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Natural radionuclide distribution and analysis of associated radiological concerns in rock samples from a rocky town (Dutsin-Ma) in the North-Western region of Nigeria

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Abstract

Natural radionuclides in rock samples, especially ²²⁶Ra (Radon), ²³²Th (thorium), and ⁴⁰K (potassium), are the primary source of exposure to radiation for employees and the public, and their detection is critical for radiation safety. The gamma (γ) radiation from natural radionuclides was quantified using the γ-ray spectrometry method in twenty rock samples obtained from Dutsin-Ma, a rocky town in Nigeria's north-western region.
The data obtained were used to estimate the radiological parameters and the ex The data obtained were used to estimate the radiological parameters and the excess lifetime cancer risk (ELCR). The results obtained show that the minimum activity concentrations of ⁴⁰K, ²³²Th, and ²²⁶Ra are respectively 36.7 Bq/kg, 25.99 Bq/kg, and 11.59 Bq/kg, with their maximum values being 73.23 Bq/kg, 92.81 Bq/kg and 100.93 Bq/kg respectively. The average activity of 40 K, 232 Th, and 226 Ra in the rock samples were found to be 73.5781± 3.38 Bq/kg, 40.7848± 3.32 Bq/kg, and 33.6616± 3.58 Bq/kg respectively, which were all below the World average values of (33, 45 and 420) Bq/kg respectively. The ELCR was found to be 0.185 ± 0.01 , while the average absorbed dose rate, as well as the mean annual effective dose rate, were calculated to be 43.13± 2.7 nGy/h and 0.015± 0.003 mSv/y respectively. The outcomes for internal hazard, external hazard, annual absorbed and effective dose equivalent (ABEDE), alpha and gamma index values were below the maximum values allowed limits of 1 mSv/y for ABEDE and 1 for representative alpha & gamma index values, internal and external hazards as recommended by the International Commission on Radiological Protection (ICRP). This study shows that the radiation contamination of rock particles in Dutsin-Ma LGA does not pose much significant risk.

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1. Introduction

The term "radioactive contamination of the environment" refers to the spike in background radiation that results from either naturally existing or artificially created radioactive materials often employed by humans [\[1\]](#page-10-0). All living things are exposed to radiation,

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which is a relatively regular occurrence in nature. Natural radioactive materials are present in our environment in higher quantities as a result of human activities like drilling, processing, and burning fossil fuels. These activities also include mining, milling, and processing of uranium ores and mineral sands, smelting of metalliferous ores, manufacturing of fertilizers, and manufacturing of fertilizers [\[2\]](#page-10-1). In places with significant concentrations of naturally occurring radionuclides, these substances may be found in high concentrations in the rocks, soil, groundwater, and surface water [\[3\]](#page-10-2). Numerous elements have resided in the Earth's crust ever since it formed. Human exposure to these elements occurs from both anthropogenic and natural sources, and their quantities in the environment reflect geomorphological factors [\[4\]](#page-10-3). The primary sources of background radiation are uranium, thorium, and the potassium isotope ${}^{40}K$ [\[5\]](#page-10-4). Through consumption and inhaling, naturally, present radionuclides can reach the living organism [\[6\]](#page-10-5). Inhaling radon decay products typically results in the biggest internal natural background dose. As a member of the uranium decay series, radon is created when radium in the soil decays. It can travel into the atmosphere and cause contamination, although typical atmospheric mixing keeps concentrations very low. However, radon may be retained and accumulate to large levels if it diffuses from the soil beneath a confined structure, like a house [\[7\]](#page-10-6).

It is interesting to note that scientists from different fields, including [\[8](#page-10-7)[–10\]](#page-10-8) all agreed that there are several naturally occurring radioactive materials (NORMs) dispersed in our surroundings because they most often occur in different rock formations. Additionally, soil radionuclides have a considerable impact on terrestrial gamma radiation levels and are mostly dependent on the geology of a given area understanding radionuclides $(^{40}K, ^{232}Th,$ and ^{238}U and their progenies) when evaluating the consequences of radiation exposure from natural and man-made sources, hence, distribution levels in the environment are crucial. Additionally, studies showing researchers employing gamma spectroscopy using either HPGe or NaI(Tl) detectors are obvious.

