Contribution of Metasilicates Containing TENORM to Radon Activity Concentration in Building Materials

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Contribution of metasilicates containing TENORM to radon activity concentration in building materials

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Abstract

Radon exhalation from soil and ores is among the most dangerous risks for the public health care. The impact becomes even more powerful when technological enhanced naturally occurring radioactive materials (TENORM) are used for public and private building. Here, we report the realization of a down-scaled model of a building, whose construction materials contain TENORM harvested in a site in Crotone (Italy). We observe an increase of the radon activity in the model when TENORM residues are employed. These results have then been compared to a real use case. The correspondence found between the values of radon activity concentration in the model and in the use case suggests that estimating the radon concentration is an useful method to target TENORM presence inside buildings.

Keywords: Radon activity, NORM and TENORM, Phosphorites, Indoor exposure, Crotone, Building materials

1 - Introduction

Radon is an inert natural gas whose detection finds application in several fields from geology (Noguchi 1977; Fleischer 1978; Dimova 2011) to public health care (Rutherford 1995) and (although controversially) in astrophysics (Pommé 2019). All these applications exploit the activity of the Radon-222 isotope (^{222}Rn) that belongs to the Uranium-238 (^{238}U) decay chain and has an half-life time of about 3.8 days. For this reason, ^{222}Rn is radioactive and therefore it is dangerous: ^{222}Rn activity has been shown to provide damages to health, attracting a worldwide interest of the scientific community (Lysandrou 2007; Yanchao 2020). A particular focus has been given by the European Commission to radon in the directive 59/2013 that reinforces the regulation about the population exposition to radioactivity (Euratom 2013).

The ^{222}Rn activity can be easily monitored in uranium mines, where the concentration may become remarkably high (Sahu 2013). Nevertheless, in the last decades, many studies have been carried out to estimate the impact of the ^{222}Rn exhalation over the ground from soil (Bossew 2003), building materials (Zhang 2012; Keller 2001) and NORM (Tufail 2010; Dentoni 2020), acronyms of Naturally Occurring Radioactive Materials. Among the latter, particularly interesting is the case of phosphorite rocks that are rich of ^{238}U and Thorium-232 (^{232}Th) (Menzel 1968). Their extraction and processing were the base for producing phosphoric acid, finding application in fertilizers, detergents, feeds, food additives and pesticides (Silva 2006; El Afifi 2009).

Processing NORM may results in the production of TENORM (Technological Enhanced Naturally Occurring Radioactive Materials) residues that, in the case of phosphorite rocks, contain naturally radioactive phosphogypsum and metasilicates (Nero 1984; Stoulos 2003).
Remarkable is the case of Crotone that since 1928 until the beginning of 1990 was one of the most important chemical industrial city in Europe, specialized in the treatment of phosphorite rocks, imported from Algeria and Morocco. For this reason, nowadays Crotone hosts in its territory residues of processed materials containing TENORM, enhancing the environmental radioactive level (Caridi 2017). Furthermore, over the years, because of their useful mechanical properties, these residues were used in the construction of public and private roads as well as closed spaces, such as dwellings, warehouses and public buildings. Thus, an important mass of phosphoric metasilicate is dangerously present all over the city, possibly determining an increase in the effective radiation dose for the population (Santagati 2016; Andresz 2019).
In this work, we propose a way to estimate the presence of TENORM in building by monitoring the indoor ^{222}Rn concentration. For this purpose, we built an in-scale model of a house including phosphorite residues harvested in the city as building materials. The effect of such residues on the environmental radioactivity has been monitored by means of both active and passive devices. We found a significant increase of the ^{222}Rn activity concentration inside the house model. The experimental values obtained in the model are compared to a real use case, i.e. an industrial warehouse where the employment of TENORM had been previously checked. A rather good agreement is found, indicating that ^{222}Rn can be used as a TENORM precursor to indirectly determine and to map the phosphorite contamination.

2 - Materials

The materials used in our experiments are phosphorite residues coming from a site targeted as a potential radioactive pole in Crotone: a square in front of a former high school (named Ciapi, whose exact location is reported in Fig.1a,b). Despite the amount of residues originally present in the city is unknown, a rough estimation of thousands of tons can be done by seeking the factories production history (Santagati 2016). Phosphorite TENORM are easily recognizable by their unique blue color (see the picture in Fig.1c).

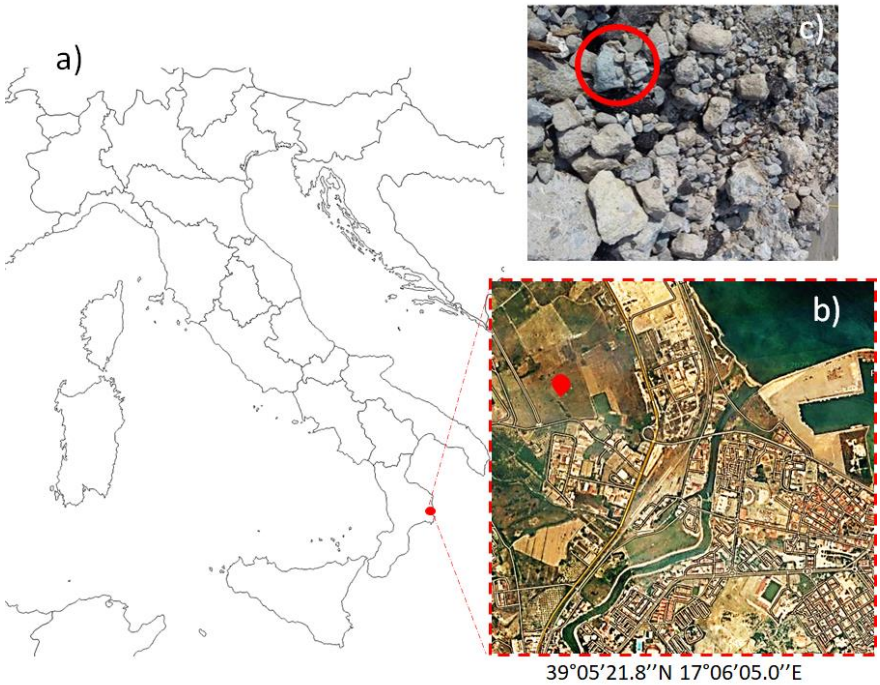
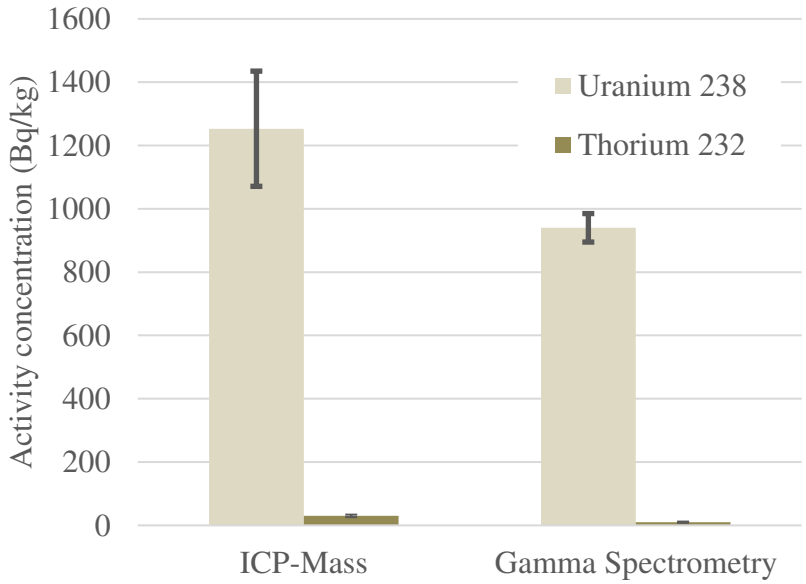


Figure 1:a) Crotone position in the Italian map. b) Harvesting site location. c) Picture of the phosphorite rocks showing their typical blue colour.

