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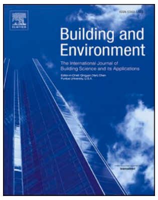

Inner wall filler as a singular and significant source of indoor radon pollution in heritage buildings: An exhalation method-based approach

Borja Frutos [a,*], Fernando Martín-Consuegra [a], Carmen Alonso [a], Gloria Perez [a], Joaquín Peón [b], Alberto Ruano-Ravina [b], Juan M. Barros [b], Ana M. Santorun [c]

[a] *Eduardo Torroja Institute for Construction Science, IETcc (National Research Council, CSIC), Madrid, Spain*
[b] *Area of Preventive Medicine and Public Health. University of Santiago de Compostela, Spain*
[c] *Tower of Hercules, Turismo de A Coruña, Spain*

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ABSTRACT

The presence of radon in buildings is a matter of growing concern in the industry. A further layer of complexity is present in heritage buildings, where sources of exhalation other than those observed in more modern conventional buildings may render diagnosis and intervention even more difficult. This study explored the high exhalation rates originating in the inner fillers in thick elements such as bearing walls and structural floors and vaults characterising historic construction. They were found to be close to the values observed in soils and one to two orders of magnitude greater (range: 32.5 mBq/m2·s to 149.7 mBq/m2·s) than found in the construction materials themselves, such as granite. The radon emitted into building interiors by those members exhibited more or less uniform concentration profiles on all storeys, irrespective of elevation and consequently distance form the soil. Further to the results delivered by an accumulative model, the only explanation for the empirical findings is that the inner filler in structural members sources a substantial fraction of the high exhalation rates. That would open a new exploratory pathway for remedies that should necessarily address all emissions, rather than deeming the soil as the sole or primary source of radon gas. The issue is broached in this article on the grounds of a case study of the Tower of Hercules at Corunna, Spain, a building dating from Roman times and presently used as a museum and monument open to the public.

1. Introduction

Radon is a colourless, odourless, insipid, gaseous decay product of the radium present in rocks on the Earth's crust. It is a carcinogen for humans [1], for whom a dose-response ratio has been defined between concentration and risk of lung cancer [2,3]. Specific studies have begun to be published stressing the importance of verifying indoor air quality in historic buildings [4,5]. In countries with a vast legacy of heritage works such as Italy, papers dating from the nineteen nineties have reported higher radon concentrations in castles than in other types of structures [6]. More recent studies conducted to meet European requirements [7] on worksite protection have assessed environmental, anthropic and construction-related factors that may affect radon ingress in such buildings [8]. Concern has also been expressed around the possible effect of the techniques used to clean monuments and their materials on the rise in radon exhalation rates attendant upon stone decay. One paper on that subject assessed the radon emitted during microwave treatments to verify and monitor alterations [9].

The radon concentration in a given space is essentially determined by the equilibrium between the ingress and ventilation-induced dilution rates [10–13]. Ingress is defined as the sum of all the gas emitted by the elements in place [14], with the soil identified as the major source under most circumstances [15–17]. As a decay product of the radium on the Earth's crust, radon gas travels across the pores in soil and permeates buildings through the weak points in their envelope such as fissures, cracks or unsealed joints [18–22]. Advective transport from the soil, induced by differences in soil and indoor building pressures, is normally the predominant mechanism [23–27].

Construction materials are also deemed a source of indoor radon concentration. Their radon content and characteristics of their internal structure determine their exhalation rate and with it the fraction of indoor concentration that can be attributed to them. As diffusion, usually

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* Corresponding author. Eduardo Torroja Institute for Construction Science, IETcc-CSIC, C/ Serrano Galvache 4, MADRID, 28033, Spain.
E-mail addresses: borjafv@ietcc.csic.es (B. Frutos), martin-consuegra@ietcc.csic.es (F. Martín-Consuegra), c.alonso@ietcc.csic.es (C. Alonso), gperezaq@ietcc.csic.es (G. Perez), joaquin.peon@usc.es (J. Peón), alberto.ruano@usc.es (A. Ruano-Ravina), juanm.barros@usc.es (J.M. Barros), a.santorun@coruna.es (A.M. Santorun).

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the transport mechanism that prevails in such cases [28], is a slower process, materials generally account for a smaller share of concentration than the soil [29,30].

Construction materials were discovered as a source of radon exhalation in the nineteen eighties [31–33]. In the interim many studies have classified these sources by their radionuclide content and emission potential based on parameters including diffusivity, radon emanation fraction, permeability, thickness, porosity, moisture and temperature [34–42].