Despite the fact that according to [\[11\]](#page-10-9), the crystalline basement rocks that make up Dutsin-Ma and her region are primarily metamorphic, hence prone to radiation hazards, there is no report on the systematic statistics on concentrations of naturally occurring radionuclides to determine the hazards from rocks in Dutsin-Ma. Therefore, a study on naturally radioactive materials in Dutsin-Ma is required to provide baseline information on natural radioactivity, allowing any kind of contamination to be identified and further quantified. The activity of ⁴⁰K, ²²⁶Ra, and ²³²Th has been measured in this work using a NaI(Tl) detector.

2. Material and Methods

2.1. Geology and location of the study area

The samples were obtained from Dutsin-Ma LGA, which is located between 12◦ 17.00′N and 12◦ 17.84′N and 007◦ 26′E and 007◦ 27′E. It is bordered to the north by the Kurfi and Charanchi LGAs, to the east by the Kankia LGA, to the west by the Safana and Dan-Musa LGAs, and to the southeast by the Matazu LGA. According to the national census of 2006, Dutsin-Ma LGA has a land area of around 552.323 km^2 and a population of about 169829 [\[12\]](#page-10-10).

2.2. Sample Collection and Preparation

Twenty rock samples were collected randomly within selected areas of Dutsin-Ma LGA (Fig. 1) and analyzed by gamma spectrometry to determine the activity concentration of radionuclides. A GPS was used to record the location of all the sampling points as shown in Table [1.](#page-2-0) At the laboratory, each of the rock samples was air-dried and then oven-dried at a temperature of 105°C until all moisture is completely lost. The samples were then grounded into a fine powder using a pulverizer and sieved through a 2-mm pore size mesh into a Marinelli beaker. The Marinelli beakers with the samples were then hermetically sealed and stored for 24 days for the short-lived daughters of ²²⁶Ra (in the ²³⁸U decay series) and ²³²Th decay series to attain secular equilibrium with their long-lived parent radionuclides. The weight of the samples was recorded as shown in Table [1.](#page-2-0)

2.3. Sample Analysis

The sample analysis was performed at the Center for Energy Research and Training (CERT), Ahmadu Bello University, Zaria, Nigeria. Low-level gamma counting spectrometer with a sodium iodide NaI(TI) detector housed in a 6 cm thick lead shield, cadmium lined assembly, and copper sheets for background radiation reduction was used for the measurements. It is connected to the computer-based multichannel analyzer (MCA) via a preamplifier base and counted for 29,000 seconds.

The quantitative determination of 40 K, 226 Ra, and 232 Th of the rock samples was accomplished following the use of IAEA standard reference materials for calibration respectively. After evaluating the spectral lines of various emissions using the MAESTRO software, the activity concentrations of the natural radionuclides were estimated. The system was configured with an all-energy resolution of 7.2% and a working energy range of 0–3000 keV. Using the formula in equation [\(1\)](#page-1-0), the activity concentration of gamma-emitting radionuclides in the samples was determined:

$$
E_e = \frac{N(cps)}{I_{\gamma}A(Bq)},\tag{1}
$$

where E_e is the detector's efficiency at a given energy, N is the peak's net area (count per second), I_v is gamma intensity and A is the standard's activity. For the NORMs, the efficiency curve was obtained at various energies. This curve was used to calculate the detector's efficiency. The outcome is computed by a computer and displayed in tables.

Figure 1. Map of the study area showing sampling sites.