Tracing their historical origin, we found that such residues come from the industrial zone where mainly two chemical processes used to be developed: the extraction of phosphorus from sulphuric acid and the thermal production of elemental phosphorus. The first results in the production of phosphogypsum that, due to its flour-like nature, was not useful as construction material. Conversely, the second exploits a thermal reaction in blast furnaces between phosphorite and silica (SiO_2) using either iron or carbon (C) as catalysers (Erkens 1999):

$$2Ca_3(PO_4)_2 + 6SiO_2 + 10C \longrightarrow 6CaSiO_3 + 10CO + P_4. \quad (1)$$

The silica main role consists of extracting the calcium (Ca) from the calcium phosphate ($Ca_3(PO_4)_2$). At the same time, this insulates the elemental phosphorus giving a glassy nature to the metasilicates ($CaSiO_3$). These are the only solid residues of the reaction, since both the carbon oxide (CO) and elemental phosphorus (P_4) are gases. Reaction (1) is only one of the chemical process that take place during the thermal production of elemental phosphorus. Indeed, phosphorites are complex minerals, constituted of many chemical elements that are involved in the reaction (e.g., aluminium and lead) (Tayibi 2009). Among them, despite some naturally radioactive elements like the Polonium-210 are ejected during the process, other elements, such as ^{232}Th, ^{238}U, Radium-226 and some of lead isotopes, are possibly accumulated inside the metasilicates. This provides a natural radioactivity to the residues that are therefore labelled as TENORM (El Afifi 2006). As a first step of our study, we characterized the radiometric features of the residues, having a density ρ = 1220 kg⁄m^3. In Fig.2 we report the activity concentration of ^{238}U and ^{232}Th, measured both by ICP-Mass and γ-spectrometry. The results given by the two techniques are compatible: the ^{238}U contribution is about 1000 Bq/kg while that of ^{232}Th is two order of magnitude smaller. Coherently, one can expect an appreciable ^{222}Rn activity. Once characterized, these rocks were employed in the model building, as it will be discussed below.



	Activity concentration [Bq/kg]	
	ICP-Mass	γ-spectrometry
^{238}U	1253±182	940±45
^{232}Th	30±4	10±2

Figure 2: Uranium and Thorium activity concentration measured by ICP-Mass and γ-spectrometry.

3 - Model

Our model is a down-sized reproduction of a typical city dwelling, shown in Fig.3a. The house, having a 1.00 m x 1.00 m square area, was named Mary's House, being Mary the most popular name in the city. In Fig.3b, we show the model planimetry. A door, properly sealed during the measurements, was added to the square perimeter in order to access the inside volume where the experiments are carried out. Significant accuracy was spent on details such as the thickness of the walls, the relative air chamber, the interior and the exterior plasters. In particular, the wall thickness is about 10 cm in the foundations and 9 cm in the house so that the accessible area is 82 cm x 82 cm. The inside space is about 50 cm high without considering the roof and the foundation volumes, as depicted in Fig.3c. The figure also details the different materials employed. After interviewing building workers, we replicated the techniques used in the '80s and '90s in order to obtain a re-production as faithful as possible. According to these techniques, the residues were systematically used as an inert material for filling buildings crawlspaces and foundations. For sake of simplicity, in our model we employ phosphorite residues only to fill the crawlspace, being 7 cm deep. In this way, we could have a better estimation of the background activity as described in the following sections. Filling the crawlspace required a mass of (61 ± 1)kg, on top of which a 3 cm thick screed of cement mortar has been made (see the different heights reported in Fig.3c and 3d). Conversely, we made the foundation of reinforced concrete (with a specific weight of 23.5 kN⁄m^3) on a raft of the same material. The curtain walls are made of forated bricks (with a specific weight of 8 kN⁄m^3), vertically arranged in order to increase the volume of the air present inside the walls, whereas the roof slab is constituted of a flooring blocksand and a cement mortar screed. After plastering, the whole house was painted both internally and externally with washable paint. The plaster has been made as thin as possible in order to avoid possible ^{222}Rn absorption. Finally, consisting of two layers of corrugated sheet, the roof has only an aesthetic function.

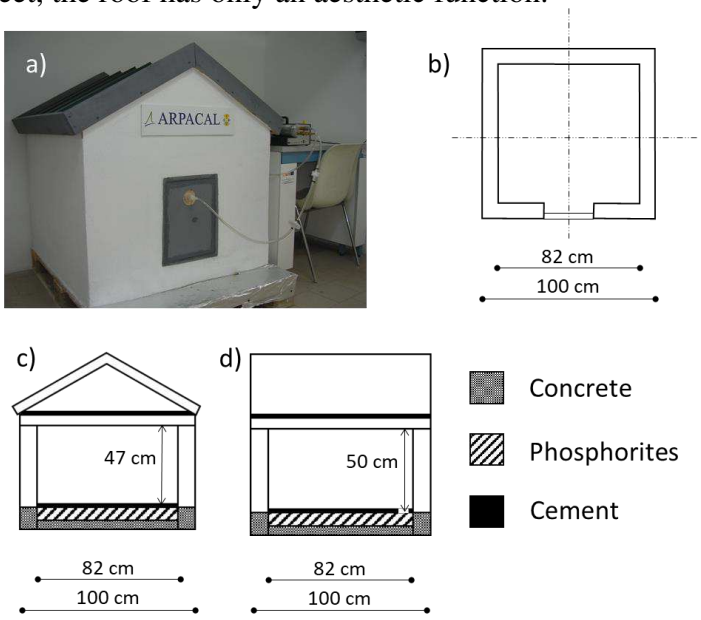


Figure 3:Building Details of Mary's House. a) Picture of the in-scale house model; b) Planimetry of the house; c-d)Model sections along the two dashed lines in b) with building material details.

