ACTA IMEKO
ISSN: 2221-870X
September 2026, Volume 15, Number 3, 1 - 7

Evaluation of Radon exhalation rate, natural radioactivity content, and radiological hazard for untreated and aged Viterbo tuff stone

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ABSTRACT
This research investigates the radon exhalation rate, natural radionuclide content, and associated radiological hazard to human health linked to Viterbo tuff stone, a material of notable historical and architectural relevance. Particular attention was given to radon exhalation behavior by examining both untreated and accelerated aged (through a salt crystallization test) samples. Radon exhalation rates were quantified using the Closed Chamber Method (CCM) in combination with a Durridge RAD7 real-time continuous radon detector. The measured exhalation rates were subsequently employed in the Markkanen room model to estimate potential indoor radon concentrations.
In addition, the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K were determined by means of high-purity germanium (HPGe) gamma-ray spectrometry. Radiological hazard was evaluated using several indices, including the absorbed gamma dose rate (D), the annual effective dose equivalent ($AEDE$), the activity concentration index (I), and the alpha index (I_{α}).
Noteworthy, the findings of this study provide a valuable basis for future research aimed at developing a systematic framework for assessing radon release from building materials. Such an approach is essential for supporting decision-making processes, as it enables the identification of buildings that may be particularly susceptible to radiological risks, and therefore require targeted mitigation and protection measures.

Section: RESEARCH PAPER

Keywords: Radon exhalation rate; natural radioactivity; radiological hazard; Viterbo tuff; accelerated aging

Citation: F. Caridi, D. Chiriu, S. Da Pelo, Giuliana Faggio, M. Guida, G. Messina, M. Ponte, S. A. Ruffolo, D. Majolino, V. Venuti, Evaluation of Radon exhalation rate, natural radioactivity content, and radiological hazard for untreated and aged Viterbo tuff stone, Acta IMEKO, vol. 15 (2026) no. 3, pp. 1-7. DOI: 10.21014/actaimeko.v15i3.2327

Section Editor: Fabio Leccese, Università Degli Studi Roma Tre, Rome, Italy

Received February 18, 2026; **In final form** September 11, 2026; **Published** September 2026

Funding: This work was performed in the framework of the project PRIN 2022 PNRR ATHENA (A novel approach Towards the management of building materials of particular Historical-artistic interest: assessment of the radon Exhalation and the radiological risk due to Natural radioActivity content), CUP J53D23014560001, funded by the European Union - Next Generation EU, PNRR - Mission 4, Component 2, Investment 1.1 - PRIN 2022 PNRR Call for Proposals - Directorial Decree No. 1409 of 14-09-2022.

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1. INTRODUCTION

Human exposure to ionizing radiation arises from both natural and anthropogenic sources. According to the UNSCEAR 2000 report, the average annual effective dose is estimated to be 2.4 mSv from natural radiation and 0.6 mSv from artificial sources [1].

Natural background radiation accounts for the majority of the external dose received by the global population. Naturally occurring radionuclides, such as ^{238}U, ^{232}Th, and ^{40}K, are widely distributed in environmental matrices including soil, sand, rocks, water, and building materials [1]. Their concentration and spatial distribution vary significantly worldwide, depending on regional geological characteristics. Specifically, different rock types contain uranium and/or radium in concentrations ranging from 2–5 parts per million, typical of background levels, up to approximately 1000 parts per million in black shales [2]. Among igneous formations, crystalline rocks such as granites, granitic pegmatites, and syenites are particularly enriched in uranium [3]. In these lithologies, uranium and radium are predominantly hosted in accessory minerals, including zircon, monazite, allanite, and sphene [4].

Conversely, exposure to artificial sources of ionizing radiation primarily results from the application of radiation in medical diagnostics and therapy, including imaging procedures and radiotherapy [5], [6]. Additional contributions arise from nuclear accidents, such as those at Chernobyl and Fukushima, which led to the release of several artificial radionuclides into the environment, including ^{131}I, ^{137}Cs, ^{134}Cs, and ^{90}Sr [7].

