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Estimation of the Concentrations and Risks of Natural Radionuclides in Rocks Used as Building Materials

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Abstract: ***Radiation exposure is exceedingly detrimental to inhabitants, especially at elevated doses. Diverse materials, including soil, water, rocks, vegetation, sand, and construction materials, subject us to radiation daily. Construction materials account for a substantial proportion of radiation exposure, particularly in regions with elevated natural radioactivity, due to prolonged indoor occupancy, which can markedly elevate the risk of health complications, including cancer, over time. This study seeks to assess terrestrial radioactivity levels in diverse construction material samples and to evaluate the associated health risks to humans from gamma radiation exposure. To achieve this objective, samples from the Al-Mahra region in eastern Yemen were collected and analyzed using high-resolution gamma-ray spectrometry with a high-purity germanium (HPGe) detector system. The results indicate that the mean activity concentrations of the primary radioisotopes (radium-226, thorium-232, and potassium-40) in the ignimbrite and ceramic materials exceed global averages, while the average value in the black cement sample aligns with the global mean. The radiological hazards encompass Raeq, DR, AEDE (indoor and outdoor), Hex, Hin, ELCR, and AGDE, which were computed to assess radiation risks to the population. In the ignimbrite rock, twice the threshold limit was observed; similarly, in the ceramic sample, certain radiological hazards exceeded globally permissible limits, while others remained below the recommended international threshold. The black cement sample significantly fell short of the recommended threshold. This study has established a base guideline regarding the levels of naturally occurring radionuclides within the investigated area, which can be used as background data source for future planning and research work in the field of environmental safety and public health***.

Keywords: Gamma spectrometry, natural radionuclides, radiation hazard, Building materials

1. Introduction

The phenomenon of radiation and the consequences associated with radioactivity have been known for over a century. Collaborative international investigations conducted throughout this timeframe have produced a substantial corpus of knowledge regarding radiation and its biological implications; consequently, this scholarly pursuit has facilitated the identification of numerous applications in the fields of medicine and industry, encompassing cancer treatment and medical diagnostic applications [1]. Radiation is the process of releasing or transmitting energy in the form of waves or particles that travel through a vacuum or a medium. It has the ability to penetrate a variety of materials and is generally categorized as either ionizing or non-ionizing depending on the energy level of the atomic particle [2]. Individuals are exposed to radiation in their daily lives. Natural radiation originates from cosmic sources, subsurface materials, and various objects that harbor radioactive elements. Additionally, humans are subjected to different types of nuclear radiation emanating from manufacturing and medical practices which utilize radioisotope substances alongside artificially produced radioisotope substances [3]. Natural radiation sources represent the primary source of radiation exposure for the global population. The spatial distribution of parent rocks and the geological processes that facilitate their erosion and migration from terrestrial environments significantly influence the prevalence of natural terrestrial radionuclides within ecological systems [4].

The assessment of radiological hazards associated with construction substances is vital because the potential exposure to radiation is harmful to individuals. The activity levels of natural happing radionuclides exist in the environment, like U-238, Th-232, and K-40, may be elevated as a result of human and industrial activities [5]. Studying the radiation levels present in the environment is vital for the evaluation of radiological exposure to the public. Thus, this investigation aims to quantify the levels of terrestrial radionuclides for the purpose of estimating radiological hazard indices in rock materials utilized for construction. This investigation provides a baseline reference for assessing natural radiation exposure in the surrounding environment. Accordingly, we focused on the determination of concentration of primordial isotopes (radium-226, thorium-232, and potasium-40) as well as various radiological risk parameters such as Ra_{eq}, D_R, H_{in}, H_{ex}, AEDE indoors and outdoors, AGDE, and ELCR in the investigated building materials. The quantitative assessment of these health hazards, based on data concerning exposure and dose-response correlations, is vital for informing policy decisions regarding acceptable risk levels and optimal management strategies [6, 7].