4 - Methods

The ^{222}Rn activity was measured by means of both active and passive devices. In particular, for a real-time monitoring, we used a Lucas cell device (one may noting it connected to the door in the picture of Fig.3a). This allows us to simultaneously monitor the ^{222}Rn activity and the environmental parameters such as the temperature, the pressure and the humidity. On the other hand, passive devices, such as CR39s and electrets, provide long time and more accurate activity

concentration estimation. The electret-based method involves electrostatic measurements so that the ^{222}Rn activity concentration [^{222}Rn] can be obtained as (Caresana 2005)

$$[^{222}\mathrm{Rn}] = \left(\frac{V_i - V_f}{C_f \cdot t_e} - C\gamma\right) \cdot H \quad (2)$$

where V_i and V_f are the initial and final surface potential respectively, C_f is a calibration coefficient, C_γ is the γ-radiation equivalent ^{222}Rn concentration, H is a factor taking into account the altitude, and t_e is the exposition time. The parameter C_γ is measured by putting an electret, dubbed "witness", in an hermetic closed plastic bag in order to evaluate the electrostatic potential drop due to γ-radiation. On the other hand, the CR39-based method consists of counting the traces formed on a plastic material by α particles emitted during the ^{222}Rn decay. These are directly proportional to the activity concentration (Kropat 2015)

$$[^{222}\mathrm{Rn}] = \frac{\rho_{net} \cdot k}{t_e} \quad (3)$$

where ρ_{net} is the density of clear traces in the detector sensitive area (in our case 25 mm x 25 mm) and k is an instrumental calibration factor. In order to enlarge the traces and make them visible at the microscope, the CR39s were soaked for one hour in a sodium hydroxide (6.25 molar) bath at 98°C. The number of traces was then recorded by an automatic optical reader (Politrack microscope from Miam srl). In Fig.4 we report pictures of the microscope and the detectors used in the experiments. When using passive devices, we kept tracking the ^{222}Rn concentration as well as the pressure, humidity and temperature by the Lucas cell. In this way, the experimental condition stability was ongoingly monitored. Significant variations of these parameters were never observed during all the experimental campaigns.

Figure 4 - Pictures of CR39 devices (left) and the optical microscope Politrack (right)

5 – Results

The house model was built in the central structure of Arpacal, Agency for the protection of the environment of Calabria Region, in Italy, where the annual average concentration of the ^{222}Rn activity is 35 ± 12 Bq/m^3. However, in order to increase the accuracy of the background estimation, we monitored the ^{222}Rn activity inside the model before filling the crawlspace with the phosphorite residues. In Fig.5, we plot the values of [^{222}Rn] measured every hour, for 5 days, by the Lucas cell. In the spite of some fluctuation ascribable to environmental parameter variation, the measurements show that an activity concentration plateau is reached after a couple of days (solid line in Fig.5). However, the saturation value might over- or underestimate the actual activity, that can be more precisely determined in long term measurements by means of passive devices. A comparison of [^{222}Rn] measured by Lucas Cell, electrets and CR39s is reported in Fig.6 for three different configurations: crawlspace free, residues-filled crawlspace and residues-filled crawlspace covered by a 3 cm thick cement screed. The values reported are averaged over 11 CR39s and 6 electrets used in two different campaigns, each of which being about 3 months long.

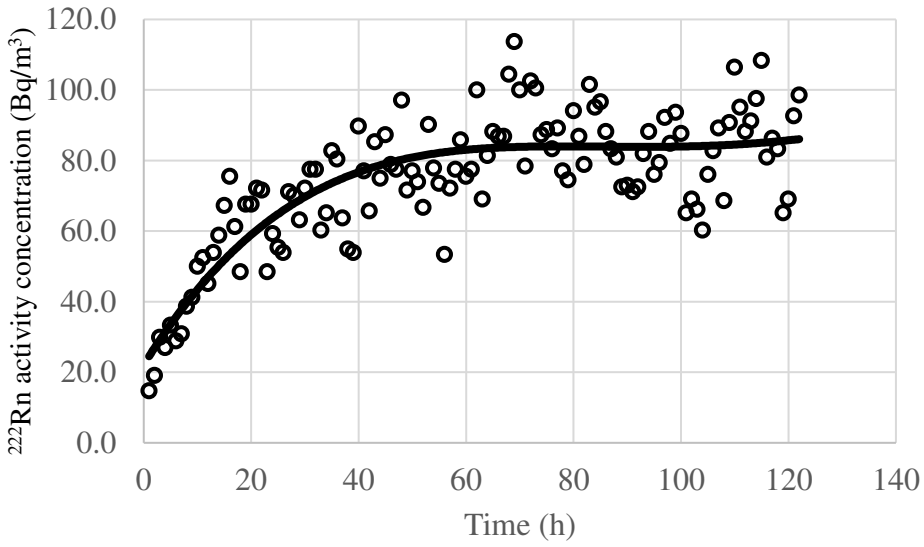
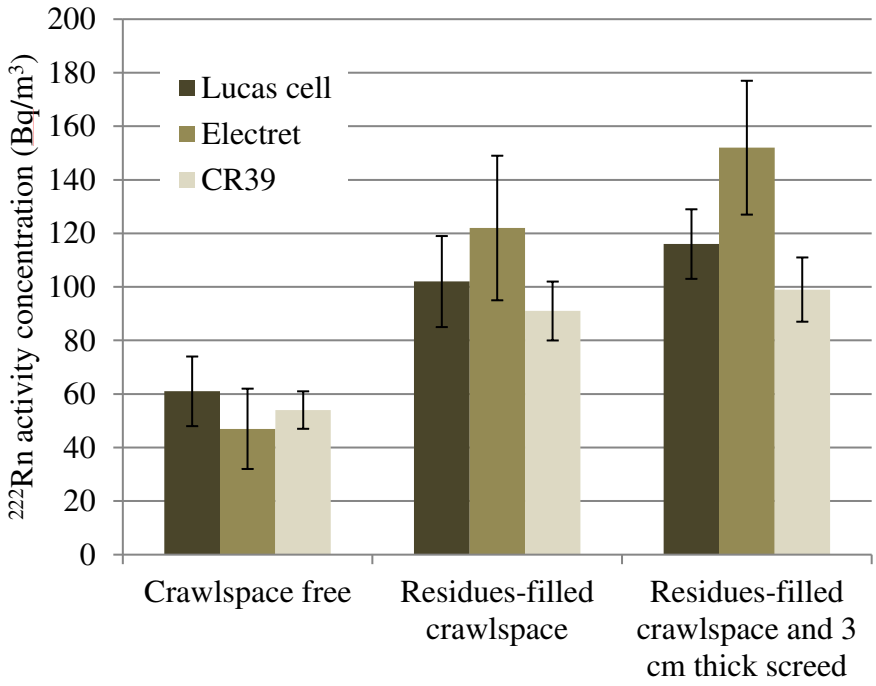


Figure 5 - Ongoing measurement of the background ^{222}Rn activity concentration (circles). The solid line is a guide for the eye highlighting the saturation.