The findings can be entered into predictive models to assess the indoor concentration attributable to materials. Nonetheless, given the fairly minor magnitude of such contributions compared to the amounts sourced from the soil [43–46], they are sometimes deemed negligible to simplify calculations on the understanding that such an approach introduces no substantial deviation in the end result. One example of that procedure can be found in a case study of the rehabilitation of heritage buildings dedicated to public use at Pushkin, Russia, where only geological sources were explored [47].

While largely valid for most standard buildings, the simplification at issue may not be suitable for singular, structurally massive buildings with high radon content materials. That is the case of historic buildings where structural engineering was based on the stability afforded by gravity and building to high elevations entailed the construction of very thick, normally stone, walls. Given its abundance but especially its strength and stiffness, granite, whose high uranium content and radon exhalation potential are well known [37], was used for such structures in many areas of the world. In these buildings, the double wythe of granite was reinforced by filling the space in-between with mortar-bound discards comprising smaller rocks or local soil. That technique, known as *opus caementicium*, was used extensively in Roman construction [48–50]. In such cases, the stone itself would not be the only (slow and scantly relevant) source of diffusive radon exhalation, for the filler would also contribute to that process across the joints in the ashlars. Under such circumstances the advective mechanism might be reinforced by the higher porosity and permeability of the medium (greater than in compact stone), which would raise rates considerably, particularly in material drawn from radon-prone areas [51–54].

In the West, Roman construction systems [55] remained in place for many centuries until lighter weight structures were introduced with Gothic architecture. Many monasteries, palaces, places of worship or castles built in the interim and often occupied today by (local or national) officials or teaching (including universities), religious or military staff, may therefore exhibit high radon concentrations induced by their structural systems and materials.

That concern informed the study described here: given the large fractions of smaller sized, high radon content material in historic building structures, the exhalation attributable to their construction systems and materials should be taken into consideration when diagnosing their radioactive status and proposing remedial action. Such buildings constitute a singular case where construction materials contribute to indoor radon concentration at a rate comparable to that of the soil. Intervention under those circumstances poses a particular challenge, in light of the degree of heritage protection or structural singularity involved. Standard mitigation action such as soil depressurisation [26,56–59] or the installation of membranes [60] is often difficult to implement or simply not feasible.

2. Objectives

The research described here explored radon exhalation and accumulation in historic heritage buildings whose structural systems and materials may constitute a source of the gas comparable to the soil.

The concentration profiles published in the literature for a number of historic buildings suggest the presence of sources other than the soil. The knowledge acquired with their identification was applied to the Tower of Hercules at Corunna, Spain, a building dating from Roman times and listed as a UNESCO heritage monument.

The methodology proposed and used detected and weighted the sources of radon based on the study of gas flows and accumulation. It may also serve as guidance for intervention, which should be tailored in such buildings to the protection accorded their heritage value, adding a further layer of complexity where radon levels are high.

3. Materials and methods

The methodology included collecting data which, entered into a model simulating ingress, accumulation and dilution, were used to assess the contribution of structural systems and respective materials to the indoor radon concentration in this manner of buildings. The materials and methods applied are described below.

3.1. Case study

Site: TOWER OF HERCULES, Corunna, Spain, (https://whc.unesco.org/es/list/1312#top).

This first-century Roman building has undergone a number of remodels over its two millennia history. Originally it was a lighthouse. Today, while still used for that purpose, it is a monument that can be visited nearly throughout, from the base with its archaeological site up to but excluding the upper storey that houses the actual light, accessible only to Corunna Port Authority staff. See Fig. 1 for an exterior image of the building.

Further to the plan view and cross-section of the building reproduced in Fig. 2, the structure is divided into two units. One, a basement located at the foot of the tower, is surrounded by a larger enclosure approximately 19.5 m in diameter with clearances ranging from 2 m to 2.5 m. It houses an archaeological site with the remains of walls and other Roman and Medieval constructions. A staircase connects this space to the other unit, the 11.4 m × 11.4 m tower itself. The tower comprises a ground plus three open plan storeys interconnected by a staircase, plus an upper storey that houses the light and has no such direct connection to the rest. Air circulates along the staircase from the basement to the third storey, as well as across openable façade windows and the entrances on the basement, ground and three upper storeys.

Granite is the major constituent material. The basement floor is the bare soil. The rest of the tower is floored with different forms of granite. Both the 2.3 m thick outer and 1.4 m thick inner walls consist in two wythes of structural stacked granite ashlars (*opus vittatum*) sandwiching a filler. In this Roman construction technique the space between two very thick walls was filled with smaller stones bound with lime mortar (*opus caementicium)*.