Table 1. Or S Coordinates of the fock samples.								
Sample ID	Latitude	Longitude	Sample ID	Latitude	Longitude			
Rock 1	12°26'35.N	$007^{\circ}38'$ E	Rock 11	$12^{\circ}28'42.N$	$007^{\circ}27'$ E			
Rock 2	$12^{\circ}27'12.N$	$007^{\circ}64'$ E	Rock 12	$12^{\circ}26'72.N$	$007^{\circ}83'$ E			
Rock 3	12°26'72.N	$007^{\circ}58'$ E	Rock 13	$12^{\circ}27'67$.N	$007^{\circ}58'$ E			
Rock 4	12°26'55.N	$007^{\circ}47'$ E	Rock 14	12°27'75.N	$007°96'$ E			
Rock 5	$12^{\circ}27'33.N$	$007^{\circ}57'$ E	Rock 15	$12^{\circ}26'66$. N	$007^{\circ}29'$ E			
Rock 6	$12^{\circ}26'81$.N	$007^{\circ}25'$ E	Rock 16	$12^{\circ}27'59$.N	$007^{\circ}61'$ E			
Rock 7	$12^{\circ}27'56.N$	$007^{\circ}69'$ E	Rock 17	$12^{\circ}26'83.N$	$007^{\circ}36'$ E			
Rock 8	12°27'25.N	$007^{\circ}30'$ E	Rock 18	$12^{\circ}27'27.N$	$007^{\circ}75'$ E			
Rock 9	$12^{\circ}26'21.N$	$007^{\circ}48'$ E	Rock 19	$12^{\circ}27'51.N$	$007^{\circ}29'$ E			
Rock 10	12°27'56.N	$007^{\circ}30'$ E	Rock 20	$12^{\circ}26'77$.N	$007^{\circ}48'$ E			

Table 1. GPS Coordinates of the rock samples.

2.4. NORM-related radiation risks in the rock samples

*2.4.1. Annual e*ff*ective doses (E)*

Equation [\(2\)](#page-2-1) was used to calculate an individual's annual effective dosage

$$
\text{AEDE}(mSv/y) = D \times T \times f_c \times 10^{-3} \times 8760,\tag{2}
$$

where *D* is the rate of absorbed dosage, *T* is the 20% outside occupancy period, and *fc* is the conversion factor of 0.7, all of which are in SvG/y.

2.4.2. Absorbed dose rate (D)

The activity concentrations of radium-226, thorium-232, and potassium-40 were converted into dose rates using the conversion coefficients 0.462, 0.604, and 0.0417 for radium, thorium, and potassium, respectively. These factors were used to calculate the total absorbed dose rate (D) (nGy/h) using the equation [\[4\]](#page-10-3):

$$
D(nGy/h) = 0.462K_{\text{Ra}} + 0.604A_{\text{Th}} + 0.0417A_K. \tag{3}
$$

Table 2. Weight of samples in grams (g).

2.4.3. Radium equivalent activity

The radiation hazard index Raeq was used to evaluate the gamma-ray radiation risks associated with the radionuclides 226 Ra, ²³²Th, and ⁴⁰K. Based on the assumption that the gamma-ray dose rates produced by 370 Bq/kg of ²²⁶Ra, 259 Bq/kg of ²³²Th, and 4810 Bq/kg of ⁴⁰K are equal, the Radium equivalent activity is the weighted sum of the activities of the three radionuclides [\[13,](#page-10-11) [14\]](#page-10-12):

$$
Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_k.
$$
\n(4)

2.4.4. Hazard indices

In this study, four indices will be generated to evaluate the risks from gamma radiation caused by specific radionuclides of ²²⁶Ra, 232 Th, and 40 K.

2.4.5. Representative level index

Equation [\(5\)](#page-3-0) was used to generate the hazard index of the representative level index I_y , which is used to evaluate the gamma radiation hazards related to environmental radioactivity [\[14\]](#page-10-12):

$$
I_{\gamma} = \left(\frac{A_{\text{Ra}}}{150} + \frac{A_{\text{Th}}}{100} + \frac{A_k}{1500}\right). \tag{5}
$$

2.4.6. Alpha index

The alpha index, defined as follows, was adopted in the measurement of the extra alpha radiation brought on by exposure to radon from used soils in construction [\[15\]](#page-10-13):

$$
I_{\alpha} = \frac{A_{\text{Ra}}}{200}.\tag{6}
$$

2.4.7. External Hazard Index

Two indices that describe exterior and internal radiation dangers were defined by Beretka and Mathew [\[13\]](#page-10-11). These indices provide a dose equivalent upper limit of 1 mSv/y for the radiation dosage. The following equation is used to determine the external hazard index (Hex):

$$
H_{\rm ex} = \left(\frac{A_{\rm Ra}}{370} + \frac{A_{\rm Th}}{259} + \frac{A_k}{4810}\right). \tag{7}
$$

2.4.8. Internal Hazard Index

Internal exposure to carcinogenic radon and its transient offspring is indicated by the internal hazard index (H_{in}) [\[16\]](#page-10-14). Equation [\(8\)](#page-4-0) was used to determine the maximum allowed concentration for ^{226}Ra , which must be decreased to 185 Bq/kg, which is half of the typical limit, to account for this concern [\[17,](#page-10-15) [18\]](#page-10-16):

$$
H_{\rm in} = \left(\frac{A_{\rm Ra}}{185} + \frac{A_{\rm Th}}{259} + \frac{A_k}{4810}\right),\tag{8}
$$

where the activity concentrations of ²²⁶Ra, ²³²Th, and ⁴⁰K are represented by A_{Ra} , A_{Th} and A_K , respectively.

