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Natural Radioactivity and Radon Exhalation Rate of Sardinian Dimension Stones

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Abstract

Due to the ever-growing public concern about radon risk arising from building materials, the radon exhalation rates and the natural radioactivity of eighteen Sardinian dimension stones mostly available in the international market (granite, marble, basalt and trachyte) have been estimated. First, the radon accumulation inside a sealed chamber containing the rock sample in its natural form has been monitored, according to the specifications of IAEA (*International Atomic Energy Agency*) for measurement and calculation of radon release from NORM (Naturally Occurring Radioactive Material). After reducing the natural stone samples to powder, smaller samples of 100 g were isolated to determine the content of natural radionuclides ^{226}Ra, ^{232}Th, and ^{40}K by means of gamma-ray spectrometry. The *Activity Concentration Index* (I_γ) defined by EC-BSS (European Commission - Basic Safety Standards for protection against the dangers arising from exposure to ionizing radiation) together with the *External Radiation Hazard Index* (H_{ex}) and the *Effective Specific Activity* (A_{eff}) were finally calculated and compared with the reference level

introduced by the 2013/59/Euratom Directive. A good correlation between the radon release rate and ^{226}Ra activity was found for fourteen samples out of eighteen. The contribution to indoor radon accumulation was also simulated for a reference room made of Rosa Limbara (the dimension stone with the highest radon exhalation rate) and the resulting concentration compared with the limit value establish by law.

Keywords: *Sardinian dimension stones; natural radioactivity; indoor radon level; gas radon exposure; Activity Concentration Index.*

Introduction

Prolonged exposure to radon gas (Rn), known as the main contributor to the overall natural radiation dose absorbed by population, may increase the risk of developing lung cancer [1]. The main source of indoor radon is the pressure-driven influx from subsurface soil, though building materials with high concentrations of Radium (Ra) may represent another significant font of natural radiation [2]. The contribution of building materials to radon exposure can be expressed either as whole-body exposure to gamma radiation or as an internal dose of inhaled radon [3]. To evaluate the exposure to gamma radiation, which mainly originates from the presence of primordial radionuclides (i.e. ^{226}Ra, ^{232}Th and ^{40}K) in the building material, the following indexes, expressed by equations from 1 to 3 respectively, are commonly used: the activity concentration index (I_γ) [4], the external radiation hazard (H_{ex}) [5] and the effective specific activity (A_{eff}) [6].

$$I_\gamma = \frac{C_{Ra}}{300^{Bq/kg}} + \frac{C_{Th}}{200^{Bq/kg}} + \frac{C_K}{3000^{Bq/kg}} \quad (1)$$

$$H_{ex} = \frac{C_{Ra}}{370^{Bq/kg}} + \frac{C_{Th}}{259^{Bq/kg}} + \frac{C_K}{4810^{Bq/kg}} \quad (2)$$

$$A_{eff}\left(\frac{Bq}{kg}\right) = C_{Ra} + 1.31\ C_{Th} + 0.085\ C_K \quad (3)$$

where C_{Ra}, C_{Th} and C_K are the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K, respectively. The building materials should be restricted in their use if their activity concentration index is higher than 1 ($I_\gamma > 1$), which corresponds to an effective annual dose exceeding 1 mSv

[4]. The H_{ex} index must be less than the unity for the radiation hazard to be negligible [7] and A_{eff} should not exceed the value of 370 Bq/kg for the material to be used for newly built dwellings and public buildings [8]. Radon (^{222}Rn) is the decay product of ^{226}Ra and a member of ^{238}U decay chain. Traces of uranium/radium can be found in different rock types, with variable concentrations in a wide range from low to very high. In the case of igneous rocks, the occurrence of uranium is more common in crystalline rocks, like granites, granitic pegmatites, and syenites [9]. Uranium content in those rocks is mainly incorporated into accessory minerals, such as monazite, allanite, sphene, and zircon [10]. Measurements of U/Ra activity can provide useful information about radon release potential [9]. However, in order to determine the radon real-time exhalation rate from the surface of given building material, direct measurements need to be performed [11]. It is worth noticing that the exhalation rate of radon from a given stone does not only depend on U/Ra activity but also on the petrographic and petrophysical characteristics of the sample under examination: micro-fissure condition, grain size, arrangement, alteration degree and contact surfaces between constituents [9]. The present study discusses the results of the experimental tests carried out on 18 Sardinian dimension stones, aimed at estimating their natural radioactivity and radon emission potential. Two criteria were taken into consideration for the selection of the rock to be investigated: to be commonly available in the market and, secondly, to be among those rocks that are believed to pose a higher risk (i.e.: potential radon sources), according to the results of previous studies [2, 3 and 11] in which the amount of radon exhalation from various building materials has been studied.

