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Assessment of the natural radioactivity content in typical building materials employed in the Italian cultural heritage.

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***Abstract* – Natural radionuclides from the uranium (^{238}U) and thorium (^{232}Th) families, as well as the radioactive isotope potassium, are present in varying concentrations in building materials and other construction products obtained from rock and soil. In Italy, in historical monuments, natural building materials have been largely employed for decorative and structural purposes.**

In this study the natural radioactivity content in six different materials (grey tuff, basalt lava stone, lava stone, sandstone, magmatic rock and travertine) was measured. The activity concentrations were determined by high purity germanium (HPGe) detector-based gamma spectrometry.

The radium equivalent activity (R_{aeq}), indoor absorbed gamma dose rate (D_{in}), the annual effective dose (D_e), and the internal and external hazard indexes were evaluated to assess the radiation hazard according to the international recommendations.

I. INTRODUCTION

Naturally occurring radionuclides of the uranium (^{238}U) and thorium (^{232}Th) series, as well as the radioactive isotope potassium (^{40}K), are present everywhere in varying concentrations in the environment and building materials [1].

Radon-222, with a half-life of 3.8 days, is one of the important members of the uranium (^{238}U) radioactive decay chain which are the primary sources of indoor radon found in soil, rocks, drinking water, building materials and natural gas. The natural origin of radon comes from rock salts or sediment [2]. Radon gas is one of the most effective sources of natural radiation exposure indoors and it is listed by the International Agency for Research on Cancer as the second leading cause of lung cancer after smoking and by the World Health Organisation (WHO) as a major environmental carcinogen [3].

All building materials, especially those derived from rock and soil, contain natural radioactive nuclides in various quantities, mainly ^{238}U and ^{232}Th series radionuclides and the ^{40}K radioactive isotope [4]. Radiation exposure from natural radionuclide in building and construction materials is generally unavoidable. The radionuclide content of such materials and the resulting doses can varies widely depending on the geological structure of the natural materials used and the industrial processes used in the production of building materials [5]. In this work, a total of 6 different building materials, typically used for decorative or structural use in the Italian historical buildings, were analysed using gamma-ray spectrometry to determine the activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K. The potential radiological hazards of these materials were determined by calculating the indoor absorbed gamma dose rate (D_{in}), the annual effective dose rate (D_e) and the activity utilization index.

The obtained results were compared to the recommended values to determine the radiation hazard to humans and the potential risk.

II. MATERIALS AND METHODS

Six samples of about 5cmx5cmx3cm of different building materials from quarries of several Italian regions were selected among the most famous and used ones, in the national cultural heritage, for structural and decorative use (Figure 1 and Table 1).

The samples were transferred to the laboratory (Nuclear Physics Laboratory, Central Research Laboratory of Kırklareli University, Turkey) where the measurements of the natural radioactive content were performed.

According to the protocol measurement, firstly, the samples were dried in an oven at approximately 105 °C for 24 hours till to constant weight. After that, they were grinded under 0.63mm. The samples were transferred into a leak-proof beaker, and stored for more than 30 days, to achieve the secular equilibrium between ^{226}Ra and its progeny (Figure 2).

