2025 IMEKO TC26 International Conference on
Metrology for Archaeology and Cultural Heritage
Bergamo, Italy, October 15-17, 2025

# Radon exhalation, natural radioactivity content and radiological hazard assessment for the *Viterbo tuff* stone: a case study

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***Abstract*** **– This study evaluates the radon exhalation rate, natural radioactivity content, and radiological hazard associated with the *Viterbo tuff* stone, a material of historical and architectural significance. The radon exhalation rate was measured using the Closed Chamber Method (CCM) with the Durridge RAD7 Real-time continuous radon monitor, while the specific activities of $^{226}$Ra, $^{232}$Th, and $^{40}$K were determined using High Purity Germanium (HPGe) gamma-ray spectrometry. Radiological hazard was assessed through the following indices: absorbed gamma dose rate (D), annual effective dose equivalent (AEDE), activity concentration index (I), and alpha index ($I_\alpha$). The results revealed activity concentrations above global average values and an AEDE exceeding the 1 mSv $y^{-1}$ public exposure threshold set by Italian legislation. While the activity concentration index also surpassed the recommended limit, the alpha index remained below the safety threshold, indicating a low risk from indoor radon exposure. These findings suggest that further radiological assessment is required before using *Viterbo tuff* in buildings with high occupancy.**
**Noteworthy, the study reported in this paper was developed in the framework of the PRIN 2022 PNRR ATHENA project, funded by the European Union - Next Generation EU.**

## I. INTRODUCTION

Natural background radiation is responsible for almost all the global population's external dose; $^{238}$U, $^{232}$Th, and $^{40}$K radionuclides can be found naturally in soil, sand, rocks, water, building materials, and so on [1]. The distribution and abundance of these radioisotopes are subject to variation according to the geology of each region on a global scale. In detail, different types of rocks contain traces of uranium and/or radium, ranging from 2–5 parts per million (background) to 1000 parts per million (black shales) [2]. In the case of igneous rocks, specifically, crystalline rocks such as granites, granitic pegmatites, and syenites are more likely to contain uranium [3]. Most of the uranium/radium concentration in these rocks is found in accessory minerals like zircon, monazite, allanite, and sphene [4]. With regard to the exhalation of $^{222}$Rn from a given rock surface, it is worth noting that it is strongly influenced by the petrographic and petro-physical characteristics of the rock itself (i.e., micro-fissures, grain size, arrangement, alteration degree, and contact surfaces between constituents), even though measurements of U/Ra activity can provide useful information about radon release potential [5].

Going on, building materials can release gamma radiation in both indoor and outdoor environments, and extended exposure to low levels of ionizing radiation can impair human health [6]. Since individuals spend the majority of their time indoors, it is imperative to assess the concentration of ionizing radiation emitters in natural building materials (BM) used in residential constructions [6]. Since 1989, the European Union (EU) has produced a series of regulations and directives aimed at limiting the radiation generated by building materials while considering the potential health effects of radiation exposure in indoor environments [7]. In particular, in 1999 the European Commission (EC) announced the ALARA (As Low As Reasonably Achievable) principle, which established radiation exposure limitation from substances containing high levels of naturally occurring radionuclides as the ultimate goal of radiation control in building materials [8-9] To achieve this goal, the EC first introduced the radiation activity concentration index (I), which is commonly used as a screening tool to limit gamma radiation exposure of construction materials. Furthermore, various new indexes have been developed over time to assess the radiological hazard associated with radiation exposure from such samples [7]. More recently, the EU has carefully considered radiological concerns affecting public health in the European Directive 2013/59 EURATOM, which has been translated into Italian legislation as D.Lgs. 101/2020 and its successive modifications [10].

In this context, the PRIN 2022 PNRR ATHENA (A novel approach to the management of building materials of particular historical-artistic interest: assessment of radon exhalation and radiological risk due to natural radioactivity content) project, funded by the European Union - Next Generation EU, provides the context for the development of this paper, where we assess and report the radon exhalation from the *Viterbo tuff* natural stone, largely employed as building material of particular historical and artistic interest, by adopting the Closed Chamber Method (CCM) using the experimental setup of the Durridge Real-time Continuous Radon Monitor Rad7 for the short-lived radon progeny alpha spectrometry. Moreover, its natural radioactivity content was evaluated by using the High Purity Germanium (HPGe) gamma spectrometry technique, and the radiological hazard for humans was then assessed through the calculation of the absorbed gamma dose rate (D), the annual effective dose equivalent (AEDE), the activity concentration index (I), and the alpha index ($I_\alpha$).

