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Radiation protection considerations on radon and building materials radioactivity in Near Zero Energy Buildings

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Abstract

Recent updates of the E.U. Basic Safety Standards, stated in the Council Directive 2013/59/EURATOM, are focusing on risks related to radon gas concentration inside dwellings and working places, as well as radioactivity of building materials. In particular, the new E.U. Basic Safety Standards are based on last recommendations from the International Commission on Radiological Protection (ICRP), and from the World Health Organization (WHO), which consider that radon issues, and external irradiation from building material, as topic aspects to population's health. Further, ICRP Publication 126, by using bio-kinetics models for estimating the effects of radon intakes, has drastically reduced the reference level for radon concentration in dwellings and working places.

Radon issues have recently gained particular attention due to current orientations in constructing buildings with energy consumptions lower and lower. Radon gas emerges from the ground, penetrates building's basements, and accumulates itself into the indoor air, being breathed by people.

Taking care of windows' airtightness allows the radon concentration to build up, in some cases beyond reference levels, together with other chemical pollutants, i.e. combustion residues and solvents.

On considering that Council Directive 2013/59 EURATOM has to be transposed into law by each EU Member State by February 2018, it is recommended that radon issues have to be considered during the design phase of the building construction, particularly for NZEB applications. Further, external irradiation from building materials, i.e. tuff, marbles, tiles, pozzolana, coal ashes and so on, may be a reason of concern also.

This paper describes radiation protection issues focusing on public and domestic environments, where people are supposed to spend a considerable amount of time. About radon, real measurements are shown, both in domestic and working scenarios. Dealing with external irradiation due to building materials, calculations and simulations have been performed and results are presented.

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1. Introduction

Today, energy production by renewable energies, low-carbon-emitting energy sources, together with energy savings, appear to be some key-approaches to a more sustainable future of the Earth planet.

Such efforts can theoretically be applied at every level, from a large industrial sector (e.g. electricity production, manufacturing installation, etc.) dealing with few plants that manage large quantities of energy, till people's dwellings, dealing with many customers managing a very small amount each.

Only in the last decade, such a culture has begun to be transferred in buildings industry also, developing new construction trends that include, starting from the design phase, solar panels for hot water or electricity production, wind generators, shadowing systems and optimized orientation to sun, thermal insulation materials, and technologies with improved performances, etc. All those strategies to minimize building's consumption and energy losses while maximizing its energy production, giving finally a net balance near to zero.

In such a contest, particular interest assumes the aspect of the indoor air quality: current trends tend to care about windows and doors' sealing, rendering the internal environment an air-tight system. In such a way, if a periodical air renewal (natural or artificial) is not present, internal air pollutants may build-up to concentrations-in-air that could be reason of concern to people's health.

Alongside chemical pollutants (e.g. combustion residues from kitchen fires or fireplaces, solvents from paints and finishing, etc.), radon gas plays the role of radioactive pollutant, being a noble gas that emerges from the ground or building materials, easily diffusing in structures and accumulating in closed environments. According the World Health Organization, WHO, radon is the second cause of lung cancer to the general population, being cigarettes smoking the first one. Epidemiological studies have provided evidence of an association between indoor radon exposure and lung cancer, even at the relatively low concentration levels commonly found in residential buildings [1]. The International Commission on Radiological Protection, ICRP, revised the risk assessment about radon in [2], confirming WHO concerns about residential exposure for the public in domestic environment. Such considerations were merged in the EU 2013/59 Euratom Council Directive, that introduced new recommendations to be transposed into law by each E.U. Member State by February 2018. In particular, the Directive introduces lower reference levels for radon concentration in air, and ratify the need for radon control not only for workplaces but for dwellings also. Moreover, the Directive introduces reference levels for gamma radiation emission from building materials.