Geological and petrographic characteristics of tested samples

Sardinian geology covers a wide range of paleontological features. About half of the surface area of the island (13,000 km^2) dates back to the Paleozoic era, while the remaining part (11,000 km^2) mostly belongs to the Tertiary and Quaternary [12, 13]. The granitoid rocks are one of the most important lithotypes outcroppings in Sardinia and constitute the calico-alkaline plutonic complex, extended for about 6,000 km^2 [14]. This outcrop is a sequence of Corsican- Sardinian batholite formed in the late stages of the Hercynian orogenesis (Upper-Permian Carboniferous). The Sardinian batholite is made up of 65%

monzogranitic magmas, and subordinately, for about 34%, of leucogranites, tonalites, and granodiorites, and finally, for less than 1%, of the rare gabbroid masses [15]. A basic geological map of Sardinia with the location of the main quarries is reported in Figure 1 [13].

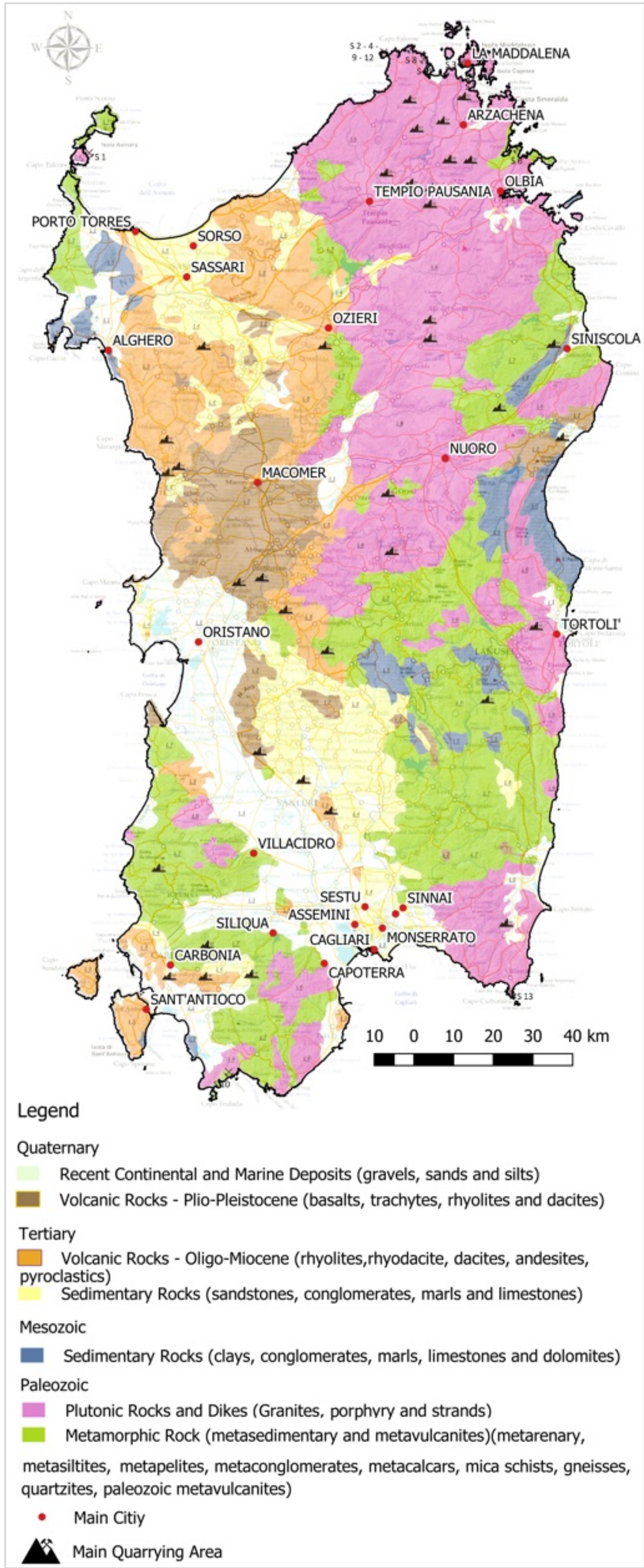

Fig. 1 Simplified geological map of Sardinia and main quarrying areas [13]

The most important dimension stones extracted in Sardinia are granite, marble, basalt and pyroclastic stones (generally marketed as trachyte, in part without distinction). Marbles and granites are mainly quarried in central-eastern and northeastern Sardinia, while extraction sites of basalts and pyroclastic rocks are widespread in a large portion of central-western Sardinia [15]. Figure 2 represents the Sardinian dimension stones mostly marketed worldwide.