Among natural radiation sources, radon (hereafter referred to as ^{222}Rn, the isotope with the highest abundance) represents the most significant contributor to internal human exposure. Radon is a colorless, odorless, and chemically inert noble gas generated by the radioactive decay of ^{238}U. It can migrate through different environmental compartments, including soil, water, and rocks. The primary source of radon is soil and underlying geological formations, which account for more than 69 % of total radon emissions [8]–[11]. Geological history and weathering processes play a crucial role in determining the geogenic radon potential (GRP) [12], [13]. A thorough understanding of these factors is fundamental for predicting radon release, supporting land-use planning, and implementing construction practices aimed at reducing indoor radon accumulation, thereby improving building safety and indoor environmental quality [14]. In addition, quarrying activities must consider the potential for radon release from extracted materials, which may affect both workers and the built environment. Radon has been classified as a carcinogenic agent, and prolonged exposure to elevated concentrations is strongly associated with an increased risk of lung cancer. This health risk is primarily related to radon decay products, particularly ^{218}Po and ^{214}Po, which emit alpha particles. While radon itself is chemically inert, these positively charged metallic progeny can attach to airborne particulate matter, facilitating inhalation and deposition in the pulmonary alveoli. Once retained in lung tissue, these particles can induce significant cellular damage, substantially increasing the probability of lung cancer development [15], [16].

Going on, building materials can emit gamma radiation in both indoor and outdoor environments, and long-term exposure to low doses of ionizing radiation may adversely affect human health [17]. Given that individuals spend a considerable proportion of their time indoors, it is essential to evaluate the concentration of naturally occurring radionuclides in building materials (BM) used for residential construction [18]. Since 1989, the European Union (EU) has issued a series of regulations and directives aimed at limiting radiation exposure from building materials, taking into account the associated health risks in indoor environments [19]. Notably, in 1999, the European Commission (EC) introduced the ALARA (As Low As Reasonably Achievable) principle, establishing the minimization of radiation exposure from materials containing elevated levels of naturally occurring radionuclides as a key objective of radiation protection policies for building materials [19]–[20]. To support this objective, the EC introduced the activity concentration index (I), which is widely employed as a screening parameter to control gamma radiation exposure from construction materials. Over time, additional indices have been developed to provide a more comprehensive assessment of radiological hazards associated with such materials [18]. More recently, radiological protection of public health has been further addressed in European Directive 2013/59 EURATOM, which has been implemented in Italy through Legislative Decree D.Lgs. 101/2020 and subsequent amendments [21].

In this study, radon exhalation from Viterbo tuff, a natural stone extensively used as a building material of significant historical and artistic value, was investigated in both untreated and accelerated aged (through salt crystallization) samples. Radon exhalation rates were determined using the Closed Chamber Method (CCM), employing the experimental setup of the Durridge RAD7 real-time continuous radon monitor for alpha spectrometry of short-lived radon progeny. The Markkanen room model was subsequently applied to estimate potential indoor radon concentrations based on the measured exhalation rates.

Finally, the natural radioactivity content of the analyzed stone was determined using high-purity germanium (HPGe) gamma-ray spectrometry. The radiological hazard to humans was then evaluated by calculating the absorbed gamma dose rate (D), the annual effective dose equivalent ($AEDE$), the activity concentration index (I) and the alpha index (I_{α}).

2. MATERIALS AND METHODS

2.1. Sample description

The Viterbo tuff formation is characterized by a massive and homogeneous structure, with coloration ranging from yellowish-reddish tones to dark grey. It contains large fragments of black porphyritic scoria composed mainly of leucite/analcime and sanidine, embedded within a coarse cineritic matrix. In addition to altered leucite and sanidine, the phenocrystal assemblage includes clinopyroxene, plagioclase, and biotite, while accessory minerals are represented by iron and titanium oxides, and sphene. Lava inclusions (leucitite) of variable size are irregularly distributed throughout the formation [22].