2. Experimental details

The analysis samples of this study consist of three construction materials, which were selected from the Al-Mahra region, eastern Yemen. Tests show that building material samples are classified as ignimbrite rock, ceramic, and black cement. Ignimbrite rock and ceramic samples were washed using distilled water and dried carefully to ensure the sample was empty of humidity. The samples were crushed to a level of homogeneity of 36 µm in particle radius. Powder of samples is converted to a plastic container and then stored at room temperature for a period of one month to ensure equilibrium between Ra-226 and Th-232 and their disintegration products. A gamma spectrometer system equipped with a high-resolution semiconductor detector (HPGe) was employed to identify levels of natural radionuclides in the building materials. The background or activity measurement period was 64800 seconds. The γ-peaks of 186.3 keV of Radium (Ra-226), 351.9 and 296.20 keV of Lead (Pb-214), and 609.3 and 1120 keV of Bismuth (Bi-214) were employed to measure the concentration of radium-226. Similarly, the γ-peaks of 911 and 338.4 keV of Actinium (Ac-228), 239 keV of Lead (Pb-212), and 727.3 keV of Bismuth (Bi-212) were employed to measure the radioactivity level of thorium-232. The specific level of potassium-40 was found using its own single γ-peak, which was in the gamma energy of 1460 keV. The radioactivity levels for investigated isotopes (Ra-226, Pb-214, Bi-214, Ac-228, Pb-212, Bi-212, and K-40) were expressed in terms of Bq/kg [8].

3. Theoretical Background

3.1 Computation of radiation hazard parameters

Radium equivalent (Raeq) represents a common popular radiological risk indicator. It depends on the concept that activities of 370 Bq kg^{-1} of radium-226, 259 Bq kg^{-1} of thorium-232, and 4810 Bq kg^{-1} of potassium-40 generate uniform γ radiation dose rates. The activity is a weighted total of the concentrations of Ra-226, Th-232, and K-40. It is possible to calculate Raeq by the following relationship [9]:

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

In the above equation, A_{226Ra} represents the radium-226 activity concentration, A_{232Th} is the concentration of thorium-232, and A_{40K} is the concentration of potassium-40. All of them are expressed in Bq/kg. For a regular distribution of primary terrestrial radionuclides, the gamma absorbed dose (D) resulting from γ rays in air one meter above the earth's surface was computed using the UNSCEAR 2000 guidelines. We considered the contribution of other natural radionuclides to be ignorable. Thus, D (nGy y^{-1}) was computed by the following relation [10]:

$$D_R = 0.462ARa + 0.621ATh + 0.0417AK \quad (2)$$

External exposure (Hex) assesses the potential risk from external radiation related to rocks used in construction and is derived from the γ radiation dose measured one meter away from the rock's surface. The H_{ex} was determined, ensuring that the radiation risk remains minimal, and this index remains below unity. The external hazard can be determined by the relation as follows:

$$H_{ex} = A_{Ra}/370 + A_{Th}/259 + A_K/4810 \quad (3)$$

Similarly, another exposure index called the internal index (Hin), which estimates the internal exposure to Rn-222 and their daughter products, is calculated as follows [11]:

$$H_{in} = A_{Ra}/185 + A_{Th}/259 + A_K/4810 \quad (4)$$

The radiation exposure parameters, specifically external and internal indicators, should not be above one in order to be accepted [11]. The annual effective dose equivalent both indoors and outdoors is determined by equations 5 and 6, which are expressed in mSv y^{-1}. The outdoor and indoor task factors of 0.2 and 0.8 are taken into account when calculating AEDE as follows:

$$AEDE_{out} = \text{Absorbed dose} \times 8760\ h\ y^{-1} \times 0.2 \times 0.7 \times 10^{-6}\ Sv\ Gy^{-1} \quad (5)$$

$$AEDE_{in} = \text{Absorbed dose} \times 8760\ h\ y^{-1} \times 0.8 \times 0.7 \times 10^{-6}\ Sv\ Gy^{-1} \quad (6)$$