Larger square-hewn granite ashlars (*opus quadratum*) were used around openings and at corners, whilst the *opus caementicium* described above is directly visible in the vaults. The three techniques described are illustrated in Fig. 3.

3.2. Spatial distribution of radon concentrations

CR-39 passive trace detectors were installed to monitor radon concentrations in the tower storeys. The data collected during exposure were subsequently laboratory studied with a RADOSYS2000 (RSV-10) analyser able to determine the mean concentrations for a given period of time Thanks to that feature concentrations could be studied irrespective of daily or seasonal fluctuation. Measurements were made further to ISO standard 11 665–4 at the Laboratorio de Radón de Galicia (www.radon.gal), accredited by Spain's National Accreditation Agency (https://www.enac.es/). All the measurements were recorded during >2 months of exposure as recommended by WHO and Spain's National Nuclear Safety Council. This study used the 11 datasets recorded at the Tower of Hercules.

The purpose was to analyse radon concentration and its spatial distribution as a first step in studying their relationship to the sources. A

Fig. 1. Tower of Hercules, Corunna, Spain. (Source: Ana Maria Santorum).

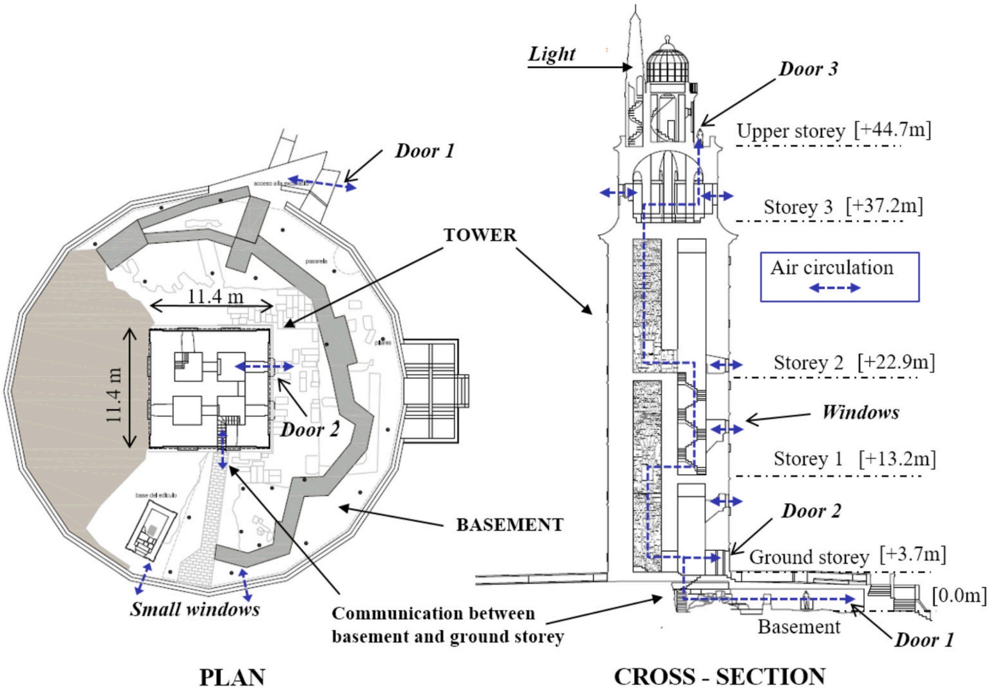

Fig. 2. Plan view and cross-section of the Tower of Hercules showing clearances, entrances and indoor and indoor/outdoor air circulation pathways.

vertical profile in which concentrations decline with rising elevation might identify the source as the soil, whereas a more uniform profile might indicate that a significant share of the concentration is attributable to the materials enclosing all the spaces in the building.

3.3. Wall and floor exhalation rate estimation

The indoor walls consist largely in unfaced granite. The ashlars' 0.5 cm–1 cm wide joints provide a pathway connecting indoor areas to the inner filler materials in the walls. Radon chambers were installed in areas with and without joints to determine the contribution to exhalation made both by the granite ashlars in walls, floors and vaults, and by the filler materials (Fig. 4).

The physics involved in the study drew from the mass balance equation [61], whilst the test was conducted to the procedure laid down in ISO standard 11 665–7 [62]. The 25 cm^3 methacrylate boxes used were secured to the respective surfaces with an oily clay paste. An FTLAB RD200P continuous radon monitor positioned inside the boxes recorded the readings every 10 min. In light of the scant presence of back diffusion, the slope on the accumulation curve used to estimate surface exhalation rates was obtained from Equation [1]:

$$C(t) = \frac{\Phi \times S}{V} \times \ t \qquad [1]$$

Fig. 3. Construction systems used in building structure.