Radon gas and typical materials used in constructions (tuff, marbles, granitoides, porphyries, pozzolana, glaze and stoneware tiles, bricks, etc.) are defined as Natural Occurring Radioactive Materials –NORM. When dealing with exposure to such radiation sources, special evaluations about delayed-in-time health effects are to be carried on. The current Italian law (D. Lgs. 230/95 s.m.i.) considers a radon annual-averaged concentration-in-air of 500 Bq/m³ for working places as a reference value, resulting in an effective dose value of ~3 mSv/a (considering a conversion factor from concentration-in-air to effective dose of $3 \cdot 10^{-6}$ mSv/h/(Bq/m³) [3] and a residence time of 2000 h/a in working spaces). It should be noted that, currently, residential dwellings are not accounted for. From [4], the reference level for radon indoor concentration, including both working and residential, shall be lowered from 500 Bq/m³ to not more than 300 Bq/m³; in the meantime, a reference level for external exposure to gamma radiation emitted by building materials, including both indoor and outdoor contributions, is introduced and set at 1 mSv per year.

If the latest issue appears fairly new, radon pollution is known by a long time. In 1988, the Italian ANPA (the former ISPRA, The Italian National Institute for Environmental Protection and Research) and ISS (The Italian National Institute for Health) realized a survey campaign about radon indoor concentration in a sample of 5000 Italian dwellings. Such a campaign was a first screening only, because a sample of 5000 dwellings is surely not representative. Anyway, results reported in [5] have shown interesting reasons of concern: 1) radon issues are strongly related to local peculiarities (soil origin and constitution, microclimate conditions [6]) and building

materials used; 2) radon benefits of air draught by windows, and a very small air-exchange with the external environment, typical of the old-fashioned wooden windows, could mitigate significantly radon build-up at indoor locations [7]; 3) data should be updated to the current situation of the Italian dwellings: experimental evidences by authors' works show that doors and windows finished in sealing could appreciably increase radon indoor concentration-in-air.

Now, due to extreme local variation of radon concentrations in the whole national territory, radon measurement currently is institutionally carried out by regional agencies (ARPAs).

At Dept. of Basic and Applied Sciences for Engineering in Sapienza – University of Rome, the Laboratory for Radiation Protection is endowed with the most updated instrumentation and calculation tools for managing all radiation exposures, including situations in domestic environments as depicted before. In this paper, a real case scenario of a country house in Viterbo is presented, showing radon measurements in normal conditions, and repeated measurements when ventilation or air-tight sealings are artificially introduced [8]. A second case study about radon examines a working location where radon concentration is widely varying in time, showing an innovative solving-approach to preserve both people's health and energy consumption.

Moreover, an operative evaluation of gamma exposure by the most utilized building materials is presented, and a gamma spectrometry characterization of a set of 9 different sands for concrete preparation is shown.

Results here presented are compared to reference levels by [4]. Consequent considerations could be taken as first indications when approaching radiation exposure situations in dwellings, giving some hints to recognize all scenarios that may need actions or suggestions by a Radiation Protection Expert. (RPE – in Italy called “Esperto Qualificato”).

2. Case study #1: radon in a country house in Soriano (VT, Italy)

The municipality of Soriano is set at the hillside of the Cimino Mountain, between the Tyrrhenian Sea and the Central Apennine mountains. During the Pleistocene Age, the Volsino, Vicano and Cimino explosive-effusive volcanism gave origin to the present orographic layout and soil composition. Particularly, the subsoil stratigraphy is characterized by large presence of tuff, lava, lapillus layers and pumices [9].

Volcanic materials are characterized by high contents in natural radioactive elements, and then the country house analyzed in such paragraph resides on a land characterized by high radiation background. Considering that rural dwellings are usually built with local materials, the main construction type in Viterbo uses tuff and bearing walls, with the result of increasing radiological issues about radon build-up and gamma irradiation at indoor locations.

The country house here presented is organized in three levels, i.e. basement, ground floor, and first floor. The planimetry layout is presented in Figure 1, showing radon-222 concentration-in-air averaged on the overall measurement periods, without any change in the normal usage of spaces. All specifications about this first set of measurement are reported in Table 1.

The current Italian law (D.Lgs. 230/95 s.m.i.) is relying on radon annual-averaged concentration in air, usually measured with passive instruments (i.e. CR-39) in times ranging from 6 to 12 months.