From the above formula, the value of 0.7 × 10^{-6} Sievert per year refers to the transformation agent between gamma absorbed dose and annual dose, and 8760 hour/year represents the period spent in the houses each year [10]. Excess lifetime cancer risk (ELCR) was computed by:

$$ELCR = AEDE \times DL \times RF \quad (7)$$

In equation 7, AEDE represents the effective dose; DL refers to the long life, approximately seventy years; and RF represents the factor of risk. ICRP 60 employs public values of 0.05 for stochastic effects. Annual gonadal dose equivalent (AGDE) offers an approximation of the amount of radiation the reproductive organs could be exposed to, which is figured out using the energy absorption rate of the gonads in conjunction with the γ radiation dose rate. The formula displayed below is employed to determine AGDE in terms of mSv/y [12]:

$$AGDE = (3.09ARa + 4.18ATh + 0.314AK) \times 10^{-3} \quad (8)$$

3.2 Results and Discussion

Activity concentrations in building material samples:

Table 1 shows the specific levels for radium-226, thorium-232, and potassium-40 determined from the obtained samples. It is noted that ignimbrite rock and ceramics have higher quantities of radium-226, thorium-232, and potassium-40 than the global mean [13], whereas black cement has lower amounts of these radionuclides compared to the worldwide mean.

The mean activity of radium-226, thorium-232, and potassium-40 for the ignimbrite sample was 201.2, 144.8, and 1459 Bq/kg, which were above the global mean values. The average concentrations of radium-226, thorium-232, and potassium-40 in the ceramic sample are 98.13, 48.15, and 610.5 Bq/kg, which indicates exceeding the global average value. The average concentrations of radium-226, thorium-232, and potassium-40 in the black cement sample were 30.77, 17.27, and 273.6 Bq/kg; consequently, these findings fall within the global average. Mean radioactivity levels in samples of construction materials are displayed in Figure 1.

Radiation risk variables in building materials:

Table 2 lists the estimated radiological risk parameters of building material samples under investigation, including (Ra_{eq}), (D_R), (AEDE indoor and outdoor), (H_{ex}), (H_{in}), (ELCR), and (AGDE).

Table 2 shows that the value of Ra_{eq} in the ignimbrite sample was 512.64 Bq kg^{-1}, which was higher than the permissible value of 370 Bq kg^{-1} advised by OECD [14]. The ceramic sample and black cement sample were 215 Bq kg^{-1} and 76.53 Bq kg^{-1}. These numbers were below the limit threshold of 370 Bq kg^{-1} defined by the OECD [14]. The findings of DR for ignimbrite and ceramic were 240.87 and 99.68 nGy/h. These results were elevated above the global mean of 58 nGy h^{-1} recommended by the UNSCEAR [13]. In contrast, the black cement sample had a value of 35.996 nGy/h, which was less than the reported value [13]. AEDE has been estimated, and the results show that for the ignimbrite sample it was 0.297 mSv y^{-1} outdoors and 1.182 mSv y^{-1} indoors, which indicates that it is above the recommended permissible value for the general public (UNSCEAR, 2000) [13]. Whereas in the ceramic sample and the black cement sample, the annual effective dose indoors was 0.408 and 0.177 mSv/y. Outdoors, 0.122 and 0.044 mSv y^{-1}, consequently, which were under the recommended limit value [13].

For ignimbrite samples, the external hazard index was 1.41, while the internal hazard index was 1.95. The average of H_{ex} and H_{in} is greater than one. This indicates that the use of ignimbrite rock in the building of homes is deemed a threat and there is no advice for construction purposes. The external hazard index for the ceramic sample is 0.578, while the internal hazard value is 0.843. The value of H_{ex} for the black cement sample is 0.207, while the H_{in} value is 0.29. It is observed that both H_{in} and H_{ex} have their average below unity for ceramic and black cement samples. This indicates that the use of ceramic and black cement samples in the construction of homes is deemed to pose no risk and thus may be employed for building homes safely. This demonstrates that black cement samples are safe to use as building materials when building homes. For the ignimbrite and ceramic, the estimated ELCR values were 1.04 and 0.429 mSv/y, which fall above the globally accepted limit of 0.290 mSv/y (UNSCEAR-2000). The black cement sample shows that the ELCR is 0.155 mSv/y, which falls under the global reported limit of 0.290 mSv/y (UNSCEAR-2000). The calculated values of AGDE for ignimbrite and ceramic samples were determined to be 1.68 and 0.696 mSv/y. The values of AGDE were above the worldwide limit of 0.36 mSv/y [15, 16]. The value of AGDE for the black cement sample was 0.253 mSv/y, which was below the critical limit of 0.36 mSv/y [15, 16]. Figure 2 displays the average of estimated radiological risk variables for the examined building material samples.