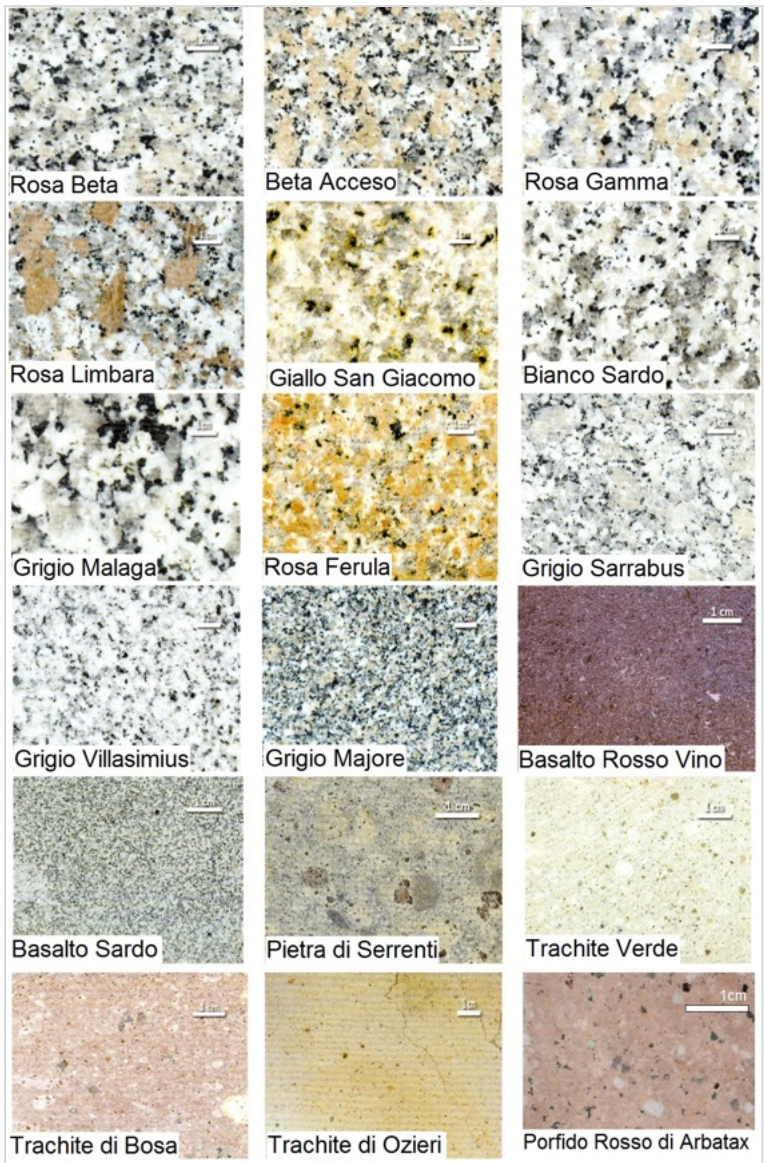

Fig. 2 Sardinian dimension stones available in the market [13].

This study includes eighteen dimension stone samples and specifically: ten granites, four pyroclastic rocks, two basalts, a syenite, and a porphyry rock. Among the above-mentioned dimension stones under investigation, granites represent the most interesting case study, as they are widely exported to be used for interior decoration, due to their natural beauty, while the presence of radioactive minerals in granite outcrops poses some concerns in terms of radon emission potential.

EXPERIMENTAL TEST

Radon exhalation measurements

Direct measurements of radon exhalation rate were performed on each rock sample of acknowledged volume and surface, maintained in its natural form, first polished and then enclosed in a 10.4 liter chamber together with a real-time radon-monitoring instrument (Radex MR-107). This approach is based on the principle of hermetically enclosing or covering a sample of soil or building material with a container/accumulator and measuring the radon activity growth inside [11, 16, and 17]. The container was sealed with a chemically resistant adhesive rubber. Radon concentration, temperature and relative humidity inside the chamber were registered (1 value each hour, for 120 h test duration). The radon activity concentration inside the sealed chamber can be described with the two-dimensional diffusion theory [18]; considering both back diffusion and radon leakage, the following formula can be used to express the variability of the radon concentration over time:

$$C_{(t)} = C_0 e^{-t\lambda_e} + C_m(1 - e^{-t\lambda_e}) \quad (2)$$

where λ_e is the effective radon decay constant (h^{-1}), which accounts for the radon decay, the leak rate of the system and the so-called ‘‘back diffusion”; C_0 and C_m are the radon activity concentration ($Bq.m^{-3}$) at time = 0 and its maximum value, respectively [16]. As shown in Fig.3, using the nonlinear least squares fitting of the experimental data with Eq.2 [18-20], for each sample the activity growth was modeled and the parameters λ_e and C_m extrapolated. Finally, the exhalation rate (E_{Rn}) from sample surface was estimated [21], according to Eq. 3:

$$E_{Rn} = C_m \lambda_e V_{eff}/S \qquad [Bq/m^2h] \qquad (3)$$

where V_{eff} is the effective volume of the calibration container [m^3] and S is the total surface area of the sample [m^2] [16]. The radon exhalation rates were determined for all samples under exam, according to the procedure above described.

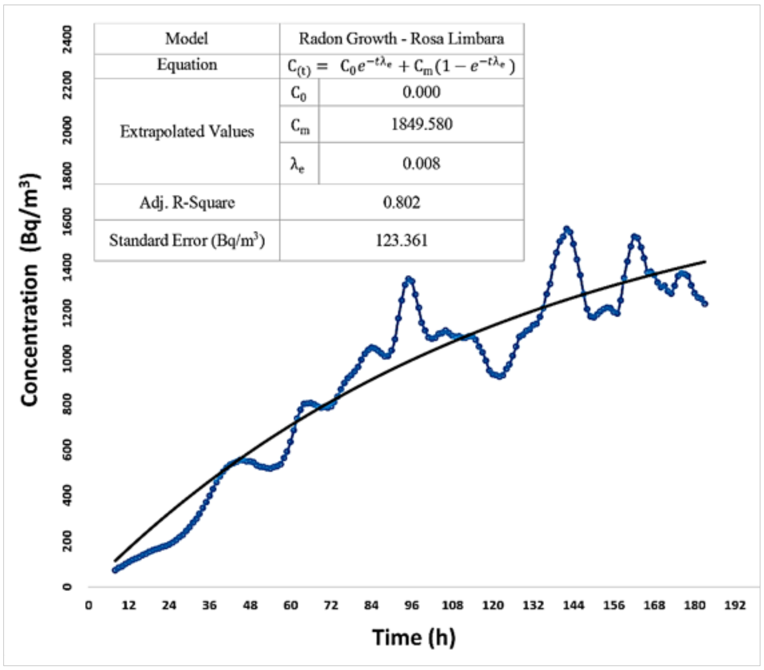

Fig. 3 Radon activity concentration vs. time (Rosa Limbara)

Measurement of Radionuclide activity concentrations

Each rock samples was cut, crushed and grounded to a fine powder. The powdered samples were oven-dried and about 100 gr of powder with grain size less than 1 mm was separated from each sample. The 100 gr were placed inside polyethylene cylindrical containers, sealed and left undisturbed for four weeks to reach the radioactive equilibrium between radon and its progenies. The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K were analyzed by performing gamma-ray spectrometry with a 1024 channels NaI(Tl) scintillation detector (3 × 3 inches), which was protected from background radiation with a lead shield. Analysis of the spectra was carried out using Ortec MAESTRO software. Radioactive sources of ^{137}Cs and ^{60}Co have been used for energy calibration. The energy resolution was calculated to be 10.36 % with reference to the full width of the peak at half of the maximum count level at 661 keV photopeak of ^{137}Cs source. The RGU1, RGTh1 (uranium and thorium reference sources from IAEA) and high pure potassium nitrate (KNO_3) salt were employed to calibrate the spectrometer. The spectrum of each reference material was acquired. It can be assumed that the overall spectrum due to the superposition of the various radioisotopes is obtained by the linear combination of the spectra of the single radioisotopes. In order to consider the background effect, further to three base spectra (Uranium, thorium, and potassium), the background spectrum was also acquired. Then the spectrum of each sample was obtained. Using a specially designed analysis program and the coefficients of the linear combination of the three base spectra plus the background spectrum, a new spectrum was built for each sample.