The measurements registered in the current study exploited the Genitron AlphaGuard available at Laboratory of Radiation Protection in Sapienza. Such radon detector is an active instrumentation capable in measuring accurately ^{222}Rn concentration hourly, giving also concentration trends vs. time.

From measurements results and considering residence times, ^{222}Rn concentration can be easily converted to annual effective doses, by means of the conversion factor given above, see Table 1. Summing all contribution, and considering a person living in the house and sleeping in ‘Bedroom 1’, the annual effective dose conferred by radon inhalation in domestic dwelling only results in ~10 mSv. However, it should be noted that such evaluation is based on a short-term measurement (about 50 hours) by an active instrument. Radon can vary significantly vs. time: it could make the measurement period carried out not representative of the annual averaged concentration on which dosimetric evaluations should be based. To get a fast evaluation, a short-term measurement could be accepted considering a 40% uncertainty, as suggested in [10].

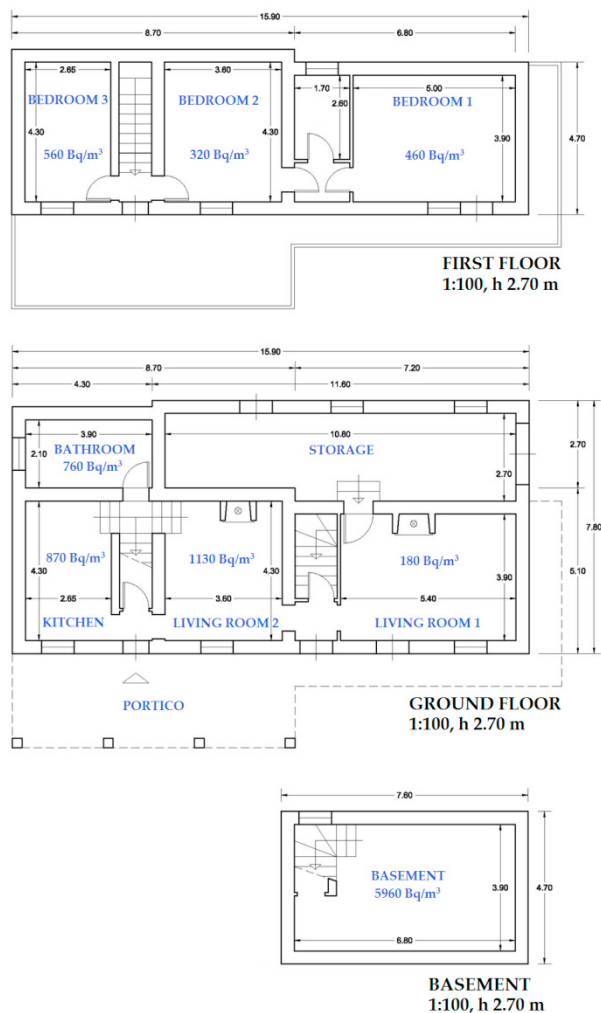


Figure 1 – Planimetry of the country house where radon measurement were taken. Each room analyzed reports the ^{222}Rn concentration value averaged on the measurement period at the normal boundary condition.

Table 1 – Specifications of radon measurements in spaces in Figure 1, with a comprehensive evaluation of residence times per space and annual effective doses from radon inhalation, considering a conversion factor of $3 \cdot 10^{-6} \text{ mSv/h}/(\text{Bq/m}^3)$.

Space label	measurement period [h]	^{222}Rn concentration [Bq/m^3]			residence time [h/a]	Effective dose, E [mSv/a]
		min.	max.	avg.		
Basement	49	428	10496	5960	60	1.07
Living room 1	49	69	324	180	1400	0.76
Living room 2	48	616	1704	1130	700	2.37
Kitchen	48	268	1608	870	400	1.04
Bathroom	48	406	6432	760	300	0.68
Bedroom 1	144	118	1923	460	3000	4.14
Bedroom 2	208	53	752	320	3000	2.88
Bedroom 3	180	111	1232	560	3000	5.04

For allowing to appreciate the order of magnitude of annual effective dose values, it can be remembered that workers in nuclear industry are considered ‘exposed to radiation field for working purposes’ because they are susceptible to receive more than 1 mSv per year, being 20 mSv the upper limit that shouldn’t be exceeded. It is evident that, even exposed to the natural radioactive background, the person living in the country house analyzed is subjected to excessive exposure values, which claims that some mitigation action should be put in place.