3.3 Figures and Tables

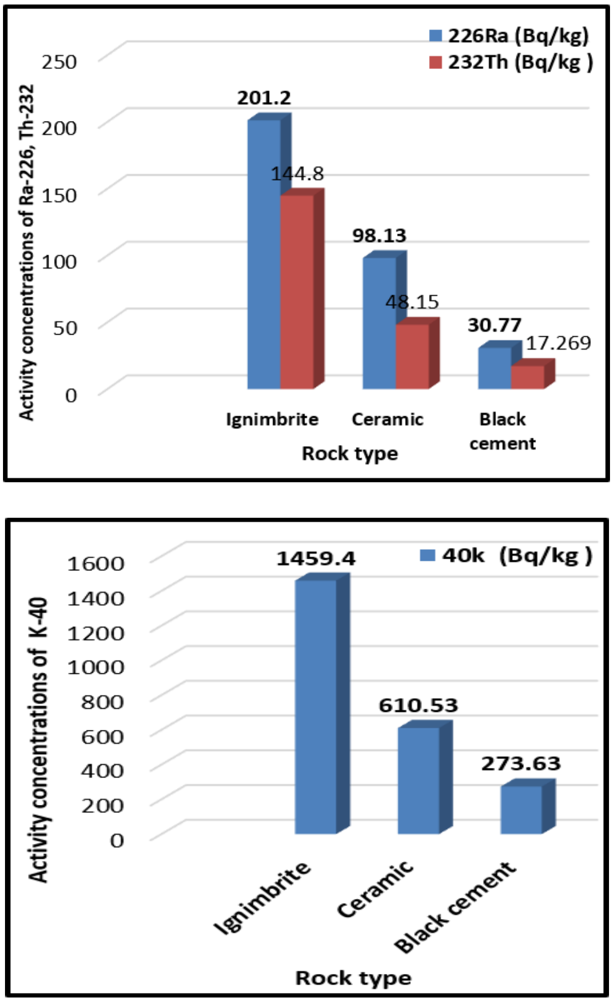
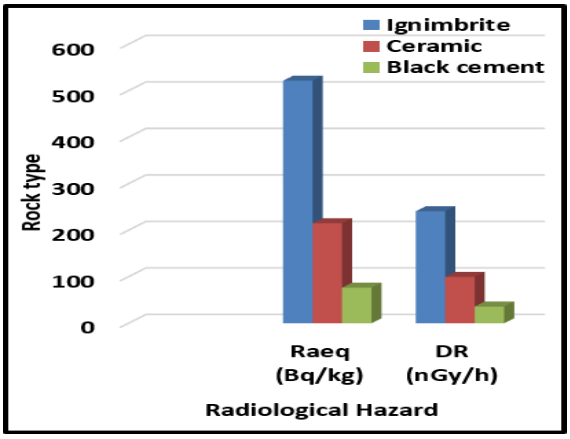


Figure 1: The measured specific activity of primordial radioisotopes in the construction materials.



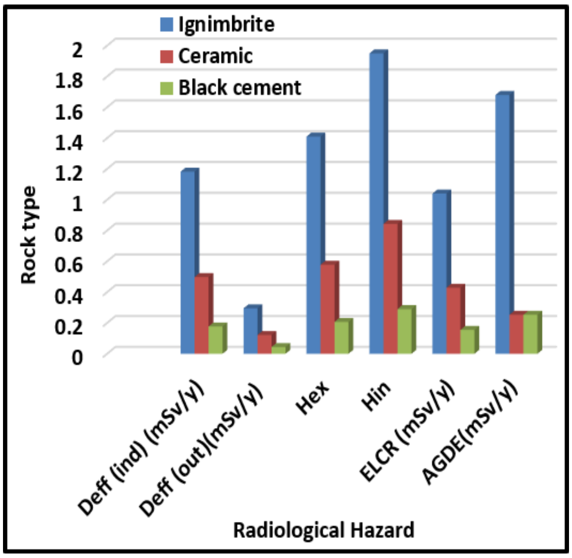


Figure 2: Shows average of Ra_{eq}, D_R, D_{eff} (indoor & outdoor), H_{ex}, H_{in}, ELCR, and AGDE in building materials.