	^{222}Rn activity concentration (Bq/m^3)		
	Crawlspace free	**Residues-filled crawlspace**	**Residues-filled crawlspace and 3 cm thick screed**
Lucas cell	61±13	102±17	116±17
Electret	47±15	122±27	152±25
CR39	54±7	91±11	99±12

Figure 6 - Values of the ^{222}Rn activity concentration measured by Lucas Cell, Electret and CR39 in three different configurations: crawlspace free, residues-filled crawlspace and residues-filled crawlspace covered by a 3 cm thick screed.

6 - Discussion

The values provided by active and passive devices are rather compatible, as shown in Fig.6. They all show the same trend, i.e. a significant increase of the ^{222}Rn activity concentration due to the presence of metasilicates. In fact, the measured activity doubles when residues are placed inside the crawlspace, growing from about 50 Bq/m^3 (just little higher than the environmental activity because of concrete, bricks and mortar used (Righi 2006)) to over than 100 Bq/m^3. An even further increase is observed when the cement screed covers the residues; the electrets provide a value over than 3 times higher than the background one. Coherently with literature (Keller 2001), it is possible to state that ^{222}Rn can diffuse inside the cement, meaning that the latter cannot actually confine the gas. Naming $[^{222}\mathrm{Rn}]_0$ the background activity, that corresponds to the crawlspace free configuration, we can write:

$$\left[^{222}\mathrm{Rn}\right] = \left[^{222}\mathrm{Rn}\right]_0 + [^{222}\mathrm{Rn}]_{res} \qquad (4)$$

where $[^{222}\mathrm{Rn}]$ is the measured value of the activity in the final configuration and $[^{222}\mathrm{Rn}]_{res}$ is the residues contribution. If we consider the most conservative situation, i.e. the highest value of the activity given by the electrets, the ^{222}Rn component generated by the phosphorous materials reads 105 Bq/m^3. Such a value is significantly high, being the triple of the original environmental activity. One may object that even the cement screed contributes to $[^{222}\mathrm{Rn}]_{res}$, since it might contain some radionuclide. However, comparing the residues-filled crawlspace configurations with and without the screed shows that the cement contribution is relatively small, and can be neglected as a first approximation.

On the other hand, the screed presence is crucial for shielding α-, β- and γ-radiations (Han 2017). By means of a radiometer, we estimated that the 3 cm thick screed is able to attenuate the γ-rays generated by the phosphorous material by a factor 2. Analogously, we observed a complete damping of the β contamination over the screed surface, passing from an initial value of 43 to 2 cps. Finally, one may observe that the device displacement could affect the validity of the experiment. This would be a non-negligible drawback in real cases, where the volume involved are much higher while the number of devices remains limited. To highlight such a crucial aspect, we homogeneously displaced the 11 CR39s inside the house-model and analyse the vertical distribution of the measured activity, depicted in Fig.7. For sake of simplicity, we investigate in this work the sole configuration of filled crawlspace without the screed. The figure clearly shows a non-monotonic trend with respect to the height, clearly indicating that the ^{222}Rn activity concentration is homogeneously distributed around the average value (dashed red line) in the whole volume.

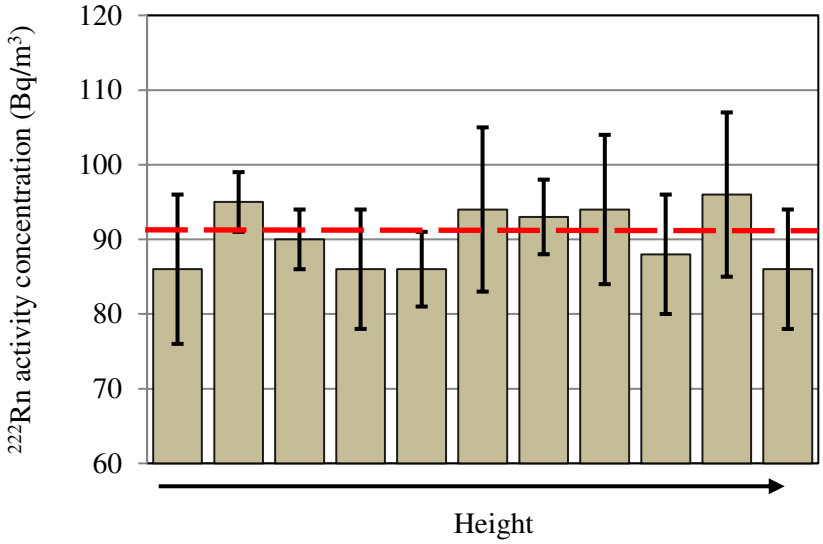


Figure 7 - Distribution of the ^{222}Rn activity concentration along the height, measured by 11 CR39s when the crawlspace is filled with the residues before covering with the screed.

7 – Use case

As a last step of our experiments, we tested the previsions given by the Mary's House model to a real use case. We chose an industrial warehouse of Crotone city, located in a zone where the presence of metasilicates had been previously checked. For privacy reasons, we do not provide in this article details about the location of the building. However, its planimetry is sketched in Fig.8a,b; the red circles stand for the exposure meter positions. We underline that the warehouse was a pretty airy environment, rich of air flows, where even the heat confinement was challenging. Therefore, in normal conditions, one may expect to find relatively close values of indoor and outdoor [^{222}Rn].

As for the model, we performed a 3 months long campaign. Our measurements were carried out by means of 14 passive devices: 9 CR39s and 5 electrets (besides the witnesses). A picture of both the devices is shown in Fig.8c while the results are reported in Fig.8d. We found a good agreement between CR39 and electret measured values, that are anomalous for the territory, where the average indoor [^{222}Rn] ranges between 40 and 50 Bq/m^3. Whereas, in our experiments values above 100 Bq/m^3 are found, as for the Mary's house. Remarkable is the case of the WC, which was under construction during the measurement campaign, and thus considerable as an outdoor environment. There, we found activity values compatible with the other (indoor) rooms, meaning that there is a significant contribution to the ^{222}Rn activity coming from the outside. In fact, we measured a remarkably high ^{222}Rn activity concentration in the square facing the warehouse: active devices registered values similar to the indoor experiments, while the outdoor average activity of the territory is only about 15 Bq/m^3. In the graph in Fig.8d, we also plot the CR39 average values of each floor. One may expect to find a sharp difference between the average activity of each floor if only the soil contributes to radon exhalation. Conversely, here we found almost the same activity concentration values, meaning that an artificial contribution is present. This arises from metasilicates inside the building structure as predicted by the model, where the [^{222}Rn] vertical spatial distribution is flat.