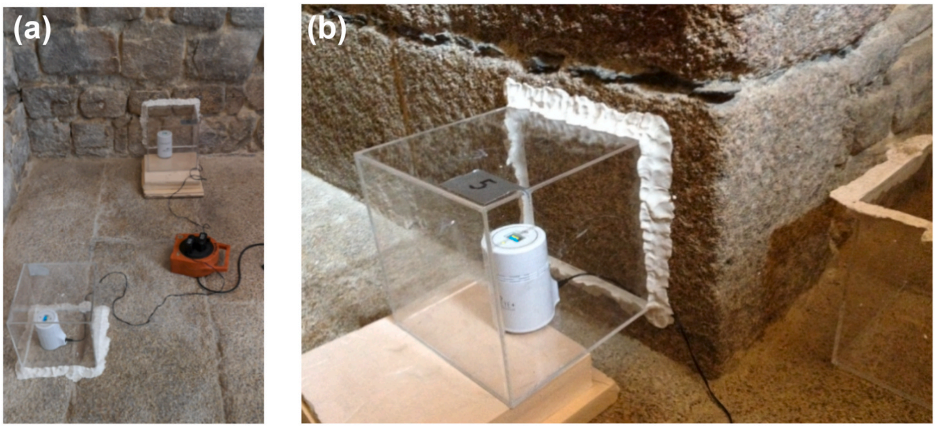

Fig. 4. Examples of test setups in the Tower of Hercules: a) *opus vittatum* wall with a joint; b) *opus quadratum* wall with no joint.

where:

C(t) = radon concentration at a given time (Bq/m^3)
Φ = exhalation rate ($mBq/m^2 \cdot s$)
S = area of exhalation surface (m^2)
V = accumulation box volume (m^3)
t = data recording time (s)

The tests were conducted on all the indoor wall and floor types in the tower, including areas with and without joints between the granite ashlars. Five tests were run in all: three in different types of walls on the ground storey and two in floors, one on the ground and the other on the first storey. The systems tested, representative of the construction types in place in the tower, were present on all storeys.

The standard procedure, in which rates are measured separately for each material, was not followed here. Rather, measurements were made on the actual walls and floors as positioned on the structure, some with and others without joints across which the inner filler might emanate radon (Fig. 4).

3.4. Ventilation rates and indoor airflows

Ventilation profiles were determined for spaces by measuring the airflows generated with a number of door and window opening/closure setups. A hot wire anemometer (velocity range 0 m/s to 25 m/s ±5%) was used to record the readings in interspace openings.

Air changes per hour (ACH), entered into the accumulation and dilution model, was calculated from Equation [2]:

$$ACH = \frac{Airflow\ rate}{V} \qquad [2]$$

where:

ACH = (air changes per hour), number of times the air is renewed per hour in a room (h^{-1}); airflow rate = airflow across openings on building exteriors (m^3/s); V = volume of air in a room (m^3).

Natural draughts in interiors were also analysed to measure the airflows in the spaces studied and hence radon gas circulation throughout. This study, conducted for all possible combinations of entrance opening/closing setups, measured air velocity at a monitoring point positioned at the connection between the basement and the ground storey (Fig. 2).

3.5. Accumulation and dilution model

Indoor radon concentration was estimated by calculating surface radon exhalation, an approach described in earlier studies. In Refs. [63–65] the authors compared the radon exhalation rates predicted by theoretical models to actual radon chamber readings, assuming certain inputs about the material. The results were subsequently validated for accuracy and consistency by comparing both types of data to the radon concentration in a number of dwellings.

The model used here deems that under steady state conditions radon concentration in a given space is determined by the balance between radon entry and dilution rates. The latter, in turn, is the sum of the air changes/hour due to infiltration and ventilation plus the radon decay rate [34,66–69]. The resulting expression is shown as Equation [3] below:

$$CRn = \frac{\Phi}{(\lambda Rn + \lambda ACH) \times V} \qquad [3]$$

where:

C_{Rn} = Steady state radon concentration (Bq/m^3); Φ = total exhalation from all surfaces (Bq/s); λ_{Rn} = radon decay rate (0.0000021 s^{-1}); λ_{ACH} = ventilation + infiltration-induced air change rate (ACH); V = accumulation volume in building (m^3).

Indoor radon concentration was predicted by entering the surface exhalation data (discussed in sub-section 4.2 below) and ventilation rates analysed (discussed in 4.3) in the model.

The model estimate was compared to the sensor readings to determine the significance of exhalation from the surfaces enclosing the spaces in the building. The originality of this study lies in the fact that it envisaged the exhalation attributable to both the exposed material and the radon released by the inner filler across inter-ashlar joints.