2.1. Case study #1: simulation of air-tight doors and windows

As discussed before, a simulation of substitution of the current doors and windows with more efficient air-tight solution has been implemented by an artificial sealing with plastic and paper scotch, Figure 2:

- in the first phase, doors and windows in Living Room 1 have been sealed and radon concentrations measured;
- in the second phase, doors and windows in Basement have been sealed and radon concentrations measured.

Results reported in Table 2 show that introducing more efficient doors and windows for energy saving purposes affect significantly radon build-up in spaces. The most significant case is given by ‘Living Room 1’ where values in normal condition were acceptable, while the action of sealing causes the same space to be of radiological concern if the energy-saving intervention of windows’ substitution is put in place.



Figure 2 – Doors and windows sealing by plastic and paper scotch, simulating air-tight doors and windows.

Table 2 – Comparison of radon measurements in normal conditions vs. doors and windows sealed conditions. Air-tights play a significant differences in radon build-up dynamics. Effective doses are computed from averaged radon concentrations (avg. values), taking the following residence times: 60 h/a for the basement, 1400 h/a for Living Room 1.

Space label	normal conditions				sealed conditions			
	²²² Rn concentration [Bq/m ³]			E [mSv/a]	²²² Rn concentration [Bq/m ³]			E [mSv/a]
	min.	max.	avg.	(ref. avg.)	min.	max.	avg.	(ref. avg.)
Basement	428	10496	5960	1.07	7700	36000	19230	3.45
Living room 1	69	324	180	0.76	392	5856	1290	5.45

2.2. Case study #1: simulation of natural ventilation

Mitigation actions against radon indoor build-up should be put in place when measured values overstep acceptable limits. In existing buildings, it is possible to seal ground foundation to limits radon produced in subsoil to enter internal spaces due to the pressure gradient. Alternatively, it is possible to create also preferential path (via an external fan mounted near ground foundations) to extract radon from the soil underneath the dwelling, avoiding its penetration into internal spaces.

If radon is produced also by building material, the interventions just discussed could be insufficient and the only way to mitigate radon concentration is to renew the indoor air by exchanges with outdoor air.

In the basement of the house analyzed, a window for limiting internal humidity and mould formations is already present; in normal conditions the window is closed. As to realize an air exchange with external air, such window is now opened, instituting a natural ventilation.

Results reported in Table 3 show that such a simple intervention is capable in mitigating radon concentration significantly. It is clear that such an action could harm the energy consumption of the dwelling if the space that needs radon dilution is also conditioned in heating or cooling. In the next case study, an idea to solve such an issue is proposed.

Table 3 – Comparison of radon measurements in normal conditions vs. natural ventilation conditions. Indoor air exchanges with outdoor air mitigate radon concentration significantly. Effective doses are computed from averaged radon concentrations (avg. values), taking the following residence times: 60 h/a for the basement.

Space label	normal conditions				vented conditions			
	²²² Rn concentration [Bq/m ³]			E [mSv/a]	²²² Rn concentration [Bq/m ³]			E [mSv/a]
	min.	max.	avg.	(ref. avg.)	min.	max.	avg.	(ref. avg.)
Basement	428	10496	5960	1.07	31	800	333	0.06

3. Case study #2: radon in a workplace at Via Scarpa (RM, Italy)

A workplace case is here presented, showing other aspects on radon issues.

The space analyzed is the building where Laboratory of Radiation Protection at Dept. of Basic and Applied Sciences in Sapienza – University of Rome resides. It belongs to the Via Scarpa district that is laying on a very particular subsoil situation: the Saint Hippolytus catacomb of the Roman age. The subsoil composition is mainly constituted by tuff, and the presence of an underground tunnels grid on 5 levels causes a large air volume where radon is free to build-up, creating a huge radon-lung.