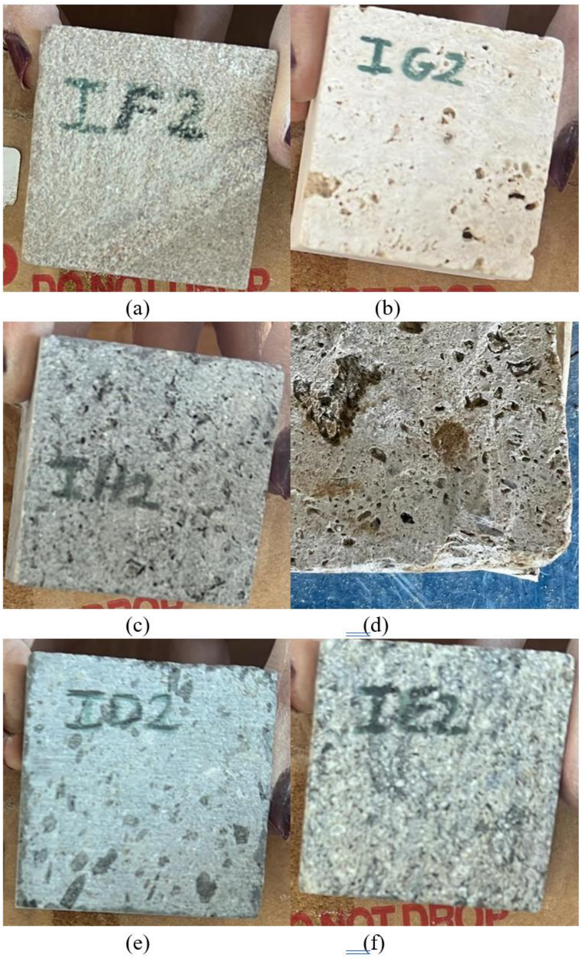

Fig. 1. Building materials samples: (a)sand stone santafiora, (b)travertine, (c)lavastone, (d)grey tuff, (e) basalt lavastone, (f) magmatic rock peperino.

Table 1. Building materials samples data.

Name	code	origin
grey tuff	IT13	CAMPANIA
basalt lava stone	IT47	LAZIO
magmatic rock-peperino	IT 48	LAZIO
sandstone-santa fiora	IT 49	TUSCANY
travertine	IT50	CAMPANIA
lava stone	IT51	SICILY

The radionuclides (^{226}Ra, ^{232}Th, ^{40}K) activity concentrations in the samples were determined using a high purity germanium (HPGe) semiconductor detector ORTEC (model GEM70P4), 70% relative efficiency and 1.9 keV resolution (FWHM) for the 1332.5 keV peak of 60Co and 1 keV resolution at 122 keV. The detector is shielded to minimize natural background radiation from the environment. The energy and efficiency calibration of the HPGe gamma spectrometer is performed with a source containing multi-nuclides distributed in 1.0 gcm^{-3} epoxy (Eckert & Ziegler Isotope Products), in the same geometry with samples, energies ranging from 80 to 2500 keV.

Fig. 2. Building materials preparation.

III. RESULT AND DISCUSSION

The specific activities of natural radionuclide in samples range from below the minimum detectable activity (MDA) to 126.01 Bqkg^{-1} for ^{226}Ra, from MDA to 260.08 Bqkg^{-1} for 232Th and from 16.94 to 2164 Bqkg^{-1} for 40K. Activity concentration results of ^{226}Ra, ^{232}Th, and ^{40}K are reported in Table 2 and in Fig. 3 and Fig.4.

Table 2. Activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K (Bq kg^{-1})

code	Ra-226	Th-232	K-40
IT13	85.28±10.95	122.43±11.71	1753.3±152.3
IT47	53.29±4.64	41.71±2.98	554.39±24.88
IT 48	126.01±7.29	182.66±9.37	1541.8±83.30
IT 49	10.21±1.10	23.90±3.14	963.94±83.94
IT50	<1.3054	<1.1469	16.94±5.75
IT51	88.47±6.12	260.08±14.01	2164.5±87.31

The highest values for the activity concentration of ^{226}Ra is 126 Bqkg^{-1}, measured in the magmatic rock. For ^{232}Th and ^{40}K, the highest values are respectively 260 Bqkg^{-1} and 2165 Bqkg^{-1}, measured in the sample of lava stone.

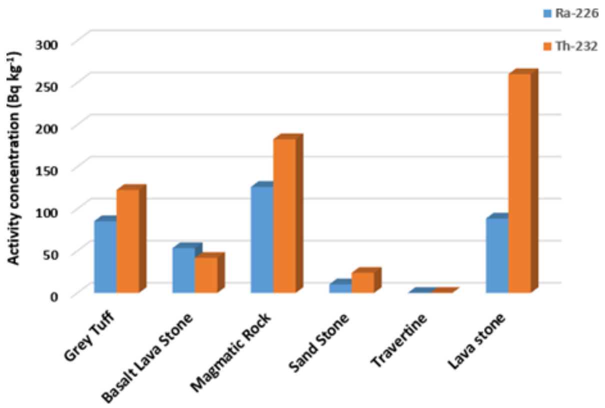

Fig. 3. Activity concentrations of ^{226}Ra and ^{232}Th.