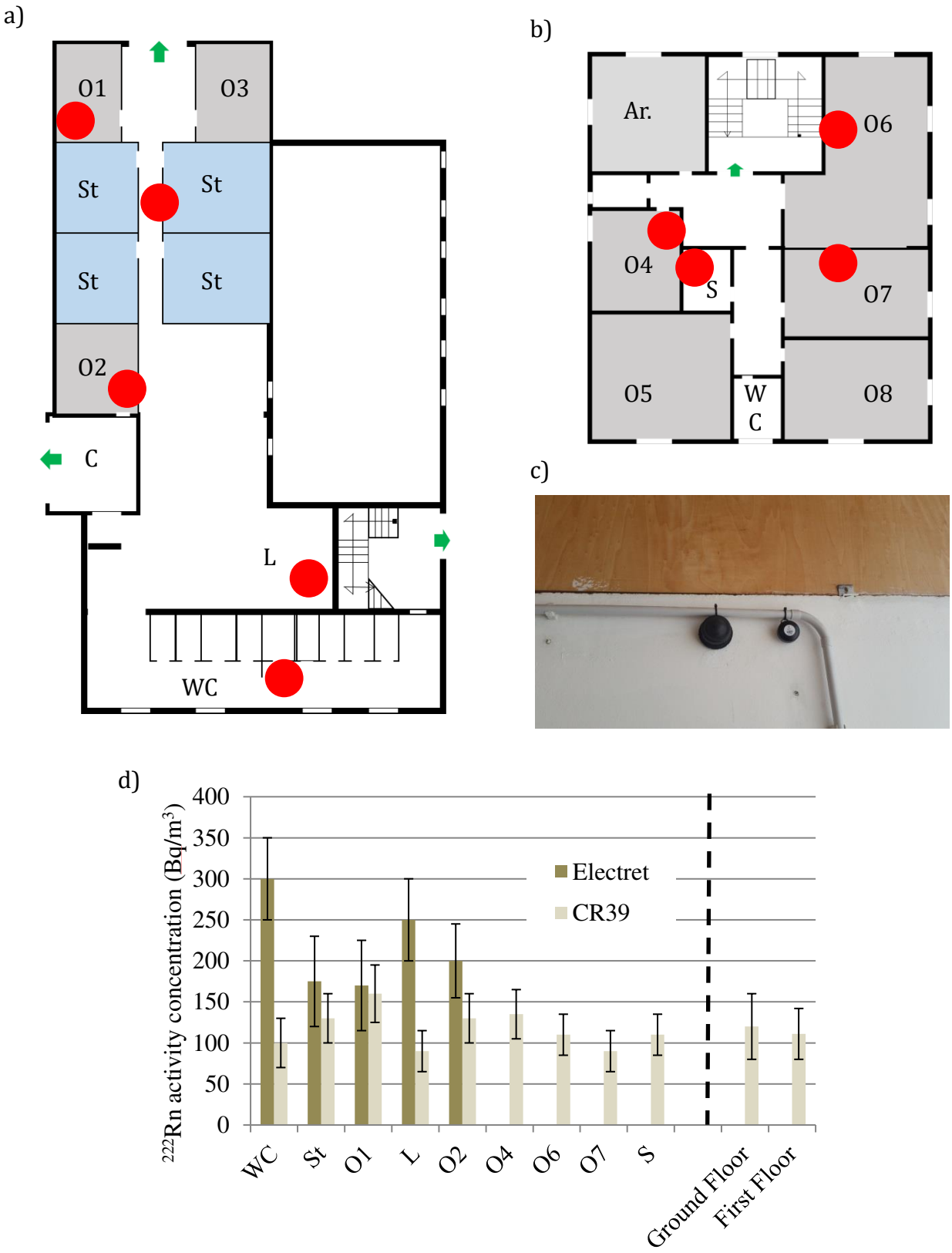


Figure 8 - a,b) Positioning of the CR39 in the industrial warehouse at the ground (a) and first (b) floor respectively. In the planimetry the letters stand for O (Office), St (Storage), C (Cafeteria), S (Server Room), Ar. (Archive). c) Picture of the Electret and CR39 used in the experiments. d) [^{222}Rn] measured in each room and average values in each floor.

8 - Conclusion

We have shown how metasilicates-containing TENORM contribute to ^{222}Rn activity. A house-model was realized by employing TENORM as building materials. These have a non-negligible ^{238}U component, responsible of a significant increase of the ^{222}Rn activity that becomes more than twice higher than the background benchmark. This result was tested by using both active and passive devices, whose measurements found a rather good agreement. The spatial distribution of the gas inside the house-model volume was investigated; we found an homogeneous distribution of the activity with respect to the height. The model results were compared with a real use case: a warehouse in the Crotone industrial zone. As predicted by the model, we found unusual values and spatial distribution of the ^{222}Rn activity, indicating the presence of metasilicates inside the building structure. In this sense, measuring the ^{222}Rn activity is a practical way to indirectly determine if and where TENORM are employed in the construction of a building. In particular, we remark that the average [^{222}Rn] measured inside the warehouse is about 120 Bq/m^3, while the typical [^{222}Rn] values of Crotone indoor environment are more than twice lower (between 40 and 50 Bq/m^3). In terms of perspective, the house-model can be used to verify whether artificial ventilation can be useful to significantly quench the ^{222}Rn activity concentration in a controlled environment with a known volume. Our results shade a light on the employing of phosphorites factories residues that, considered inert materials, were used to build roads and houses in the city, underrating the radon issue. We highlight that, if controlled, a recycle of the residues is still possible. Surely, they must not be used for school or warehouse building, as in our study case, but they might find application in outdoor environments. Nevertheless, for these applications, particular attention must be paid to water drainage, and phosphorite residues must be used only far from aquifers. Furthermore, the erosive action of the sea breeze and the physiological deterioration due to atmospheric agents must be considered. These may attack the silica component of the residues, producing an increase of the particulate matter (mainly heavy radioactive nano- and picoparticles that maybe redeposited on the surface). We conclude underlining the peculiarity of Crotone city, where a flood of the Esaro river (that is dangerously close to the industrial zone, as shown in Fig.1b) may further unearth residues and bring them to the sea and to the beaches, possibly producing an even greater exposition of the population.

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Author contribution All authors contributed to the study conception and design. Material preparation, data collection were performed by S.P. Analysis were carried out by A.N. and M.R.L. The first draft of the manuscript was written by M.F. and all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

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Data availability All data generated or analysed during this study are included in this published article.

Declarations

- **Ethics approval and consent to participate.** Not applicable.
- **Consent to publish** Not applicable.
- **Competing interests** The authors declare no conflict of interest.