Since 2000, when the Laboratory for Radiation Protection was endowed with the AlphaGuard system for ²²²Rn active measurements, the presence of concentrations exceeding acceptable values were found (concentrations in the order of ~2000 Bq/m³, [11]), especially in correspondence of two rooms of a building laying on the catacomb main duct. From then on, radon mapping in the area was subjected to periodical re-evaluations, as to monitor feasible variations.

Figure 3 reports results from the last survey campaign with passive detectors (CR-39), from January 2012 till present days. As can be easily seen, radon concentrations are strongly variable vs. time, due to subsoil modifications, or earthquakes effects, affecting the preferential paths for radon escaping from the catacomb. In 2000 a certain building was believed as critical; today major risks for radon exposure of workers are found into another structure. Many values overcome the current reference level of 500 Bq/m³, requesting correction actions to prevent radon build-up at indoor location subjected of human attending.

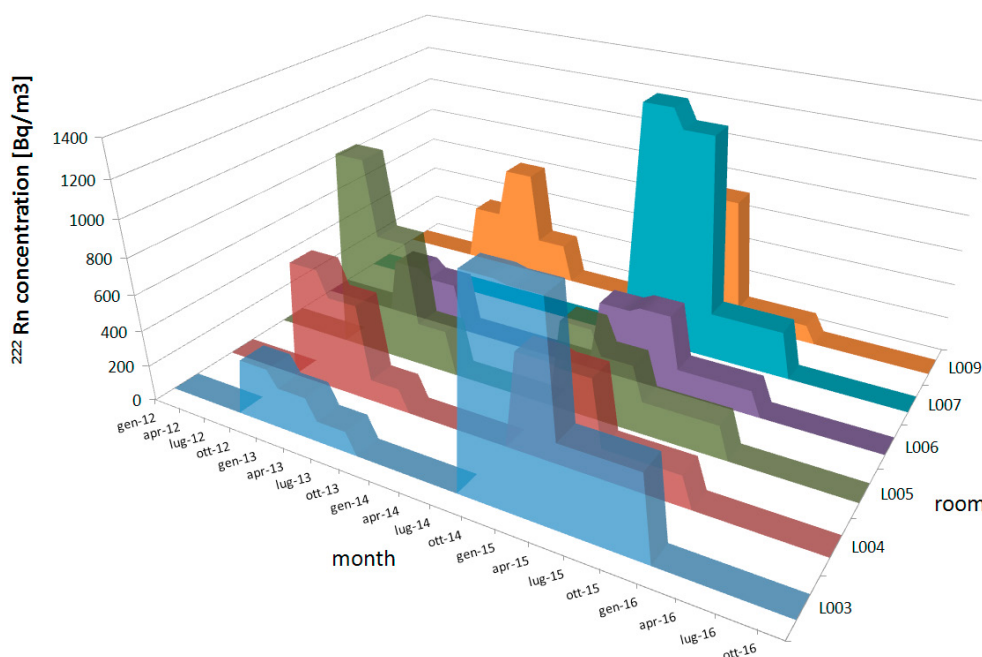


Figure 3 – Radon concentrations vs. time in some rooms of the building where the Laboratory for Radiation Protection is located, at Via Scarpa, RM. Measurements were taken with CR-39 films periodically installed and read (data courtesy of the Qualified Expert in Sapienza). Zero values correspond to periods where detectors were not placed in measurement.

Here, an active mitigation action, with an external fan to drain radon underneath foundations, or an artificial ventilation of spaces to dilute indoor radon with external air, could be inconvenient if operated continuously, without being scaled to the “entity” on radon build-up.

It is also known that indoor radon is subjected to day-night dynamics. In other words, it accumulates during the night, when doors and windows are closed, and it is diluted in the first hours of the day, when air-exchanges with outdoor air are promoted by anthropic activities.

In such scenarios, an active mitigation system, operating only when radon concentration overcome a threshold limit, could be helpful in containing the energy consumption about the electrical machinery for artificial ventilation. As to command the operation of such a “smart” system, a cheap, reliable and fast ^{222}Rn sensor (to be integrated in the dwelling automations) is currently under development at Laboratory of Radiation Protection in Sapienza.