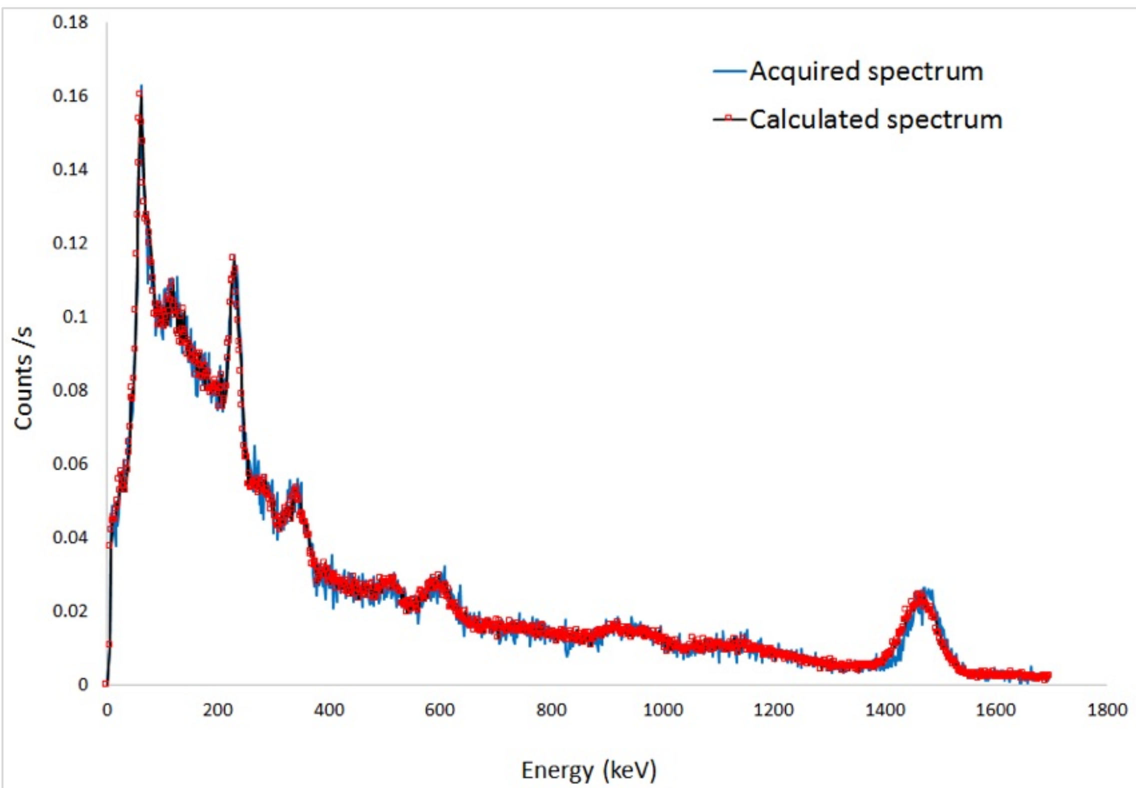

Fig. 4 Comparison of acquired and calculated spectrum (Rosa Limbara)

As can be seen in Fig.4, the built spectrum was compared with the acquired spectrum. Considering that the mass activity of the references and their base spectra were known, the specific activity of the three main radioisotopes in a given sample can be obtained by finding the optimal solution of the problem whose purpose is to identify the contribution of each of the three radioisotopes and background effect.

The ^{238}U, ^{232}Th, and ^{40}K activity concentration were estimated using the method above described. ^{226}Ra concentration is expressed in equivalent equilibrium concentration of ^{238}U. Using the ^{238}U and ^{226}Ra mass ratio ($U_{natural}$: ^{226}Ra = 1: 3.376×10^{-7} [22]), the specific activity of ^{226}Ra was also obtained (See Tab.1).

Results and discussion

Tab.1 summarizes the test results together with the relevant petrographic information and provenance for each sample [13]. The activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K is in the range of 3.03 – 79.93 Bq/kg (mean value 29.49), 4.36 – 97.49 Bq/kg (mean value 51.87) and 238.48 – 1826.31 Bq/kg (mean value 1003.70) respectively. The mean radionuclide concentration of ^{226}Ra and ^{232}Th in Sardinian dimension stones results lower than the average value worldwide (mean reference value), which is 50 Bq/kg for both ^{226}Ra and ^{232}Th [23]. However, the mean activity concentration for ^{40}K resulted twice the mean reference value (500 Bq/kg) [23], which may be correlated to high contents of potassium feldspar minerals in the tested samples. To assess the radiation hazard due to whole-body exposure or to radon intake through inhalation and ingestion, the radiological parameters I_γ, H_{ex}, and A_{eff} (Table 1) were calculated: none of those indexes exceeded the reference limits. The results are in agreement with previous studies [15, 24].

