)
IAEA	1–200	1–200	60–2000	IAEA (2003)
European Community	51(7–84)	49 (4–102)	555(59–805)	Trevisi et al. (2018)
Sudan	64(32–132)	52(26–87)	722(285–1070)	This study
Tiles /Porcelains/Ceramics used in superficial amount				
Italy	84	54	609	Righi et al. (2009)
Turkey	97	68	471	Turban et al. (2013)
Sudan	31	60	486	Salih et al. (2014)
IAEA	30–200	20–200	160–1410	IAEA (2003)
Sudan	105(8–527)	52(18–118)	393(129–812)	This study

Table 5. *Statistical summary of doses and the radiological hazard parameters: D (nGyh^{-1}), E (mSvy^{-1}), I_γ and I_α estimated from the radioactivity concentrations in building materials used in Sudan*

Type	D (nGyh-1)		E (mSvy-1)		I_γ		I_α	
	Mean	Range	Mean	Range	Mean	Range	Mean	Range
Materials used in superficial amount								
ST	13.1	9.7-14.7	0.06	0.05-0.07	0.43	0.32-0.49	0.57	0.42-0.64
CC	12.0	8.5-18.2	0.06	0.04-0.09	0.41	0.26-0.64	0.54	0.40-0.81
CP	35.3	23.8-41.9	0.17	0.12-0.21	1.23	0.81-1.50	1.57	1.06-1.85
IP	40.5	17.9-75.7	0.20	0.09-0.37	1.49	0.63-2.97	1.78	0.80-3.23
EC	18.3	14.6-22.7	0.09	0.07-0.11	0.60	0.51-0.71	0.83	0.65-1.04
Materials used in bulk amount								
SC	34.4	20.2-46.3	0.17	0.10-0.23	0.16	0.10-0.23	0.19	0.11-0.26
BC	54.0	30.1-74.4	0.27	0.15-0.37	0.22	0.12-0.32	0.31	0.17-0.43
RB	173.3	111.1-253.0	0.85	0.55-1.24	0.73	0.47-1.10	0.97	0.60-1.45

For ceramics and tiles used in a superficial amount, mean I_γ values ranged from 0.41 to 1.49 and are less than 2, which corresponds with the annual dose limit of 0.3 mSv. These materials can be exempted from regulatory control. For the cement blocks and red bricks, which materials are used in bulk amount, I_γ values ranged from 0.16 to 0.73. These materials exceed the lower limit of I_γ and, therefore, need to be subjected to regulatory control. As presented in *Table 4*, I_α for CP & IP porcelain was 1.57±0.37 and 1.78±0.98, respectively. These levels exceed the prescribed limit of < 1 indicating that the use of this imported decorative material poses risk to the safety of the occupants.

Correlations between ^{226}Ra, ^{232}Th and ^{40}K activities

Table 6 shows the Pearson correlation coefficients of ^{226}Ra, ^{232}Th, and ^{40}K radionuclides. The results are graphically depicted in *Fig. 2* and *Fig. 3*. A significant correlation was observed between ^{232}Th and ^{40}K (R = 0.42, P<0.05); and a highly significant correlation was observed between ^{226}Ra and ^{232}Th (R = 0.68, P<0.001). The correlations observed could be due to the raw materials and industrial by-product of the building materials being of the same origin.

Table 6. *Pearson correlation coefficients between ^{226}Ra, ^{232}Th, and ^{40}K*

Radionuclide	**^{226}Ra**		**^{232}Th**		**^{40}K**	
	Pearson corr.	**p-value**	**Pearson corr.**	**p-value**	**Pearson corr.**	**p-value**
^{226}Ra	1	**	0.68*	<0.001	0.15	0.28
^{232}Th	0.68*	<0.001	1	**	0.42*	0.002
^{40}K	0.15	0.28	0.42*	0.002	1	**