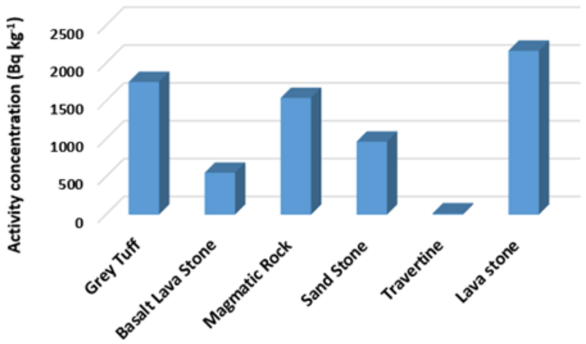

Fig. 4. Activity concentrations of ^{40}K

The assessment the radiation hazard for people living and working in buildings where these materials are potentially used is calculated by using the following Equation (1):

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

where A_{Ra}, A_{Th} and A_K are the specific activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K, respectively.
The calculated values of radium equivalent activity (Ra_{eq}) for analyzed building materials were in the range from 1.30 $Bqkg^{-1}$ (travertine sample) to 627.05 $Bqkg^{-1}$ (lava stone sample).

Table 3. Ra_{eq} ($Bqkg^{-1}$) of the investigated samples

code	Ra_{eq}
IT13	395.36
IT47	155.62
IT 48	505.93
IT 49	118.61
IT50	1.30
IT51	627.05

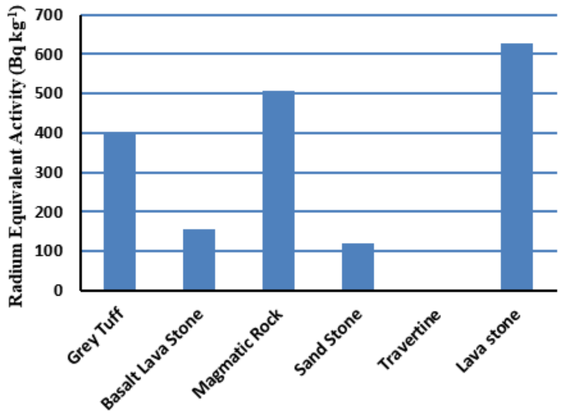

Fig. 5. Radium equivalent activities calculated for all the samples.

Table 3 and Figures 5 show the Ra_{eq} for construction materials, which has an average value of 300.65 $Bqkg^{-1}$, less than the global average range values suggested.

The absorbed dose rate in air (D_{in}) due to gamma ray emission from the radionuclides (^{226}Ra, ^{232}Th, and ^{40}K) in the building materials was calculated using Equation (2) given by European Commission Report [4]. Dose conversion coefficients were calculated for a standard room with dimensions of 4 m×5m×2.8 m.

$$D_{in}\ (nGy\ h^{-1}) = 0.92\ A_{Ra} + 1.1\ A_{Th} + 0.08\ A_K \qquad (2)$$

The annual effective dose (De, in $mSvy^{-1}$) to an occupant from the gamma radiation originating from the building materials can be assessed using the Equation (3):

$$D_e = D_{in}\ (nGy\ h^{-1}) \times 7000\ (h) \times 0.7\ (Sv\ Gy^{-1}) \times 10^{-6} \qquad (3)$$

The value of 0.7 $SvGy^{-1}$ is the conversion factor from the absorbed dose in air to the effective dose received by adults and 7000h is the exposure time, assuming that the humans spend 80% of the time in 1 year in the room (8760 h), on average, around the world [4], [6].
The estimated D_e values ranged from 0.01 to 2.65 mSv with an average of 1.30 mSv.
Radiation exposure due to building materials can be divided into external (gamma index) and internal (alpha index) exposure. The external exposure is caused by direct gamma radiation. The internal exposure is caused by the breathing of ^{222}Rn, ^{220}Rn and their short-lived decay products [4].
For building materials, the gamma index (Iγ) can be calculated using the equation (4) [4]:

$$I_\gamma = \frac{A_{Ra}}{300\ Bq\ kg^{-1}} + \frac{A_{Th}}{200\ Bq\ kg^{-1}} + \frac{A_K}{3000\ Bq\ kg^{-1}} \qquad (4)$$