2.4.9. Excess Lifetime Cancer Risk (ELCR)

The Excess Lifetime Cancer Risk (ELCR) is an incremental rise in cancer cases in the exposed population over what would happen in the absence of exposure. It is the likelihood of developing cancer as a result of exposure to a given carcinogen. ELCR is provided by:

$$
ELCR = AEDE \times DL \times RF,
$$
\n(9)

where, AEDE = Annual Effective Dose Equivalent, *DL* = The average duration of life/ life expectancy (estimated as 70 years), and RF = Risk Factor (0.05 S/v), i.e. fatal cancer risk per Sievert [\[19\]](#page-10-17).

3. Results and Discussion

The activity concentration of Radium-226, Thorium-232, and Potassium-40 in the rock samples have been measured and com-pared to world mean as shown in Figure [2.](#page-4-1) The minimum activity concentration of 226 Ra, 232 Th, and 40 K are 11.59 Bq/kg, 25.99 Bq/kg, and 36.7 Bq/kg, and the maximum values are 73.23 Bq/kg, 92.81 Bq/kg, and 100.93 Bq/kg respectively. The average activity concentrations of ²²⁶Ra, ^{2[3](#page-7-0)2}Th, and ⁴⁰K are 33.66 Bq/kg, 40.78 Bq/kg, and 73.59 Bq/kg respectively. Table 3 shows the mean activity concentrations of the three natural radionuclides ²²⁶Ra, ²³²Th, and ⁴⁰K in all the sampling sites.

Figure 2. Comparison of mean activities in rock samples of Dutsin-Ma to worldwide mean.

According to the activity concentration of each naturally occurring radionuclide, the amount of natural radioactivity varied from 11.58 to 73.23 Bq/kg for K-40, 61.29 to 100.79 Bq/kg for Ra-226, and 26.45 to 92.81 Bq/kg for Th-234. In comparison to the global measured mean values of 33 Bq/kg for Radon-226, 45 Bq/kg for Thorium-232, and 420 Bq/kg for Potassium-40, the activity concentration values of ^{226}Ra , ^{232}Th , and ^{40}K in Dutsin-Ma LGA are lower [\[4\]](#page-10-3). These results show that the activity levels were Ra-226 > Th-232 > K-40. The three radionuclide concentrations in Dutsin-Ma Town are apparently low in radiation and favorably compared other elsewhere.

Figure 3. Radium equivalent activities for rock samples analyzed in this work.

Figure 4. A graph of absorbed dose rate.

Basement complex rocks with a predominance of metamorphic genesis make up the majority of the rocks in the Dutsin-Ma region. The igneous forms of rocks are the oldest in this area, and heat and pressure eventually changed them into metamorphic rocks. Because of the impacts of weathering and erosion, the rocks in some areas of the area have altered through time. This

Figure 5. A graph of annual effective dose equivalent.

Figure 6. Comparison between internal (H_{in}) and external (H_{ex}) hazard indices.

provides the basis for lateritic capping, river valley formation, and the development of undulating plains [\[11,](#page-10-9) [20\]](#page-10-18). As a result of the rocks, granite hills and inselbergs like the one in the middle of Dutsin-Ma town later developed [\[11\]](#page-10-9). It was concluded that the crystalline basement rocks that make up the Dutsin-Ma Town and her region are primarily metamorphic [\[11\]](#page-10-9). Natural radionuclides can be found in many minerals, but in igneous and metamorphic rocks, they are mostly found in zirconite, apatite, and titanite as well as in rare minerals such monazite, pyrochlore, uraninite, thorite, and allanite [\[21\]](#page-10-19). Though some authors also reported that metamorphic rocks is majorly made up of banded gnesiss, granitic gnesiss, biotite gnesiss and quartzite augitegnesiss [\[20,](#page-10-18) [22\]](#page-11-0). Metamorphic rocks are reported to hold a large variant of radioactive potassium concentration, this depends majorly on the original nature of the parent sedimentary or igneous precursor rock. One of the major example, the politic rocks which are metamorphic are formed by the metamorphism of the mud-rocks that contains mica and consequently a significant potassium but opposite to this,