3.4 Tables

Table 1: Measured concentrations of primordial radionuclides (Bq per kg) for construction materials

Rock type	A_{226Ra}		A_{232Th}		A_{40K}	
	Avg.	sd	Avg.	sd	Avg.	sd
Ignimbrite	201.2	16.9	144.8	16.98	1459	91.2
Ceramic	98.13	7.98	48.15	5.1	610.5	28.99
Black cement	30.77	4.36	17.27	1.4	273.6	13.65
World average	35		30		400	

Table 2: Lists the estimated radiological risk parameters of building material samples under investigation, including (Raeq), (DR), (AEDE indoor and outdoor), (Hex), (Hin), (ELCR), and (AGDE)

No	Radiological Hazard	Rock type			Permissible limits
		Ignimbrite	Ceramic	Black cement	
1	Ra_{eq} (Bq/kg)	521.64	215	76.53	370
2	D_R (nGy/h)	240.87	99.68	35.996	57
3	D_{eff} (ind) (mSv/y)	1.182	0.498	0.177	0.46
4	D_{eff} (out)(mSv/y)	0.295	0.122	0.044	0.07
5	H_{ex}	1.41	0.578	0.207	< 1
6	H_{in}	1.95	0.843	0.29	< 1
7	ELCR (mSv/y)	1.04	0.429	0.155	0.29
8	AGDE (mSv/y)	1.68	0.253	0.253	0.36

3.5 Conclusion

Samples of building materials from the Al-Mahara region in Yemen have been analyzed to determine concentrations of terrestrial radionuclides Ra-226, Th-232, and K-40 using a gamma spectrometric technique. The findings show the average radioactivity of radium-226, thorium-232, and potassium-40 are 201.2, 144.8, and 1459 Bq/kg in the ignimbrite sample; 98.13, 48.18, and 610.5 Bq/kg in the ceramic sample; and 30.77, 17.27, and 273.6 Bq/kg in the black cement sample. The background data on levels of terrestrial radioactivity in the study region established in this work will serve as a future reference for additional research. Radiation hazard indices were calculated for evaluating risks to residents. The calculated dose rate in ignimbrite rock was found to be double the global average values, putting nearby residents and users of the rocks at risk for radiation exposure. A black cement sample is advised for building houses with safety for individuals, even though the estimated hazard indices are significantly lower than the critical limit values. According to the estimated hazard indices in the ceramic sample, some radiological hazards are below the reported threshold values, while others exceed the critical limit values. Therefore, the ceramic sample poses a potential health risk to the population and is considered unsafe. Overall, this research work carries significant implications from both health and scientific standpoints. It may serve as a reference for similar studies if the results yield substantial findings. Furthermore, it could facilitate the formulation of policies aimed at regulating the utilization of construction materials that may pose threats to public health.

4. Equations

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

$$D_R = 0.462ARa + 0.621ATh + 0.0417AK \quad (2)$$

$$H_{ex} = A_{Ra}/370 + A_{Th}/259 + A_K/4810 \quad (3)$$

$$H_{in} = A_{Ra}/185 + A_{Th}/259 + A_K/4810 \quad (4)$$

$$AEDE_{out} = D_R \times 8760\ h\ y^{-1} \times 0.2 \times 0.7 \times 10^{-6}\ Sv\ Gy^{-1} \quad (5)$$

$$AEDE_{in} = D_R \times 8760\ h\ y^{-1} \times 0.8 \times 0.7 \times 10^{-6}\ Sv\ Gy^{-1} \quad (6)$$

$$ELCR = AEDE \times DL \times RF \quad (7)$$

$$AGDE = (3.09ARa + 4.18ATh + 0.314AK) \times 10^{-3} \quad (8)$$

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