The excess of alpha radiation caused by the inhalation of radon exhalated from building materials was estimated through the alpha index (Iα), i.e. an internal index, calculated according to equation (5) [7-9]:

$$I_{\propto} = \frac{A_{Ra}}{200\,Bq\,kg^{-1}} \qquad (5)$$

where A_{Ra} is the activity concentration of ^{226}Ra in Bqkg^{-1} and if this value exceeds 200 Bqkg^{-1} in the building material, inhalation of radon from this material may cause the indoor radon concentration to exceed 200 Bqm^{-3} [10]. Results of the absorbed dose rate in air, the annual effective dose, the gamma and alfa index are reported in Table 4.

Table 4. Results of the D_{in}, D_e, Iγ and I_α

code	D_{in} (nGy h^{-1})	Iγ	Iα	D_e (mSvy^{-1})
IT13	353.39	1.48	0.42	1.73
IT47	139.25	0.57	0.26	0.68
IT 48	440.19	1.84	0.63	2.15
IT 49	112.79	0.47	0.05	0.55
IT50	1.35	0.005	0	0.006
IT51	540.64	2.31	0.4	2.64

The obtained gamma index (Iγ), alpha index (Iα) and the annual effective dose (D_e) of the building samples are shown in Figures 6.

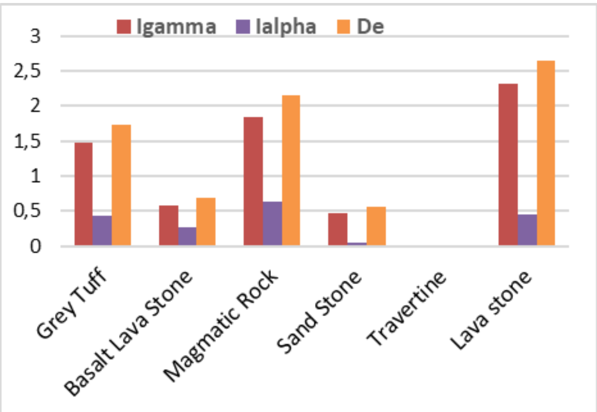

Fig. 6. Gamma index (in red), alpha index (in violet), annual effective dose (in orange) calculated for the investigated samples.

IV. CONCLUSION

In this study, the determination of the ^{226}Ra, ^{232}Th and ^{40}K activity concentrations in six samples of different building materials from Italian quarries was performed by gamma spectrometry. The obtained results were compared with other countries and world average results.

The average activity concentrations ^{226}Ra, ^{232}Th and 40K (60.54, 105.13 and 1165.81 Bqkg^{-1} respectively) of the analyzed building materials were higher than world average values (50 Bqkg^{-1} for ^{226}Ra and ^{232}Th and 500 Bqkg^{-1} for ^{40}K) [11]. Grey tuff (IT13) peperino (IT48) and lavastone (IT51) present concentrations of all the three radionuclides over the world average value, travertine (IT50) the lowest values, instead.

The estimated D_{in} values for all the materials samples ranged from 1.35 to 540.64 nGyh^{-1} with an average of 264.61 nGyh^{-1}. This value is above the world average value of 55 nGyh^{-1} for building materials [12].

In this study, the average value of Iγ obtained from some samples are above the criterion of unity corresponding to the annual effective dose 1mSv for structural applications.

It is clear that the Ra_{eq} values in the present study are generally higher than the recommended upper value of 370 Bqkg^{-1}. In literature, for a building material, dose criteria for exemption (annual effective dose) of 0.3mSvy^{-1} corresponds to a gamma index of Iγ ⩽0.5, whereas the top dose criterion of 1 mSvy^{-1} is satisfying for Iγ⩽1. The value of I γ are over the criterion of 0.5 corresponding to an annual effective dose 0.3 mSv except for the travertine sample. The values of gamma index are above unity for all samples analyzed except travertine. And also, the average value of D_{in} is higher than the dose limit value of 1.

So, since some values are over the recommended limits in order to complete the radiological characterization of the investigated samples and the assessment of the health risk related to the use of these building materials the measurements of the radon activity concentration and the calculation of the radon exhalation rate should be evaluate and discussed in future works.

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