## II. MATERIALS AND METHODS

### *A. Sample description*

The *Viterbo Tuff* formation is massive and homogeneous, its colour is from yellow-reddish to dark grey. Large fragments of black porphyritic leucite/analcime and sanidine scoria are immersed in a coarse cinder matrix. Among phenocrystals other than weathered leucite and sanidine, clinopyroxene, plagioclase and biotite with accessory minerals represented by oxides of iron and titanium and sphene are present. Inclusions of lava (leucitite) of various dimensions are distributed irregularly [11]. Rock chemistry varies between phonolitic tephrite and phonolite. The ignimbrite matrix is strongly zeolitized, but studies focusing on the inherent zeolite content are quite scarce. The rock is strongly zeolitized to chabazite and to a lesser extent to phillipsite with a total zeolite content ranging between 20% and 70% [11]. The petrographic and mineralogical characterization conducted by [11] indicate a very high degree of homogeneity of the formation and an almost total lack of zoning in the vertical sense. This bed shows a chaotic structure and a matrix texture of a coarse-grained cineritic type with a reddish coloration and an occasional tendency to yellow or intense red, but more frequently tending to grey-black. Paragenesis of the phenocrystals seems to be obvious: analcime and pyroxene are present in virtually all the outcrops, whilst mica is not always observed.

The mechanical properties are good, although porosity is high, in fact this tuff is used generally in construction and paving.

### *B. Methods*

Five aliquots of the investigated *Viterbo tuff* natural stone were obtained in laboratory by cutting larger pieces of stones by means of a circular saw. Each aliquot consisted in a cube with a side of approximately 5 cm.

The radon exhalation rate from each aliquot of the investigated natural stone was assessed by employing the Closed Chamber Method (CCM) [12,13]. In particular, the experimental apparatus comprises a small cylindrical steel vessel (Closed Chamber) (volume 2.75 L) containing the sample under investigation and connected to the Durridge Real-time Continuous Radon Monitor Rad7, by means of vinyl tubes and a Laboratory Drying Unit (LDU) containing a desiccant ($CaSO_4$), forming a closed circuit experimental set-up [13]. From the Closed Chamber the air flows, drawn by a mechanical pump internal to the monitoring instrument, into the instrument ionization chamber where Radon's and Radon progeny's alpha decays are detected, passing first through the LDU and an inlet filter (capable to avoid the unwanted suction of dust and radon's progeny). Subsequently the air is returned to the stainless-steel chamber through an outlet filter (output), thus forming a closed loop. As $^{222}Rn$ containing in the filtered air decays inside in the instrument ionization chamber, it produces alpha-emitting progeny, particularly polonium isotopes, which can be detected. A high voltage of 2500 V is applied between the ionizations chamber walls and a solid-state silicon detector, where the detected alpha radiations convert directly to electrical signals, enabling to differentiate between the electrical pulses generated by α-particles and those generated by $^{218}$Po, $^{216}$Po, $^{214}$Po and $^{212}$Po, with energies of 6 MeV, 6.7 MeV, 7.7 MeV and 8.8 MeV, respectively. Exploiting the secular equilibrium between polonium and radon nuclei this approach enables the use of the $^{218}$Po peak for the assessment of the $^{222}$Rn activity concentration in a very reasonable measurement time of 30 minutes [14].

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 [13]:

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

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). In order to minimise the leakage from the chamber, an insulating rubber was utilised.

Prior to analysis by the HPGe gamma spectrometry technique, each aliquot was first dried, in order to completely remove the moisture and to obtain constant mass. Then, it was inserted into a Marinelli hermetically sealed container of 1 L capacity and left to rest for a period of 40 days, in order to reach the secular radioactive equilibrium between $^{226}$Ra and its daughter products. After that, the specific activity of $^{226}$Ra was quantified [15].

Samples were counted for 70000 s and, to assess the $^{226}$Ra specific activity, the 295.21 keV and 351.92 keV $^{214}$Pb and 1120.29 keV $^{214}$Bi gamma-ray lines were used. Moreover, the $^{232}$Th activity concentration was determined by using the 911.21 and 968.97 keV $^{228}$Ac gamma-ray lines. Finally, regarding $^{40}$K, the evaluation was performed from its γ-line at 1460.8 keV.

Going on, the experimental setup was composed by a positive biased Ortec HPGe detector (GEM) [15], located inside lead wells to shield the environmental background radioactivity. For efficiency and energy settings, a multi-peak Marinelli γ-source (AK-5901) of 1 L capacity, energy range 60-1836 keV, custom made to replicate the exact designs of the specimens in a water-equivalent epoxy resin matrix, was employed. The Gamma Vision (Ortec) software was used for data acquisition and analysis [16].