Figure 7. Comparison between gamma (I_{γ}) and alpha (I_{α}) indices.

quartzites which are ored by the metamorphism of a clean quartz-rich sandstone (which are mostly found among the rocks in Dutsin-Ma Town and its region) contains little or no radioactive potassium [\[23,](#page-11-1) [24\]](#page-11-2). Basham and Kemp [\[25\]](#page-11-3) propose that concentration of radium found in most metamorphic rocks ranges from 0.1 to 4 ppm Ra and that diminution of uranium mostly takes palce with rising metamorphic gradient. They further concluded that the variation in radium content of this rock types are usually dependent on the parent materials. Thorium concentration in metamorphic rocks is vastly unpresictable hence variable, this depend on the original parent sedimentary or igneous rocks. It has been shown that its concentration may vary from <0.1 ppm in marble to >67 ppm to some high-grade feldpathic and quartzite augitegnesiss rocks [\[26\]](#page-11-4). The low level of thorium content was associated with the selective

Rock 20 65.8165 3.7325 19.6987 4.7509 31.0148 2.9190 69.1179

Figure 8. A: Mean activities in rock samples of Dutsin-Ma to Worldwide mean B: Radium equivalent activities for rock samples C: Absorbed dose rate D: Annual effective dose equivalent E: Comparison between internal (Hin) and external (Hex) hazard indices F: Comparison between gamma (I_{γ}) and alpha (I_{α}) indices.

loss of this radioactive element during metamorphism [\[23,](#page-11-1) [27\]](#page-11-5). Our results compliment studies from various authors, about the low activity concentration of Ra-226, Th-232, K-40 in the metamorphic rock-types. Metamorhic rocks have been generally reputed to have varied activity concentration depending on their parental sources [\[8,](#page-10-7) [21,](#page-10-19) [28,](#page-11-6) [29\]](#page-11-7).

3.1. Radiological health risk assessment

The necessity to evaluate the radiological threat posed by the naturally occurring radionuclides in the rocks of the research region results from the fact that the rocks under study are used for a variety of purposes, mostly as ornaments and building materials. With Table 4. Radiological Hazards of analysed samples.

Sample	Absorbed dose	Annual	Alpha	Gamma	Hazard indices		Excess-life
ID	rate (nGy/h)	effec-	Index	Index			cancer risk
		dose tive					
		(mSv/y)					
Rocks	D	E	I_{α}	I_{γ}	H_{in}	$H_{\rm ex}$	ELCR
1	34.1035	0.0418	0.14079	0.2676	0.2827	0.2065	0.1464
\overline{c}	37.6069	0.0461	0.13036	0.2976	0.2997	0.2291	0.1614
3	30.4663	0.0374	0.0985	0.2427	0.2399	0.1867	0.1308
4	41.0602	0.0504	0.0892	0.3313	0.3029	0.2546	0.1764
5	36.0779	0.0442	0.1060	0.5768	0.2797	0.2223	0.1547
6	37.9894	0.0466	0.1976	0.5906	0.3349	0.2281	0.1631
7	38.1431	0.0468	0.1860	0.5951	0.3332	0.2327	0.1637
8	49.2596	0.0604	0.1663	0.7831	0.3930	0.3032	0.2114
9	50.4425	0.0619	0.2503	0.7864	0.4404	0.3050	0.2165
10	57.0909	0.0700	0.2323	0.8982	0.4751	0.3495	0.2451
11	24.5228	0.0298	0.0579	0.3757	0.1802	0.1500	0.1041
12	56.4134	0.0692	0.3198	0.7649	0.5139	0.3027	0.2422
13	41.3643	0.0507	0.2225	0.5673	0.3704	0.2501	0.1776
14	60.0265	0.0736	0.3662	0.7669	0.5592	0.3613	0.2576
15	41.5531	0.0510	0.1304	0.6629	0.3259	0.2554	0.1784
16	41.3573	0.0507	0.1918	0.6501	0.3543	0.2501	0.1775
17	38.9626	0.0478	0.1188	0.6221	0.3038	0.2396	0.1672
18	75.3894	0.0925	0.1738	1.2144	0.5633	0.4693	0.3236
19	40.7552	0.0500	0.0892	0.6328	0.2599	0.2529	0.1750
20	30.4664	0.0374	0.0985	0.4525	0.2399	0.1867	0.1308
Average	43.1274	0.0147	0.6039	0.6039	0.3526	0.2618	0.1852
Error	± 2.69	± 0.003	± 0.24	± 0.053	± 0.024	± 0.016	± 0.011