From a geochemical perspective, the rock composition spans from phonolitic tephrite to phonolite. The ignimbritic matrix exhibits extensive zeolitization; however, studies specifically addressing the intrinsic zeolite content remain limited. The tuff is predominantly zeolitized to chabazite and, to a lesser extent, to phillipsite, with total zeolite contents reported to range between 20 % and 70 % [22]. Petrographic and mineralogical investigations reported in [22] indicate a high degree of lithological homogeneity and an almost complete absence of vertical zoning within the formation.

The studied bed displays a chaotic internal structure and a coarse-grained cineritic matrix texture, typically showing reddish hues with occasional yellow or intense red tones, though more frequently exhibiting grey to black coloration. The paragenesis of phenocrystals appears well defined: analcime and pyroxene are consistently observed across most outcrops, whereas mica is not ubiquitously present.

Despite its relatively high porosity, the Viterbo tuff demonstrates favorable mechanical properties, which has led to its widespread use in construction and paving applications.

2.2. Methods

For the evaluation of the radon exhalation rate, five aliquots of the investigated Viterbo tuff were prepared in the laboratory by cutting larger stone blocks using a circular saw. Each aliquot was shaped into a cube with an approximate side length of 5 cm.

Radon exhalation from each aliquot was measured using the Closed Chamber Method (CCM) [23], [24]. The experimental setup consisted of a small cylindrical stainless-steel chamber with a volume of 2.75 L, housing the sample and connected in a closed-loop configuration to a Durridge RAD7 real-time continuous radon monitor via vinyl tubing. A Laboratory Drying Unit (LDU) containing a calcium sulfate ($CaSO_4$) desiccant was incorporated into the circuit to control humidity levels [24]. Air circulation within the system was maintained by an internal mechanical pump of the monitoring instrument.

Air from the closed chamber was conveyed through the LDU and an inlet filter, designed to prevent the ingress of dust and radon progeny, before entering the ionization chamber of the RAD7 detector. Following alpha-particle detection, the air was returned to the stainless-steel chamber through an outlet filter, thereby completing the closed-loop system. Within the ionization chamber, the decay of ^{222}Rn in the filtered air generates short-lived alpha-emitting progeny, primarily polonium isotopes, which are subsequently detected.

A high voltage of 2500 V was applied between the walls of the ionization chamber and a solid-state silicon detector. The detected alpha particles were directly converted into electrical signals, allowing discrimination between pulses produced by alpha emissions from ^{218}Po, ^{216}Po, ^{214}Po, and ^{212}Po, corresponding to energies of 6.0 MeV, 6.7 MeV, 7.7 MeV, and 8.8 MeV, respectively. By exploiting the secular equilibrium between radon and its polonium progeny, the ^{218}Po alpha peak was used to determine the activity concentration of ^{222}Rn, enabling reliable measurements within a relatively short acquisition time of approximately 30 minutes [25]. The radon growth curve to equilibrium was monitored over a period of 10 days, and the ^{222}Rn specific exhalation rate (E, in Bq h^{-1} kg^{-1}) was calculated according to the following equation [24]:

$$E = \frac{(C - C_0\,\mathrm{e}^{-\lambda\,T})/m}{1 - \mathrm{e}^{-\lambda\,T}}\,\lambda\,V\,, \tag{1}$$

where C represents the equilibrium concentration (Bq m^{-3}), C_0 denotes the initial radon concentration (Bq m^{-3}), λ (h^{-1}) indicates the sum of the radon decay constant, the back-scattering constant, and the leakage constant, V stands for the total volume of the analytical system (m^3), T is the time of exposure (h), and m is the mass of the sample (kg). To minimize potential air leakage from the chamber, an insulating rubber seal was applied.

Following the measurement of radon exhalation from the untreated aliquots, the investigated specimens were subjected to accelerated aging through a salt crystallization test, also referred to as a salt weathering test. The experimental procedure was conducted in accordance with the recommendations of EN 12370:2001. Specifically, each aliquot was exposed to 15 crystallization cycles, each comprising of the following: (i) immersion for 2 hours in a supersaturated sodium sulfate solution (14 % w/w at 20 °C); (ii) drying for 16 hours in a ventilated oven at 105 °C; and (iii) cooling for 6 hours at ambient temperature.