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Figures

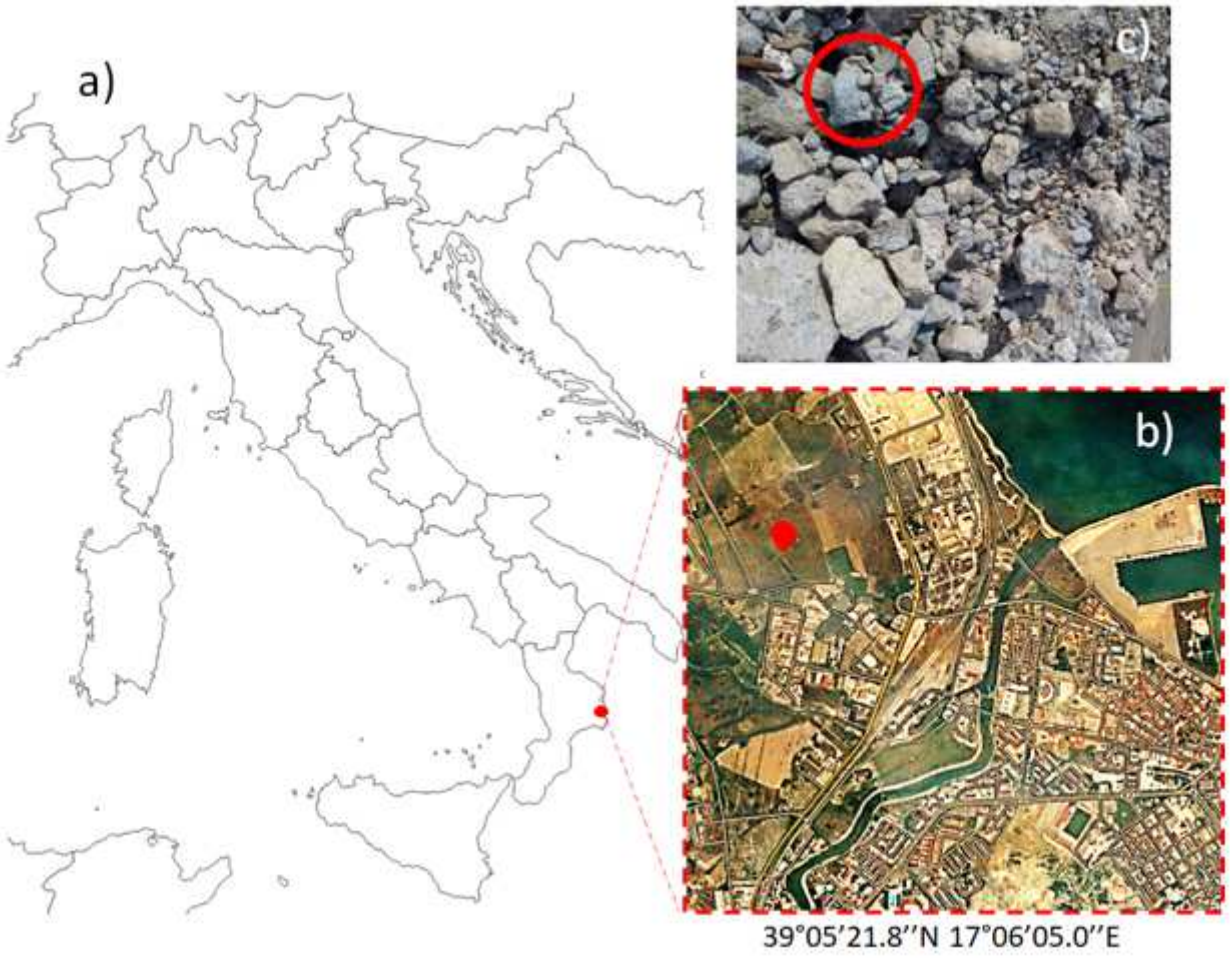
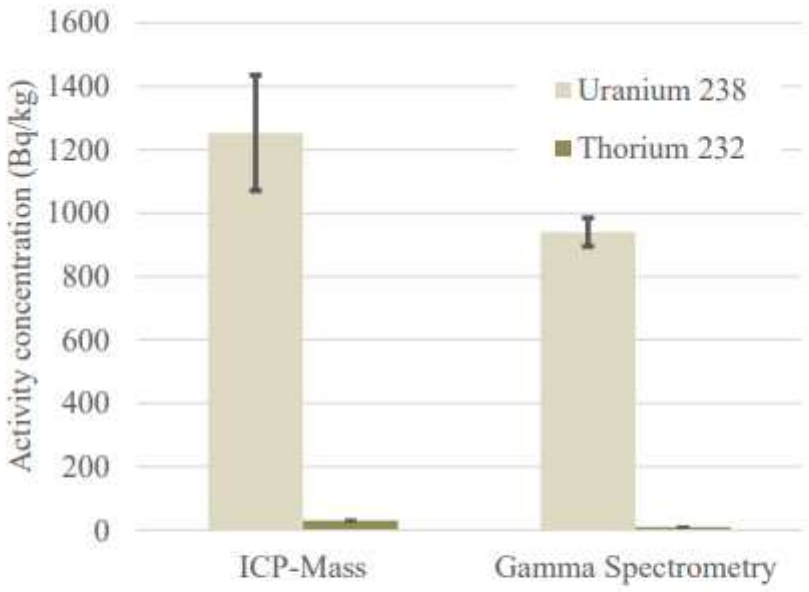


Figure 1

a) Crotone position in the Italian map. b) Harvesting site location. c) Picture of the phosphorite rocks showing their typical blue colour. Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.



	Activity concentration [Bq/kg]	
	ICP-Mass	γ-spectrometry
^{238}U	1253±182	940±45
^{232}Th	30±4	10±2

Figure 2

Uranium and Thorium activity concentration measured by ICP-Mass and -spectrometry.

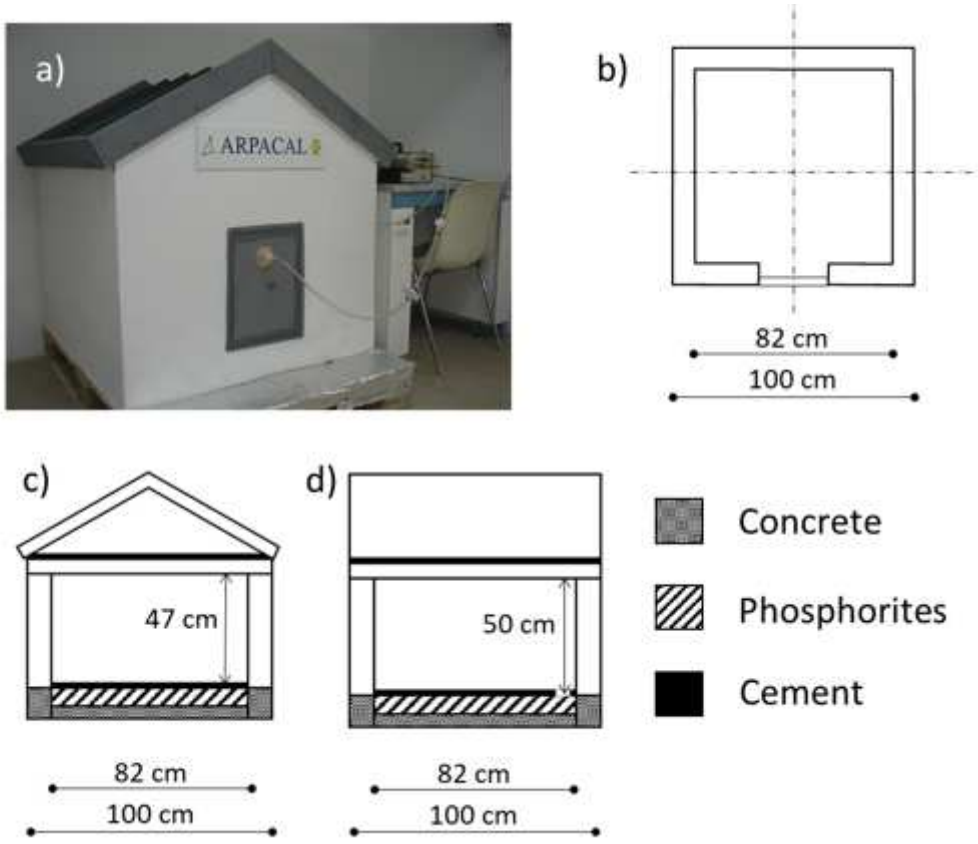


Figure 3

Building Details of Mary's House. a) Picture of the in-scale house model; b) Planimetry of the house; c-d)Model sections along the two dashed lines in b) with building material details.