4. Case study #3: gamma irradiation from building materials

As introduced before, building materials could carry significant amounts of radioactivity due to their origin. Tuff, marbles, granitoides, porphyries are naturally radioactive (NORM) while tiles, bricks, sands could be artificially enriched in radioactivity by anthropical activities that concentrate some radionuclides in products used, later, in manufacturing industries.

When radioactivity concentration in a building material overcomes certain values, the material itself could deserve radiological attention, as to evaluate its effect on people’s health. Because of that, Annex VIII in [4] provides a screening procedure easy to be applied as to determine if a building material has to be considered of radiological concern. i.e. the definition of the Activity Concentration Index, I,

$$I = \frac{C_{^{226}\text{Ra}} [\text{Bq/kg}]}{300} + \frac{C_{^{232}\text{Th}} [\text{Bq/kg}]}{200} + \frac{C_{^{40}\text{K}} [\text{Bq/kg}]}{3000}$$

where $C_{226\text{Ra}}$, $C_{232\text{Th}}$, and $C_{40\text{K}}$ are the activity concentrations in Bq/kg of the corresponding radionuclides in the building material. The Activity Concentration Index value of 1 can be used as a conservative screening tool for identifying materials that may cause the overcoming of the reference level for external exposure to gamma radiation emitted by building materials (including both indoor and outdoor contributions), set at 1 mSv/a. If the Activity Concentration Index overcomes the value of 1, special consideration by a radiation expert may be needed.

Table 4 – Radiological contents in $C_{226\text{Ra}}$, $C_{232\text{Th}}$, and $C_{40\text{K}}$ of typical building materials. For each, the Activity Concentration Index is provided, as to give an immediate idea of the corresponding radiological concern.

	Radiological content [Bq/kg]			Activity Concentration Index
	^{226}Ra	^{232}Th	^{40}K	
Concrete: standard [12]	33	45	420	0.48
Concrete: slightly above the average [12]	80	80	800	0.93
Concrete: increased in ^{226}Ra content [12]	1200	45	420	4.37
Bricks with high radioactivity content [12]	200	300	1500	2.67
Tiles [12]	1200	1500	1200	11.90
Tiles: Italian glazed	50	60	750	0.72
Tiles: Chinese glazed	70	60	600	0.73
Chinese glazing	500	600	500	4.83
Stoneware	200	60	0	0.97
Tuff (γ spectrometry in Laboratory)	380	288	1579	3.23
Sand #1 (γ spectrometry in Laboratory)	77	29	274	0.49
Sand #2 (γ spectrometry in Laboratory)	70	25	387	0.49
Sand #3 (γ spectrometry in Laboratory)	29	13	140	0.21
Sand #4 (γ spectrometry in Laboratory)	32	12	204	0.23
Sand #5 (γ spectrometry in Laboratory)	5	33	409	0.32
Sand #6 (γ spectrometry in Laboratory)	20	13	271	0.22
Sand #7 (γ spectrometry in Laboratory)	358	768	7993	7.70
Sand #8 (γ spectrometry in Laboratory)	114	71	1247	1.15
Sand #9 (γ spectrometry in Laboratory)	47	70	768	0.76

Table 4 reports the radiological content, in terms of concentration per kg of the radionuclide cited above, for typical building materials, together with the associated Activity Concentration Index. As can be seen, some of materials commonly used need radiological attention about their usage due to their capability in conferring dose to people, especially in domestic environment where people are supposed to spend a large amount of their life. When dealing with a material overcoming the value of 1 for its Activity Concentration Index, it is possible to apply different calculation approaches, as to determine the effective dose to people in the specific situation. Particularly, the simplified evaluation approach given by most updated method in the field is presented here [12], and the same “room situation” when analyzed by the authors with the Monte Carlo simulation code MCNPX [13].