Figure 1 shows the investigated natural stone, before (a) and after (b) salt weathering.

The aliquots subjected to accelerated aging were subsequently analysed using the Closed Chamber Method (CCM), in order to determine their radon exhalation rates, and to compare the obtained values with those measured for untreated specimens.

In addition, a Markkanen room model simulation was performed [26], employing the experimentally determined radon exhalation rates for both untreated and artificially aged samples, to evaluate potential health risks associated with indoor radon exposure. This theoretical model was developed to estimate the equilibrium radon activity concentration within an enclosed space. For this purpose, the model incorporates two key aspects: first, the accumulation of radon released from building materials used in the construction of the room; and second, the influence of ventilation processes on indoor radon levels.

Specifically, the following parameters are considered: (i) the volume of the room and the surface areas covered by the analysed building materials (walls, floor, and ceiling); and (ii) the ventilation rate, defined as the volume of outdoor air exchanged with indoor air per hour.

The time-dependent variation of the radon activity concentration, $C(t)$, is described by a mass balance equation that accounts for radon exhalation from the investigated natural stone, as well as radioactive decay and air exchange due to ventilation [26]:

$$\frac{\mathrm{d}C(t)}{\mathrm{d}t} = \frac{E \cdot A}{V} - \left(\lambda_{222} + \frac{1}{\tau}\right) C(t)\,, \tag{2}$$

where $C(t)$ denotes the radon concentration (Bq m^{-3}) at the time t, E is the specific exhalation rate (Bq m^{-2} h^{-1}), A is the total area of the sample exhaling radon (m^2), V is the volume of the room (m^3), λ_{222} is the radon decay constant, and τ is the air exchange time (h).

The solution of the differential equation (2) is given by:

$$C(t) = C_\infty \left(1 - \mathrm{e}^{-\left(\lambda_{222} + \frac{1}{\tau}\right)\,t}\right). \tag{3}$$

The equilibrium radon concentration, C_∞, is reached when the rate of radon release from the building material is balanced by the combined effects of ventilation and radioactive decay:

$$C_\infty = \frac{\dfrac{E \cdot A}{V}}{\lambda_{222} + \dfrac{1}{\tau}}\,. \tag{4}$$

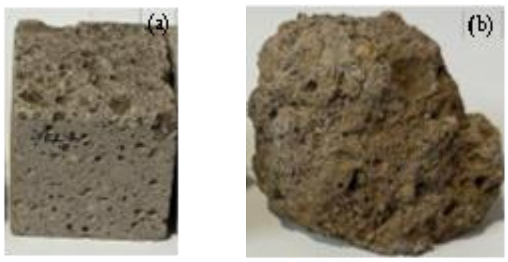

Figure 1. Viterbo tuff stone before (a) and after (b) salt weathering.

The ratio A/V is 1.6 for a standard room of 4 × 5 × 2.8 m^3, while the ventilation rate is assumed to be between 0.2 and 2 h^{-1} [26].

For the determination of the natural radioactivity content, five aliquots of the investigated Viterbo tuff were prepared in the laboratory starting from larger stone blocks, according to the procedure described above. Prior to analysis by high-purity germanium (HPGe) gamma-ray spectrometry, each aliquot was first pulverized and then oven-dried to completely remove residual moisture and to achieve a constant mass. The dried specimens were then placed in 1 L hermetically sealed Marinelli containers and stored for 40 days to allow secular radioactive equilibrium to be established between ^{226}Ra and its progeny. After this equilibration period, the specific activity of ^{226}Ra was determined [27].

Each sample was counted for a total acquisition time of 70000 s. The activity concentration of ^{226}Ra was evaluated using the gamma-ray emissions of its decay products, specifically the 295.21 keV and 351.92 keV lines of ^{214}Pb, and the 1120.29 keV line of ^{214}Bi. The activity concentration of ^{232}Th was determined from the 911.21 keV and 968.97 keV gamma lines of ^{228}Ac. The activity of ^{40}K was assessed using its characteristic gamma emission at 1460.8 keV.