The activity concentration (Bq $kg^{-1}$ dry weight, d.w.) of the investigated radioisotopes was calculated as follows [17]:

$$C = \frac{N_E}{\varepsilon_E t \gamma_d M} \quad (2)$$

where $N_E$ is the net area of a peak at energy E, $\varepsilon_E$ and $\gamma_d$ 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) [18]. The γ-ray spectrometry experimental results have been obtained according the protocol issued by the International Standards Organization (ISO) in the specific guidance ISO 19017:2015 [19]. The Italian Accreditation Body (ACCREDIA) certified the quality of the γ-ray spectrometry experimental results [20], thus ensuring continuous verification that the performance properties of the method are preserved.

It is worth noting that, in order to estimate the radiological hazard for humans, the absorbed gamma dose rate (D), the annual effective dose equivalent (AEDE), the activity concentration index (I), and the alpha index ($I_\alpha$)., were calculated. In particular, the absorbed gamma dose rate, D (nGy $h^{-1}$), was calculated using the standard room model, as previously stated in [21]:

$$D = 0.92 C_{Ra} + 1.1 C_{Th} + 0.08 C_K \quad (3)$$

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 sample, respectively.

Assuming an employment factor 80% for indoor exposure, the annual effective dose equivalent (AEDE) (mSv $y^{-1}$) of an individual may be calculated as follows [21]:

$$AEDE = (D-50) \times 8760 \text{ h} \times 0.7 \text{ Sv Gy}^{-1} \times 0.8 \times 10^{-6} \quad (4)$$

where the average dose rate value of 50 nGy $h^{-1}$ for the background was discounted. In order 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 [21]:

$$I = C_{Ra}/300 + C_{Th}/200 + C_K/3000 \quad (5)$$

This index refers to the dose from γ-radiation present in a building built by means of a given construction material, in excess of the typical external exposure. It should not be more than 1 for the radiation hazard to be neglectable [10].

The alpha index was calculated with the following formula [21]:

$$I_\alpha = C_{Ra}/200 \quad (6)$$

that enables the assessment of the alpha radiation exposure to the indoor radon exhaled from construction materials. The activity concentration of $^{226}$Ra must be lower than 200 Bq $kg^{-1}$, to prevent exposure to indoor radon specific activity higher than the threshold value of 200 Bq $m^{-3}$ [22], and then $I_\alpha$ must be less than unity for the hazard of exposure to radiation to be minimal [22].

## III. RESULTS AND DISCUSSION

The average $^{222}Rn$ exhalation rate, together with the $^{226}Ra$, $^{232}Th$ and $^{40}K$ activity concentrations (the mean value of the five analyzed aliquots), for the analyzed natural stone, are reported in Table 1.

*Table 1. The average $^{222}Rn$ exhalation rate, together with the $^{226}Ra$, $^{232}Th$ and $^{40}K$ activity concentrations, for the analyzed natural stone.*

| **Sample** | **$^{222}$Rn (Bq h$^{-1}$ kg$^{-1}$)** | **$^{226}$Ra (Bq kg$^{-1}$ d.w.)** | **$^{232}$Th (Bq kg$^{-1}$ d.w.)** | **$^{40}$K (Bq kg$^{-1}$ d.w.)** |
|---|---|---|---|---|
| *Viterbo tuff* | 0.755 ± 0.263 | 125 ± 19 | 302 ± 35 | 2021 ± 251 |

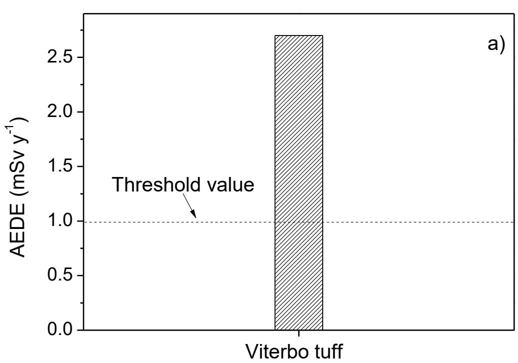
It is important to note that the specific activities of $^{226}Ra$, $^{232}Th$ and $^{40}K$, are significantly higher than the average world values [1].