Figure 4

Pictures of CR39 devices (left) and the optical microscope Politrack (right)

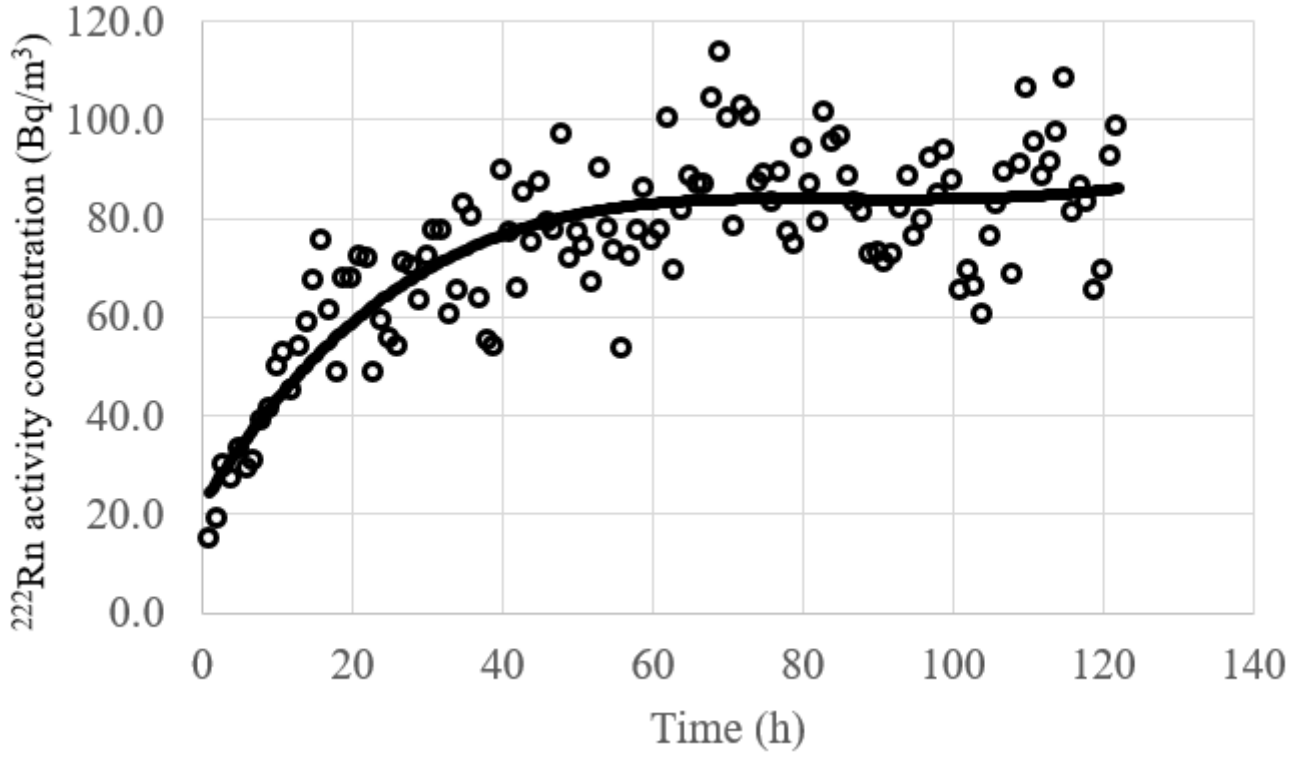
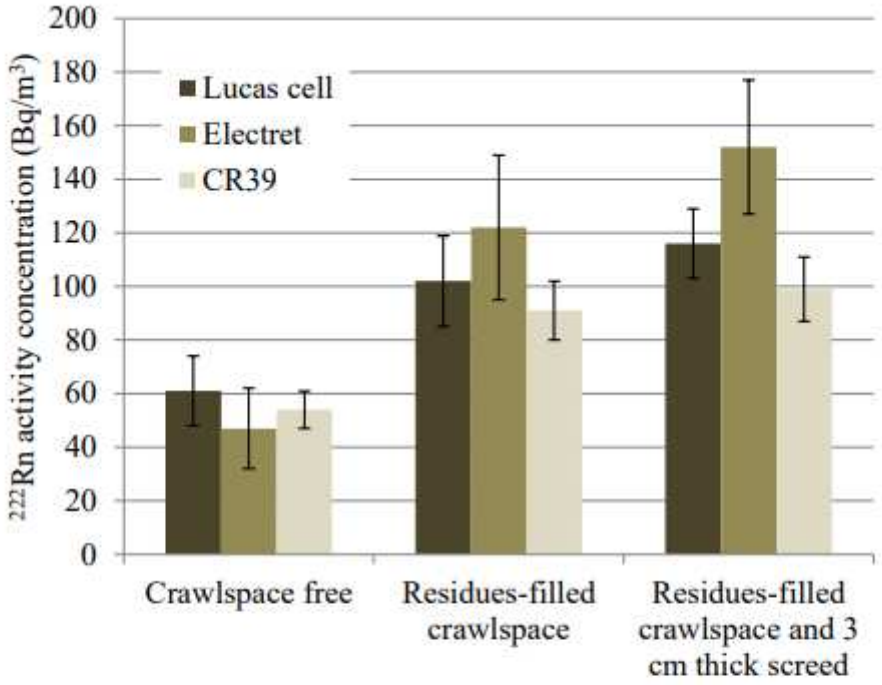


Figure 5

Ongoing measurement of the background 222Rn activity concentration (circles). The solid line is a guide for the eye highlighting the saturation.



	222Rn activity concentration (Bq/m3)		
	Crawlspace free	**Residues-filled crawlspace**	**Residues-filled crawlspace and 3 cm thick screed**
Lucas cell	61±13	102±17	116±17
Electret	47±15	122±27	152±25
CR39	54±7	91±11	99±12

Figure 6

Values of the 222Rn activity concentration measured by Lucas Cell, Electret and CR39 in three different configurations: crawlspace free, residues-filled crawlspace and residues-filled crawlspace covered by a 3 cm thick screed.