The experimental setup consisted of a positively biased Ortec HPGe (GEM) detector [27], housed within a lead shielding assembly to reduce environmental background radiation. Energy and efficiency calibration was performed using a multi-peak 1 L Marinelli gamma reference source (AK-5901), covering an energy range of 60–1836 keV. The calibration source was custom-manufactured to replicate the geometry of the analyzed samples and embedded in a water-equivalent epoxy resin matrix. Data acquisition and spectral analysis were carried out using Gamma Vision software (Ortec) [28].

The activity concentration of the investigated radionuclides, expressed in Bq kg^{-1} dry weight (d.w.), was calculated according to the following equation [29]:

$$C = \frac{N_E}{\varepsilon_E \, t \, \gamma_d \, M}, \quad (5)$$

where N_{E} is the net area of a peak at energy E, ε_{E} and γ_{d} are the efficiency and yield of the photopeak at energy E, respectively, M is the mass of the sample (kg), and t is the live time (s) [30]. The γ-ray spectrometry measurements were conducted in accordance with the protocol established by the International Organization for Standardization (ISO), following the specific guidance of ISO 19017:2015 [31]. The quality of the experimental results was certified by the Italian Accreditation Body (ACCREDIA) [32], ensuring ongoing verification that the performance characteristics of the methodology are consistently maintained.

To evaluate the potential radiological hazard to humans, several indices were calculated, including the absorbed gamma dose rate (D), the annual effective dose equivalent ($AEDE$), the activity concentration index (I), and the alpha index (I_α). In particular, the absorbed gamma dose rate, D (nGy h^{-1}), was determined using the standard room model, as described in [33]:

$$D \; = \; 0.92 \, C_{\mathrm{Ra}} + \; 1.1 \, C_{\mathrm{Th}} + \; 0.08 \, C_{\mathrm{K}} \, , \quad (6)$$

where C_{Ra}, C_{Th}, and C_{K} are the average activity concentrations (the mean value of the five analyzed aliquots) of ^{226}Ra, ^{232}Th, and ^{40}K in the analyzed stone, respectively.

Assuming an employment factor of 80 % for indoor exposure, the annual effective dose equivalent ($AEDE$) (mSv y^{-1}) of an individual may be calculated as follows [33]:

$$AEDE = (D - 50) \times 8760 \, \mathrm{h} \times 0.7 \, \mathrm{Sv \, Gy^{-1}} \times 0.8 \times 10^{-6}, \quad (7)$$

where the average dose rate value of 50 nGy h^{-1} for the background was discounted. For a radiological danger to be considered minimal, $AEDE$ must be less than 1 mSv y^{-1} [10].

The radiation activity concentration index is defined by [33]:

$$I \; = \; C_{\mathrm{Ra}}/300 \; + \; C_{\mathrm{Th}}/200 \; + \; C_{\mathrm{K}}/3000 \, . \quad (8)$$

This index represents the additional dose contribution from γ-radiation originating from a building constructed with a specific material, above the typical external background exposure. For the radiological hazard to be considered negligible, its value should not exceed 1 [10].

The alpha index was calculated with the following formula [33]:

$$I_\alpha \; = \; C_{\mathrm{Ra}}/200 \, . \quad (9)$$

This index allows for the assessment of alpha radiation exposure resulting from radon exhalation of building materials indoors. The activity concentration of ^{226}Ra should not exceed 200 Bq kg^{-1} to ensure that indoor radon activity remains below the threshold value of 200 Bq m^{-3} [34]. Accordingly, I_α must remain below unity to minimize the radiological hazard associated with indoor radon exposure [34].

3. RESULTS AND DISCUSSION

Table 1 presents the mean ^{222}Rn exhalation rates, calculated from the five aliquots of the investigated Viterbo tuff, for both untreated and accelerated aged samples. The average activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K are summarized in Table 2.