Correlation significant at p<0.05; and highly significant at p<0.001

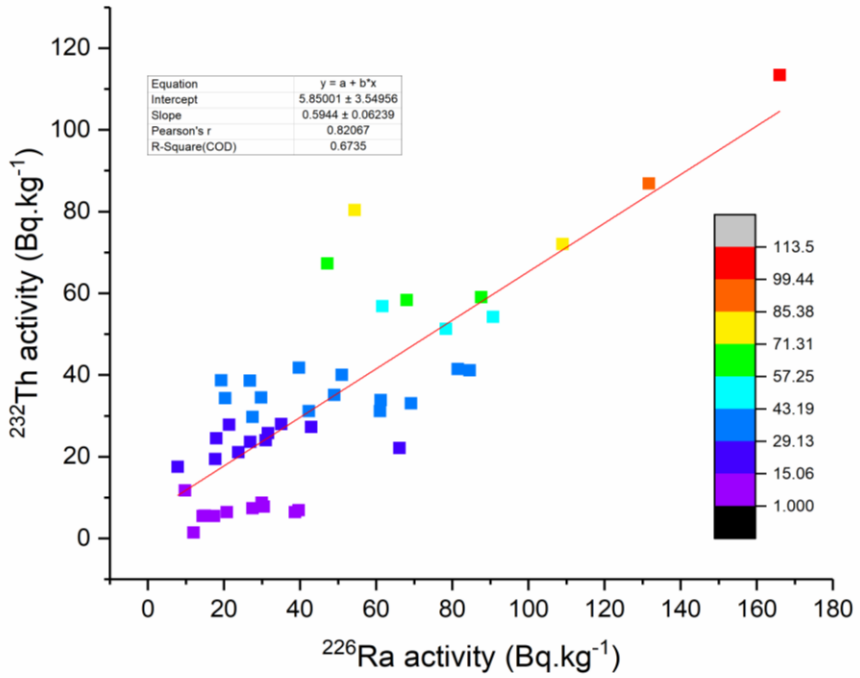

Figure 2. *Correlation between of activity concentrations ^{226}Ra and ^{232}Th natural radionuclides showing colour codes for the corresponding ^{232}Th activity*

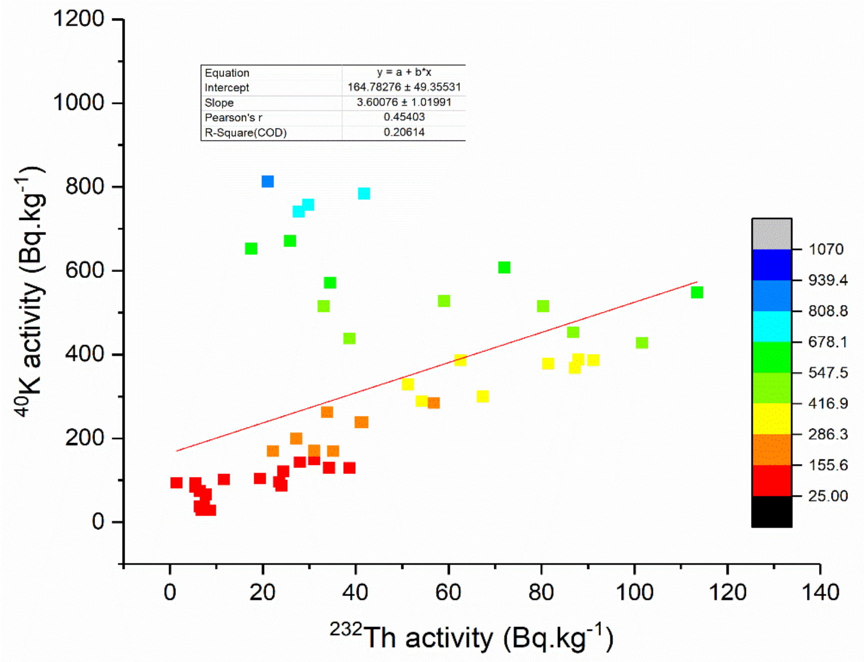

Figure 3. *Correlation between of activity concentrations ^{226}Ra and ^{40}K natural radionuclides showing colour codes for the corresponding ^{40}K activity*

Conclusion

Natural radioactivity levels in the selected building materials used in Sudan were measured using gamma spectrometry. The average values obtained are comparable to those reported in the literature. Continuous demand for both local and imported building materials necessitates regular monitoring of radioactivity levels to protect the occupants of buildings from radiation. The current data provide important radioactivity levels. Their benchmarking is the first step towards acquiring large scale data that would make it possible to set national regulatory control limits for protection of the public from radiation in buildings. These results indicate that considering the various types of dwellings constructed in various states and regions of Sudan and the variety of building materials used in their construction, establishment of a national research project to study radioactivity levels in building materials and evaluate the risk they pose to the population, is strongly recommended. Such a project should also consider investigating radon gas in dwellings and workplaces to establish safety limits to mitigate the radiological risk associated with radioactivity in building materials.

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Declaration of Conflicting Interests. The authors have no conflict of interests to declare.

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