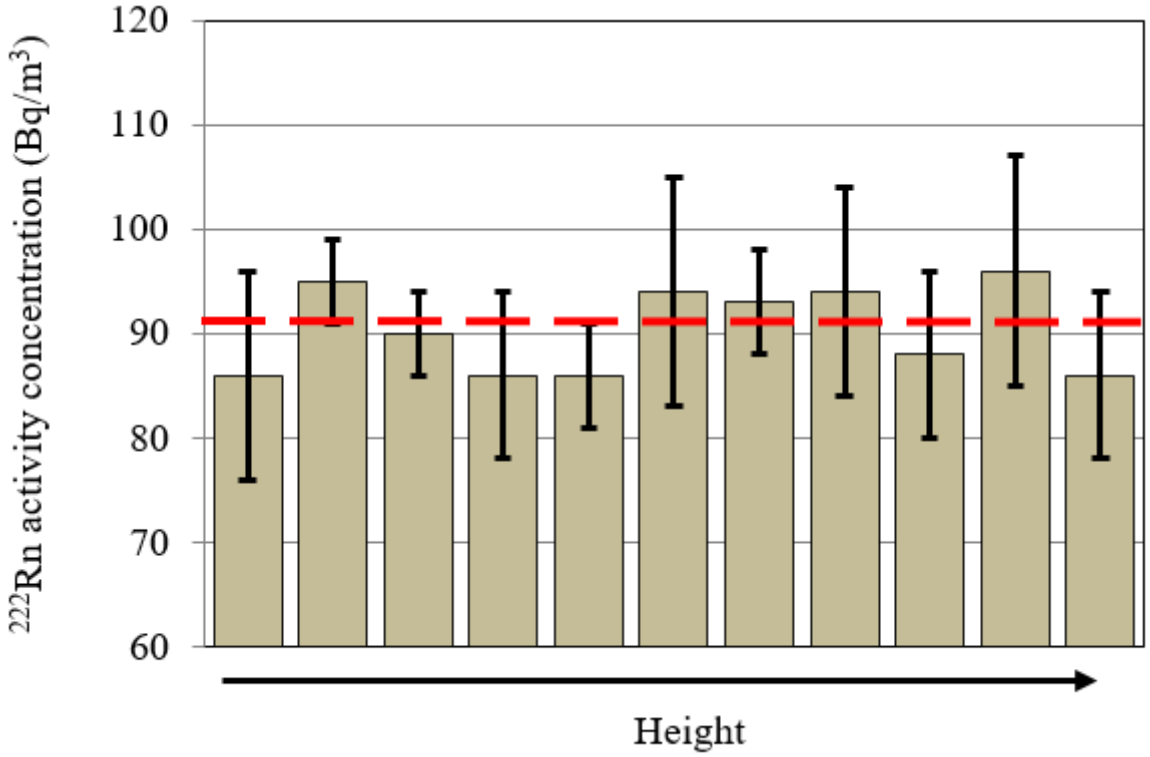


Figure 7

Distribution of the 222Rn activity concentration along the height, measured by 11 CR39s when the crawlspace is filled with the residues before covering with the screed.

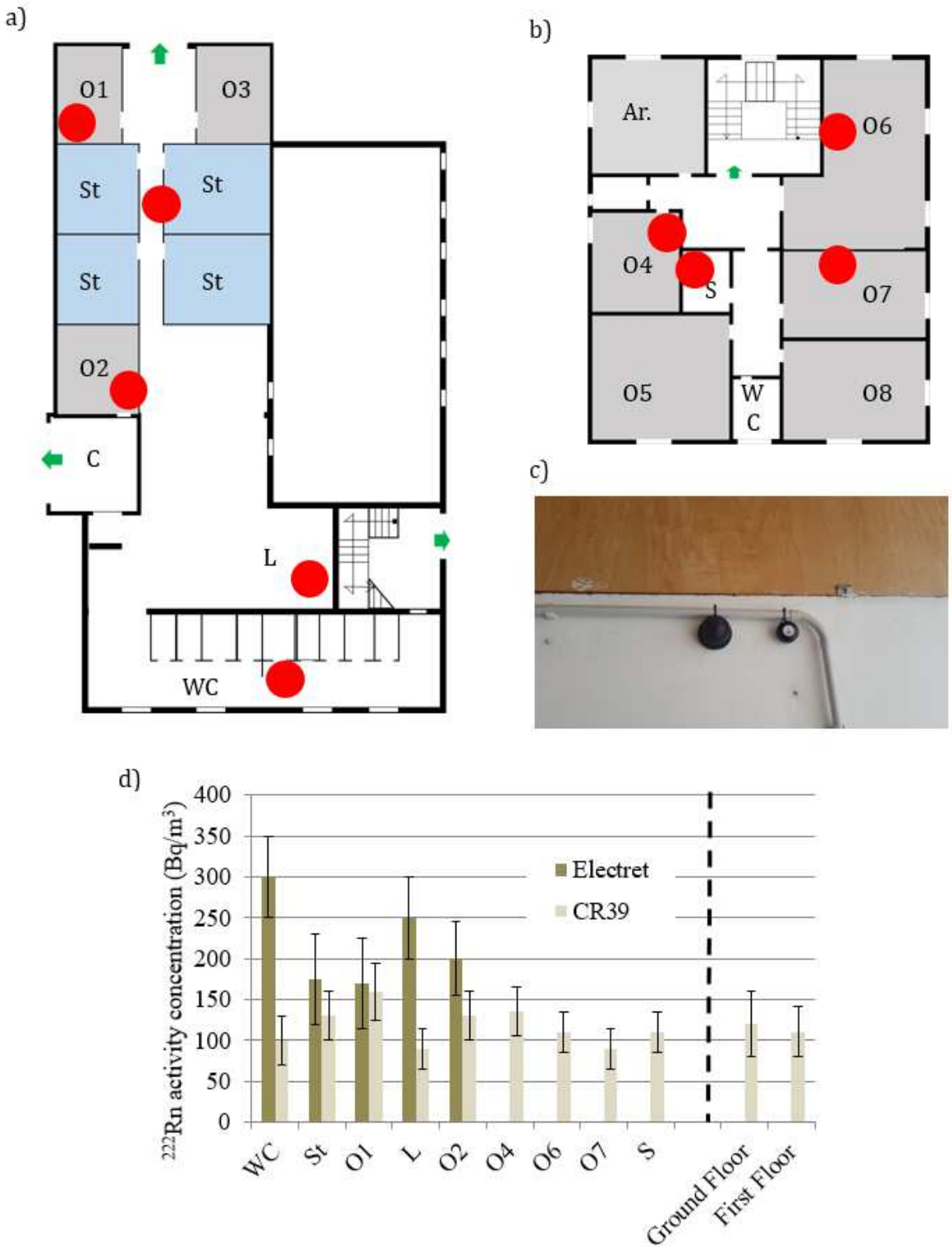


Figure 8

a,b) Positioning of the CR39 in the industrial warehouse at the ground (a) and first (b) floor respectively. In the planimetry the letters stand for O (Office), St (Storage), C (Cafeteria), S (Server Room), Ar. (Archive). c) Picture of the Electret and CR39 used in the experiments. d) [222Rn] measured in each room and average values in each floor.