It should be noted that the specific activities of ^{226}Ra, ^{232}Th, and ^{40}K are substantially higher than the corresponding global average values [1]. Previous studies have demonstrated that the chemical composition and mineralogical characteristics of natural stones significantly influence both the ^{222}Rn exhalation rate and the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K [35]. In light of these observations, further investigations have been planned to characterize the chemical and mineralogical composition of Viterbo tuff using X-ray diffraction (XRD) and micro-Raman spectroscopy techniques.

Additionally, Table 3 presents the equilibrium radon concentration, C_∞, for both untreated and accelerated aged Viterbo tuff, calculated assuming the lowest air exchange rate (τ = 0.2 h^{-1}).

Table 1. The average ^{222}Rn-specific exhalation rate for untreated and accelerated aged Viterbo tuff stone.

Sample	^{222}Rn (Bq h^{-1} kg^{-1}) untreated	^{222}Rn (Bq h^{-1} kg^{-1}) accelerated aged
Viterbo tuff	0.755 ± 0.263	0.195 ± 0.014

Table 2. The average ^{226}Ra, ^{232}Th, and ^{40}K activity concentrations, for the analysed natural stone.

Sample	^{226}Ra (Bq kg^{-1} d.w.)	^{232}Th (Bq kg^{-1} d.w.)	^{40}K (Bq kg^{-1} d.w.)
Viterbo tuff	125 ± 19	302 ± 35	2021 ± 251

Table 3. The equilibrium concentration of radon, C_∞, for untreated and accelerated aged Viterbo tuff stone.

Sample	C_∞ (Bq m^{-3}) untreated	C_∞ (Bq m^{-3}) accelerated aged
Viterbo tuff	39.8	15.6

It is noteworthy that Directive 2013/59/Euratom, transposed into Italian law by Legislative Decree 101/2020 and in force since 27 August 2020, introduced new measures for radon control in workplaces. Specifically, it established a reference level of 300 Bq m^{-3} as the annual average indoor radon activity concentration for workplaces and homes constructed before 31 December 2024. For dwellings built after this date, the reference level is set at 200 Bq m^{-3}.

Considering this regulatory framework and adopting the most conservative scenario, i.e. a threshold value of 200 Bq m^{-3}, the contribution of the investigated Viterbo tuff to indoor radon activity was found to be less than 20 % and 10 % of the threshold for untreated and accelerated aged samples, respectively. These results indicate that the radiological hazard to occupants from radon exhalation of this material can be considered negligible.

Furthermore, the absorbed gamma dose rate (D), the annual effective dose equivalent ($AEDE$), the activity concentration index (I), and the alpha index (I_α), calculated using equations (6)–(9), are presented in Table 4. The latter three indices are also illustrated in Figure 2, alongside the recommended safety thresholds.

In particular, the absorbed gamma dose rate (D) of 608 nGy h^{-1} can be attributed to the lithological characteristics of the investigated sample [36], and was subsequently used to calculate the annual effective dose equivalent ($AEDE$) according to equation (7).

Notably, under the highly conservative scenario considered, i.e. assuming continuous exposure for 8760 hours per year (24 hours per day for 365 days), the $AEDE$ was calculated to be 2.7 mSv y^{-1}, exceeding the action level of 1 mSv y^{-1} established by Italian legislation for members of the public [10]. Moreover, the activity concentration index (I) was found to be 2.6, surpassing the recommended threshold of 1. These findings indicate that, when considering the use of this natural stone in civil engineering applications, such as residential buildings or structures with high occupancy rates, a more detailed assessment of the radiological dose is warranted. This assessment should follow the provisions of Article 29, paragraph 5, of D.Lgs. 101/20 [10] and incorporate dose estimation methodologies recommended by national and international standards, accounting for factors such as material density, thickness, building type, and intended use [27], [37].

Finally, the alpha index (I_α) was determined to be 0.6, below the safety threshold of 1, thereby indicating that exposure to radon released from the investigated stone is unlikely to pose a significant health hazard.

Table 4. The calculated values of the radiological hazard indices for the investigated sample.