As regards radon exhalation rates, the detected levels were found between zero (less than detectable level) and 149.73 ± 9.99 $Bq.m^{-2}.d^{-1}$ (Rosa Limbara), with an average value of 60.40 ± 44.79 (SD). As anticipated, the lowest exhalation rate values were estimated for basalts. Although granites show a wide range of exhalation rates, in general, higher mean values were found for granites compared to the other rock types. Among granites, Leucogranites has higher mean exhalation rates compared to Monzogranites.

Tab. 1 Radon exhalation rate, radionuclide contents and radiological indexes of studied samples

Rock type	Commercial name	Petrography	Quarry location	Exhalation rate ± SE* (Bq/m^2d)	Radionuclide Concentration (Bq/kg)			I_γ	H_{ex}	A_{eff} (Bq/kg)
					Ra ± σ*	Th± σ*	K± σ*			
Granite	Rosa Beta Acceso	Monzogranite	Aggius, Arzachena, Luogostano, Luras, Sant Antonio di Gallura, Tempio P.	32.07 ± 5.63	46.21 ± 1.91	45.02 ± 1.46	761.93 ± 53.33	0.63	0.46	170
	Rosa Beta (β)	Monzogranite		26.80 ± 2.30	26.33 ± 2.04	54.83 ± 1.56	1049.10 ± 56.96	0.71	0.50	187
	Giallo San Giacomo	Leucogranite-Equigranular	Luogostano, Olbia, Sant Antonio di Gallura	74.69 ± 3.42	79.93 ± 2.05	72.58 ± 1.43	1057.42 ± 47.75	0.98	0.72	265
	Rosa Ferula	Leucogranite-Equigranular	Orosei	86.09 ± 6.49	33.01 ± 1.89	82.35 ± 1.45	1375.79 ± 52.90	0.98	0.69	258
	Grigio Malaga	Monzogranitic Granodiorite	Buddusò	41.75 ± 12.15	18.40 ± 1.68	53.28 ± 1.29	709.18 ± 47.01	0.56	0.40	148
	Grigio Sardo Sarrabus	Leucogranite	Castiadas	142.47 ± 14.28	35.43 ± 1.84	73.69 ± 1.41	944.72 ± 51.36	0.80	0.58	212
	Granito di Villasimius	Monzogranodiorite	Villasimius	23.62 ± 0.10	22.97 ± 1.72	35.78 ± 1.32	813.23 ± 48.21	0.53	0.37	139
	Rosa Limbara	Monzogranite-non-Equigranular	Aggius, Calangianus, Luras, Sant Antonio di Gallura (Priatu)	149.73 ± 9.99	24.85 ± 2.02	61.08 ± 1.55	956.16 ± 56.59	0.71	0.50	186
	Rosa Gamma	Leucogranite	Luras	20.81 ± 4.17	31.21 ± 1.92	61.94 ± 1.48	1044.93 ± 53.72	0.76	0.54	201
	Bianco Sardo	Monzogranite	Buddusò	36.89 ± 1.33	28.36 ± 1.97	97.49 ± 1.26	1078.25 ± 55.08	0.94	0.68	248
Pyroclastic Rock	Trachite di Ozieri	Pyroclast, Ignimbrite	Ozieri	24.98 ± 3.44	8.19 ± 1.93	11.79 ± 1.49	238.48 ± 54.13	0.17	0.12	44
	Pietra di Serrenti	Dacitic and Riodacitic Pyroclast	Serrenti	133.07 ± 7.41	35.36 ± 2.58	41.42 ± 1.98	1777.97 ± 72.18	0.92	0.63	241
	Trachite verde di Fordongianus	Dacitic and Riodacitic Pyroclast, Ignimbrite, Trachyte	Fordongianus	36.79 ± 1.54	32.72 ± 2.54	42.45 ± 1.95	1826.31 ± 59.23	0.93	0.63	244
	Trachite di Bosa	Dacitic and Riodacitic Pyroclast	Bosa	22.25 ± 3.29	20.49 ± 2.06	32.16 ± 1.59	1435.62 ± 57.74	0.71	0.48	185
Basalt	Sardinian basalt	Basalt	Bauladu, Mogoro, Paulilatino, Sardara	ND*	7.69 ± 1.77	9.46 ± 1.36	344.51 ± 49.60	0.19	0.13	49
	Basalto Rosso Vino	Basalt	Paulilatino	ND*	3.03 ± 1.74	4.36 ± 1.34	307.50 ± 48.61	0.13	0.09	35
Syenite	Grigio Majore	Monzogranite-Equigranular	Villagrande Strisaili, Tortoli	67.20 ± 6.02	47.19 ± 2.11	86.38 ± 1.35	1205.77 ± 53.63	0.99	0.71	263
Porphyry	Porfido Rosso di Arbatax	Porphyry Granite	Arbatax	47.13 ± 4.50	29.39 ± 1.87	67.61 ± 1.44	1139.67 ± 52.35	0.82	0.58	215