Table 5. Concentration of the radionuclides of Dutsin-Ma compared to worldwide mean.

a mean value of 0.0529 mSv/y for the rock samples, the yearly effective dosage was outside and ranged from 0.09 mSv/y to 0.03 mSv/y, with Sample 18 having the highest value and Sample 11 having the lowest. As can be seen in Figure [4,](#page-5-0) the average absorbed dose rate in the research area is less than the global average value of 1 mSv/y [\[4,](#page-10-3) [30\]](#page-11-8). The annual absorbed dose was in ranged from 24.[5](#page-6-0)2 nGy/h to 75.39 nGy/h, with Sample 10 having the highest value while Sample 3 having the least. Figure 5 shows the graph of the annual effective dose rate. The average outdoor dose rate is 43.13 nGy/h which is lower than the world average of 60 nGy/h [\[4,](#page-10-3) [30\]](#page-11-8). Radium equivalent activity (Raeq) estimated for the collected samples ranged from 55.56 Bq/kg to 173.78 Bq/kg with a mean value of 97.7 Bq/kg.

The estimated average levels fell short of the 370 Bq/kg suggested maximum value. The radium equivalent activity for each sampling site is shown in Figure [3.](#page-5-1) According to Figure [6,](#page-6-1) the typical internal and exterior danger indices for the examined rock samples are 0.3526 and 0.2618, respectively. These numbers are below 1, indicating that the local radiation exposure is very low. It is advised [\[30\]](#page-11-8) that building materials be H_{ex} and H_{in} less than 1. The values of the gamma index ranged from 0.3757 to 1.2144 with a mean value of activity index of 0.6039, while the values of the mean estimated alpha index ranged from 0.057 to 0.3198 with a mean value of 0.1682. These values are below unity, hence the rock does not seriously affect the local population in terms of radiation effects. Figure [7](#page-7-1) demonstrates that for all rock samples, gamma indices have larger values than alpha indices. The maximum allowable limit suggested by ICRP is lower for each of the radiation danger indices calculated for the rock samples. As a result, the natural radionuclides from the rock and their particles in the research region do not represent any significant health hazards to the people in the vicinity.

Though this current study, gamma indices are higher than doses reported for other foreign countries, according to a comparison of the data with those estimations (Figure [8\)](#page-8-0) for Finland (0.49 mSv/y), United States (0.28 mSv/y), Japan (0.32 mSv/y), China (0.55 mSv/y), Bulgaria (0.45 mSv/y), Canada (0.23 mSv/y), Germany (0.41 mSv/y), United Kingdom (0.35 mSv/y), Norway (0.49 mSv/y),

4. Conclusion

The radioactivity levels 226 Ra, 232 Th, and 40 K have been measured in the rock samples collected from Dutsin-Ma LGA. The activity concentration of these radionuclides was found to be in the range of $(11.59 - 73.23)$ Bq/kg, $(25.99 - 92.82)$ Bq/kg, and (36.77) - 100.93) Bq/kg respectively. Annual effective dose (mSv/y), annual dose rate (nGy/h), Alpha and Gamma radiation index (I_a and I_y), radium equivalent activity (Ra_{eq}), and internal (H_{in}) and external (H_{ex}) indices were used as variable hazard measures. The outcome of these measures is within the permitted standard limits for the world. Because of this, the public is not at risk from the detrimental radiation effects that result from the researched rocks' inherent radioactivity in the context of building and decorating or from the amount to which they affect persons who lived within the vicinity or around these places where the researched rocks are or for other applications. It is our strong belief that baseline information on the level of natural radioactivity in this rocky Town has been provided by this study.

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