Considering the approach presented in Annex VI of [12] at the Example n. 4, a room with size specified in Figure 4 with walls made in light-in-weight concrete (1000 kg/m^3 , as to simulate voids present inside a real wall construction) with increased ^{226}Ra content (Activity Concentration Index of 4.37 from Table IV) is considered. From [12], the effective dose rate at the room center is $0.117 \text{ }\mu\text{Sv/h}$; supposing a person residing in the room analyzed (as his own domestic dwelling) for 5000 h/a, the following annual effective dose is 0.584 mSv/a . Repeating the same study with a more efficient, customizable and generalized simulation instrument such as the Monte Carlo code for radiation transport MCNPX, results are slightly different: the effective dose rate at the room center is $0.151 \text{ }\mu\text{Sv/h}$; supposing a person residing in the room analyzed (as his own domestic dwelling) for 5000 h/a, the following annual effective dose is 0.841 mSv/a . Both values can be reason of concern considering that: 1) the reference level of 1 mSv/a by [4] considers both indoor and outdoor external irradiation, then others indoor contributions (e.g. working places), and outdoor contributions need to be computed case by case to complete the analysis; 2) a reference room more similar to Italian dwellings, instead of reference dimension in [12], could give more realistic results; 3) the simulation code used is completely customizable, and real situation can be reproduced with high-fidelity in details, giving more accurate results case by case.

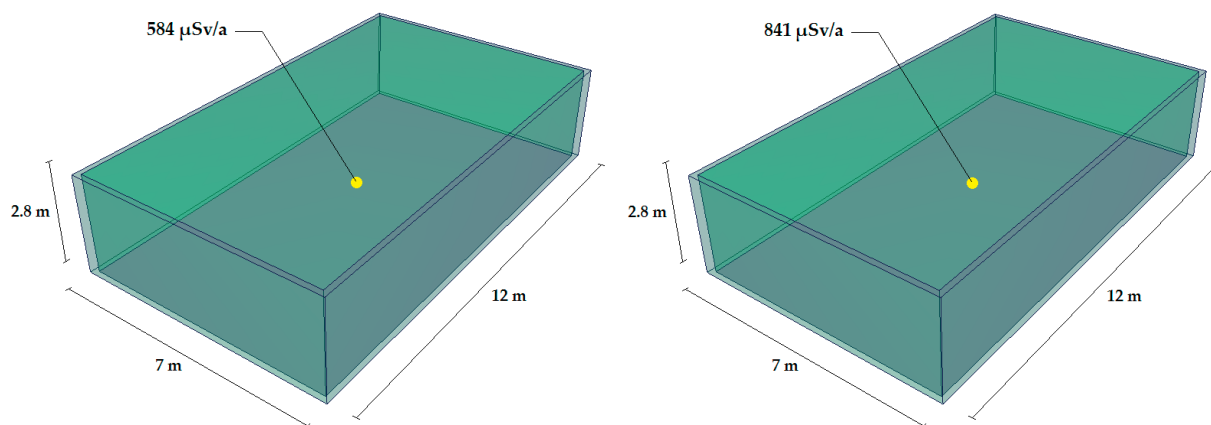


Figure 4 – Left: annual effective dose taken by a person residing 5000 h/a in the room analyzed in the Example 4 of Annex VI of [12] as his own domestic dwelling, following the analytical approach in [12]. Right: annual effective dose taken by a person residing in the same room, calculated via the Monte Carlo simulation code MCNPX by the authors.

5. Case study #3: gamma irradiation from building materials

The work here presented remarks the attention that radon gas and radioactivity in building materials deserve as ‘radioactive pollutants’ in both current Italian dwellings’ situation and new trends in buildings realization.

New perspectives by Council Directive 2013/59 EURATOM [4] and specialized Institutions, such as WHO [1] and ICRP [2], have shown that indoor locations for residential use need a specific radiological attention, due to the fact that natural radiations may harm the occupants when certain exposure limits are exceeded.

Three case studies are presented: 1) a country-house in Viterbo where high radon concentrations were measured and some mitigation actions experimented, 2) a workplace in Rome undergoing measured radon concentrations very variable vs. time; 3) a virtual scenario illustrating issues about gamma irradiation by building materials, once analyzed with an analytical model, once analyzed with a Monte Carlo simulation code.

Each situation is compared with current and foreseen effective dose limits or constraint by law, as to give a real touch on risks.

The authors intended to get the audience aware about the topics here faced, ensuring the intention to continue their information activity and on-field monitoring actions here shown.

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