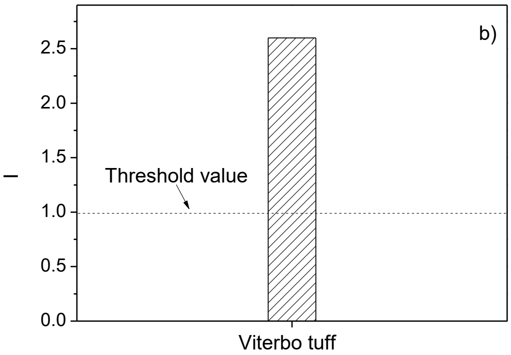
Published research indicates that the chemical composition and mineralogical characteristics of natural stones exert a considerable influence on the values of the $^{222}Rn$ exhalation rate, as well as those of $C_{Ra}$, $C_{Th}$, and $C_K$ [23]. In the light of these findings, further investigations have been planned to ascertain the chemical and mineralogical composition of the *Viterbo tuff* stone, by using X-ray diffraction and micro-Raman scattering techniques.

The absorbed gamma dose rate (D), the annual effective dose equivalent (AEDE), the activity concentration index (I) and the alpha index ($I_\alpha$), as calculated using the equations provided in (3–6), are reported in Table 2. Moreover, the last three indices are shown in Figure 1, together with recommended safety thresholds.

*Table 2. The calculated values of the radiological hazard indices for the investigated sample.*

| **Sample** | **D (nGy h$^{-1}$)** | **AEDE (mSv y$^{-1}$)** | **I** | **$I_\alpha$** |
|---|---|---|---|---|
| *Viterbo tuff* | 608 | 2.7 | 2.6 | 0.6 |



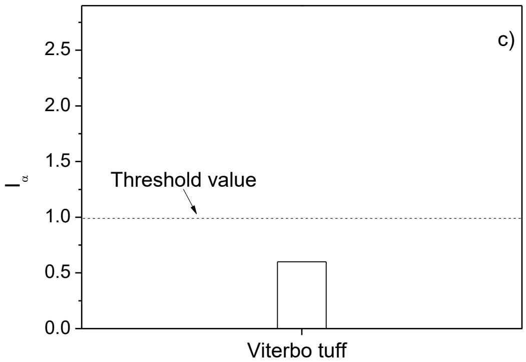




*Fig. 1. The annual effective dose equivalent (AEDE) (a), the activity concentration index (I) (b), and the alpha index ($I_\alpha$) (c) for the investigated natural stone, together with recommended safety thresholds.*

In particular, the D value of 608 nGy h$^{-1}$ can be attributed to the lithological component of the sample under investigation [24] and was used to calculate AEDE through the equation (4).

Noteworthy, in the extremely precautionary scenario assumed (8760 hours of exposure, i.e. 24 hours a day for 365 days), the latter was found to be 2.7 mSv y$^{-1}$, which is higher than the action level of 1 mSv y$^{-1}$ reported by the Italian legislation for members of the population [10]. Furthermore, the I index was found to be 2.6, higher than the threshold value of 1. These results impose, for the purposes of using this natural stone for civil engineering buildings, such as dwellings and buildings with a high occupancy factor, a more accurate assessment of the dose in accordance with Article 29, paragraph 5, of D.Lgs. 101/20 [10], applying dose estimation methods provided by national and international standards that take into account other factors, including density, material thickness, as well as factors relating to the type of building and the intended use of the material (structural or surface) [18, 25].

Finally, $I_\alpha$ was turned out to be 0.6, which is below the threshold value of 1, thus rationally ruling out any significant health impact from exposure to radon gas exhaled by the investigated natural stone.

## IV. CONCLUSIONS

Radon exhalation rate and natural radioactivity content of a natural stone of particular historical and artistic interest employed as building material, i.e. the *Viterbo tuff* stone, were investigated by using the Closed Chamber Method (CCM), applied by means of the experimental setup of the Durridge Real-time Continuous Radon Monitor Rad7 for the short-lived radon progeny alpha spectrometry and the High Purity Germanium (HPGe) gamma-ray spectrometry technique.

In particular, the specific activity concentrations of $^{226}$Ra, $^{232}$Th and $^{40}$K, were found to be higher than the average global values. Furthermore, to evaluate the radiological hazard associated with radiation exposure from the investigated specimen, the absorbed gamma dose rate, the annual effective dose equivalent, the activity concentration index, and the alpha index were estimated. In detail, in the extremely precautionary scenario assumed, the AEDE value was found to be higher than the action threshold specified by Italian legislation for the public, namely 1 mSv $y^{-1}$. Moreover, the I index was found to be higher than the threshold value of 1. These results impose, for the purposes of using this natural stone for civil engineering buildings, such as dwellings and buildings with a high occupancy factor, a more accurate assessment of the dose in accordance with Article 29, paragraph 5, of D.Lgs. 101/20. Finally, it was verified that $I_\alpha$ was less than unity, indicating that the radiological hazard associated with exposure to indoor radon concentrations exceeding 200 Bq $m^{-3}$ is extremely low.

**Funding:** This work was performed in the framework of the 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) project, 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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