Sample	D (nGy h^{-1})	$AEDE$ (mSv y^{-1})	I	I_α
Viterbo tuff	608	2.7	2.6	0.6

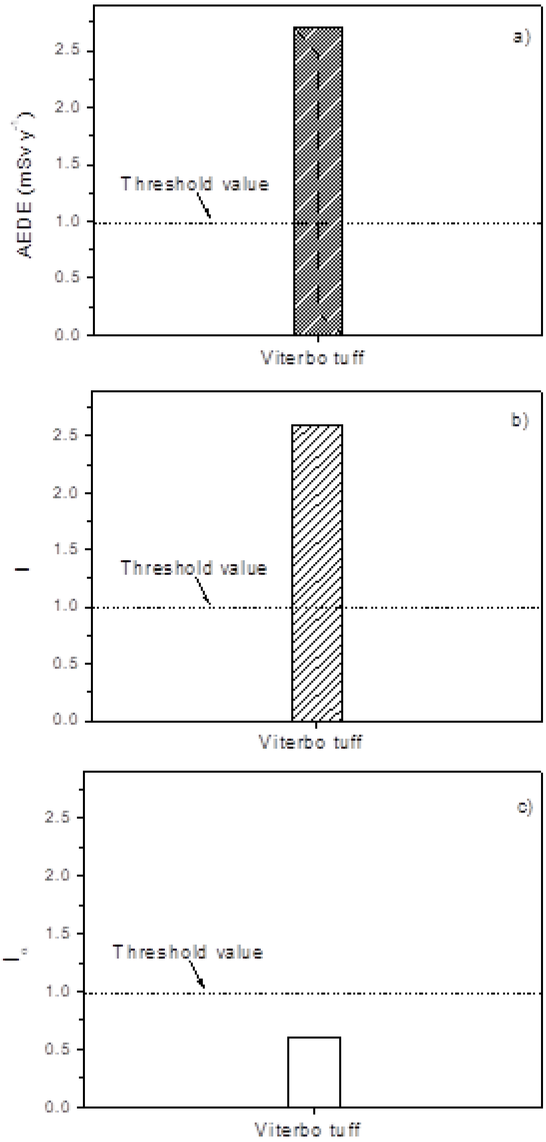

Figure 2. The annual effective dose equivalent ($AEDE$) (a), the activity concentration index (I) (b), and the alpha index (I_α) (c) for the investigated natural stone, together with recommended safety thresholds.

4. CONCLUSIONS

The radon exhalation rate of Viterbo tuff, a natural stone of significant historical and artistic value used as a building material, was investigated both in its untreated state and after accelerated aging via the salt crystallization test. Measurements were conducted using the Closed Chamber Method (CCM) with the Durridge RAD7 system, and the results are presented as a case study. Based on the measured radon exhalation rates, the contribution of this stone to indoor radon accumulation was estimated through the Markkanen room model. For untreated and accelerated aged samples, this contribution was found to be below 20 % and 10 % of the threshold value established by Italian Legislative Decree 101/2020 (200 Bq m^{-3}), respectively, thereby indicating that the radiological hazard to occupants from radon exposure is minimal. Notably, this methodology can, in principle, be applied to assess radon-related radiological hazards in a wide range of construction materials, providing a useful guideline for environmental quality monitoring and research.

Additionally, the natural radioactivity content of Viterbo tuff was determined using high-purity germanium (HPGe) gamma-ray spectrometry. The specific activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K were found to exceed global average values. To evaluate the associated radiological hazard, the absorbed gamma dose rate (D), annual effective dose equivalent ($AEDE$), activity concentration index (I), and alpha index (I_{α}) were calculated. Under the highly conservative scenario considered, the $AEDE$ exceeded the public exposure limit of 1 mSv y^{-1} specified by Italian legislation, and the activity concentration index (I) was also higher than the recommended threshold of 1. These results indicate that, for applications in civil engineering, such as residential buildings or structures with high occupancy, more detailed dose assessments are necessary in accordance with Article 29, paragraph 5, of D.Lgs. 101/20. Finally, the alpha index (I_{α}) was found to be below unity, confirming that the radiological hazard associated with radon exposure from the investigated stone, even at indoor concentrations above 200 Bq m^{-3}, is negligible.

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