SE: Standard Error of the Estimated Value, σ*: Associated Uncertainty (1σ), ND*: Not Detectable.*

Correlation between radium content and radon exhalation rate

Uranium/radium levels in a given stone depend on the genetic nature of the rock, degree of evolution and mineralogical composition [9]. The sample with the highest Ra activity concentration was Giallo San Giacomo (Leucogranite), but this rock is not the one with the highest radon exhalation rate. This can be explained by the fact that radon atoms released from Ra-bearing minerals may remain trapped in the rock, due to its textural features. In fact, when ^{226}Ra atom decays a radon atom together with an alpha particle is formed [9]. The radon atom is ejected from the crystal or molecular lattice in an opposite direction to the surface. Based on the recoil energy and its direction, not all the radon atoms but a fraction of them can escape from the rock structure [25]. Presence of microfractures and porous media in rock structure can facilitate radon escape and increase the probability to reach outside environments [9]. In the case of rock samples Rosa Limbara, Grigio Sardo Sarrabus, Pietra di Serrenti and Rosa Ferula, although the activity concentrations of Ra are not considerable, high radon exhalation rates were observed (see also Fig.5). As mentioned above, this may be explained by the structure physical features of the rock samples that can provide pathways for radon transfer and facilitate radon exhalation from source to rock surface. As shown in Fig.6, except for four samples (Rosa Limbara, Grigio Sardo Sarrabus, Pietra di Serrenti, and Rosa Ferula), a significant correlation ($R^2 = 0.60$) was found between radon exhalation rate and radium content. The result confirm that U/Ra content in the rock sample can provide valuable information on the presence of radon source, but the actual exhalation rate remains strongly influenced by the rock's physical characteristics (e.g. density, porosity, etc.) [26].

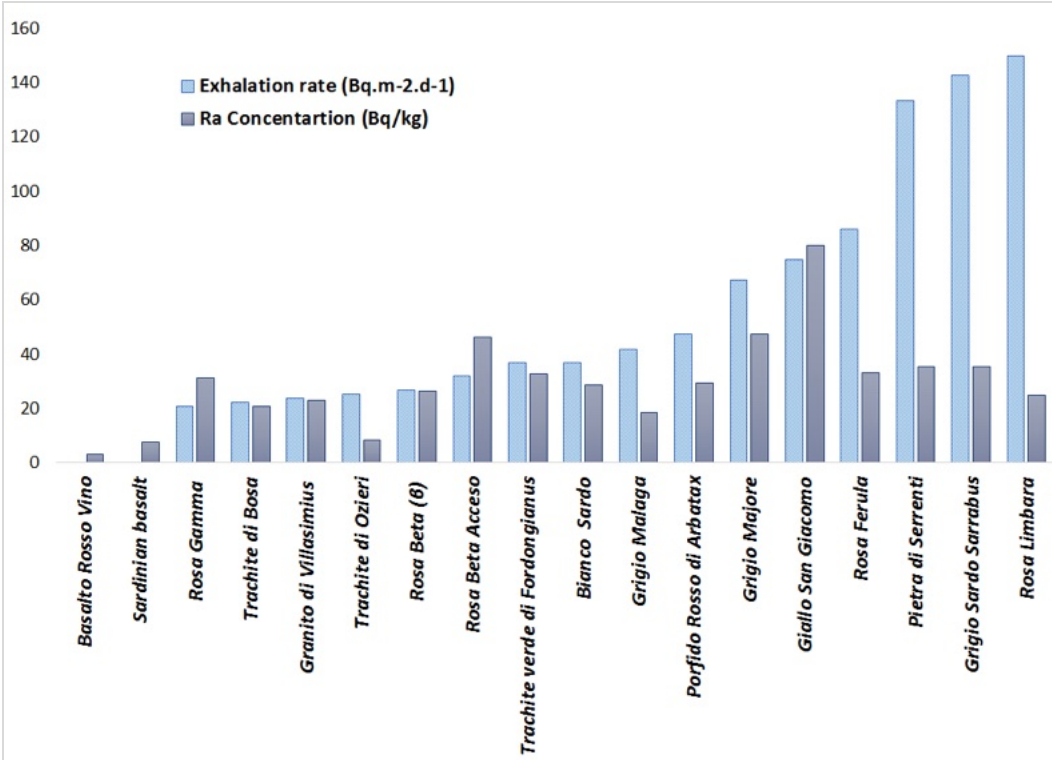

Fig. 5 Radon exhalation rate and radium concentration for each stone sample

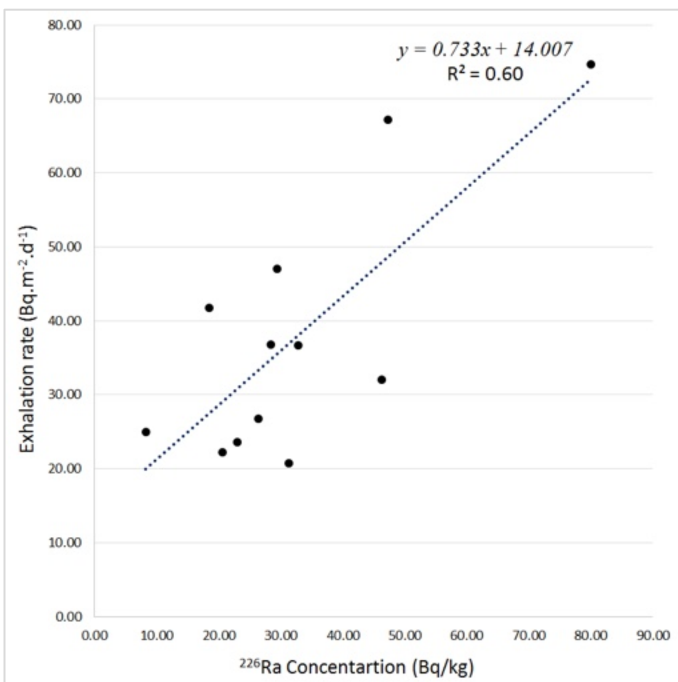

Fig. 6 Correlation between radon exhalation rate and radium content (Rosa Limbara, Grigio Sardo Sarrabus, Pietra di Serrenti, and Rosa Ferula)

Contribution of building materials to indoor radon concentration

With the necessary schematic assumptions and based on the results of the radon exhalation test, it is possible to calculate the contribution to indoor radon accumulation of a given dimension stone, hypothetically used to confine a defined volume. The indoor radon concentration originated from the confining walls can be easily estimated as follow [27, 28]:

$$C_{Rn} = E_{Rn}A_r/V_r\lambda_v \quad (4)$$

where A_r is the area of the wall surface; V_r is the volume of the confined space and λ_v is the room ventilation rate. The Ratio A_r/V_r is taken to be 1.6, considering a standard room of 4×5×2.8 m^3, while the ventilation rate is between 0.2 and 2 h^{-1} [28]. In this study, the highest calculated value of E_{Rn} was about 6.29 $Bq.m^{-2}.h^{-1}$ for Rosa Limbara rock sample. Considering the lowest air ventilation rate (λ_v = 0.2), the estimated indoor radon concentration would be about 50 $Bq.m^{-3}$, which is lower than the recommended level stated by the article 103 of the EC Directive 2013/59/Euratom (300 $Bq.m^{-3}$).

Conclusion

In order to contribute to the overall characterization of the Sardinian dimension stones mostly available in the international market, the radon release rates and the natural radioactivity of eighteen selected samples were estimated. Radon exaltation measurements were performed on the rock samples in their natural form, according to the specifications of IAEA (International Atomic Energy Agency). Smaller powdered samples (100 g) were isolated to determine the content of the natural radionuclides ^{226}Ra, ^{232}Th, and ^{40}K, by means of gamma-ray spectrometry. The Activity Concentration Index (I_γ) defined by EC-BSS together with the External Radiation Hazard Index (Hex) and the Effective Specific Activity (Aeff) were calculated and found below the reference limits introduced by the 2013/59/Euratom Directive. A comparative analysis of the test results indicates higher levels of ^{226}Ra activity for Giallo San Giacomo, Rosa Beta Acceso and Grigio Majore respectively, whereas higher radon release rates were found for Rosa Limbara, Grigio

Sardo Sarrabus and Pietra di Serrenti. A good correlation between the radon release and the ^{226}Ra activity was found for fourteen dimension stones. The essential contribution of the stone's physical characteristics in providing radon escape paths from inner rock to surface was confirmed. The indoor radon accumulation was simulated for a reference room made of Rosa Limbara; the model proved that even with poor ventilation radon concentration remains below the reference level of 50 Bq/m^3 introduced by 2013/59/Euratom Directive.

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