4. Results and discussion

4.1. Spatial distribution of radon concentrations

The 11 radon concentration readings for representative areas in the Tower of Hercules (Fig. 2) are given in Table 1: three in the basement, one on the ground, three each on the first and second and one on the third floor. The 101 d exposure period ran from July 18, 2017 to October 27, 2017.

Table 1
Mean radon concentration in the tower of Hercules by storey and elevation.

Storey	Elevation (m)	Rn concentration (Bq/m^3) expanded uncertainty k = 2 (±Bq/m^3)			Mean Rn (Bq/m^3)	SD
		1	2	3		
Basement	0	1216.9 (±86)	1160.6 (±82)	1156.7 (±82)	1178 (±83.3)	27
Ground	3.7	*666*[a]	–	–	666	–
First	13.2	1096.9 (±78)	1415.4 (±100)	1075.4 (±76)	1196 (±84.7)	155
Second	22.9	1595.8 (±113)	1187.7 (±84)	1504.9 (±107)	1429 (±101.3)	175
Third	37.2	1200.2 (±85)	–	–	1200 (±85)	–

[a] This value represents a single 6 min reading taken with a ‘sniffer’. The space has an outdoor entrance, which would explain the lower concentration.

On all but the ground storey that houses the main outdoor entrance, mean radon concentrations were high, ranging from 1178 Bq/m^3 in the basement to 1429 Bq/m^3 on the second storey. Concentration by elevation was observed to be fairly flat, a finding not usually reported in case study literature where, as noted in the introduction, indoor radon concentration is normally attributed to the soil and observed to decline with rising distance from that source [65,70–74].

Since the concentrations were found here to be fairly uniform and apparently unrelated to elevation, the working hypothesis was that significant sources of exhalation were present on all the storeys, contributing to radon concentration at rates similar to those of the soil.

4.2. Wall and floor exhalation rates

As noted in sub-section 3.3, a total of five radon chambers were installed on floors and walls, in areas with or without inter ashlar joints. The tests sampled two of the construction solutions (*opus vittatum* and *opus quadratum*) observed in the structural system, which is very uniform from top to bottom of the tower and characterised by granite tile flooring throughout.

The possible effect of back diffusion was deemed negligible in the estimates, resulting in some cases in underestimated exhalation rates. The tests were conducted at the ambient conditions prevailing in the tower.

The exhalation rates found for the representative elements in walls and floors sampled are listed in Table 2.

The exhalation rates recorded in wall and floor materials in the building were higher than reported by other authors for construction materials. In Ref. [44] for instance, construction granite was found to have a mean rate of 0.63 mBq/m^2·s, whilst the values measured for different materials in Ref. [45] ranged from 0.05 mBq/m^2·s to 0.4 mBq/m^2·s.

The readings recorded in the tests conducted in wall areas with an inter-ashlar joint, in turn, were one order of magnitude greater than the values found for jointless areas: test 3, run on a single sound ashlar (*opus quadratum*), delivered a rate of 3.6 mBq/m^2·s, whereas in tests 1 and 2, conducted in the same space but on stone surfaces bearing joints, values of 33.3 mBq/m^2·s and 50.2 mBq/m^2·s were recorded. Rates were also observed to be higher in thicker walls, perhaps in keeping with the greater amount of inner filler: the 33.3 mBq/m^2·s found for a 1.4 m thick indoor wall (test 1) and the 50.2 mBq/m^2·s façade in the same space (test 2). Photographs of the tests are reproduced in Fig. 4. These data suggest that the radon in the inner wall filler may preferentially migrate into the indoor space across the inter-stone joints (Fig. 5).

That singularity, in conjunction with the huge mass of the filler material, might explain the exhalation values of orders of magnitude more characteristic of soils than of materials. Soil exhalation potential studies conducted in France [75,76], for instance, delivered values ranging from 1 mBq/m^2·s to 100 mBq/m^2·s. The worldwide mean, according to Ref. [43], is 20 mBq/m^2·s. The mean value reported for the

Table 2
Exhalation rates on wall and floor surfaces in the Tower of Hercules.

Test	Element	Φ = Surface exhalation rate (mBq/m^2·s)	Inter-ashlar joint
1	1.4 m thick indoor wall, *opus vittatum*	33.3	Yes
2	2.3 m thick façade wall, *opus vittatum*	50.2	Yes
3	1.4 m thick indoor wall, *opus quadratum*	3.6	No
4	Floor lying directly on soil	32.5	Yes
5	Floor lying over a vault	149.7	Yes

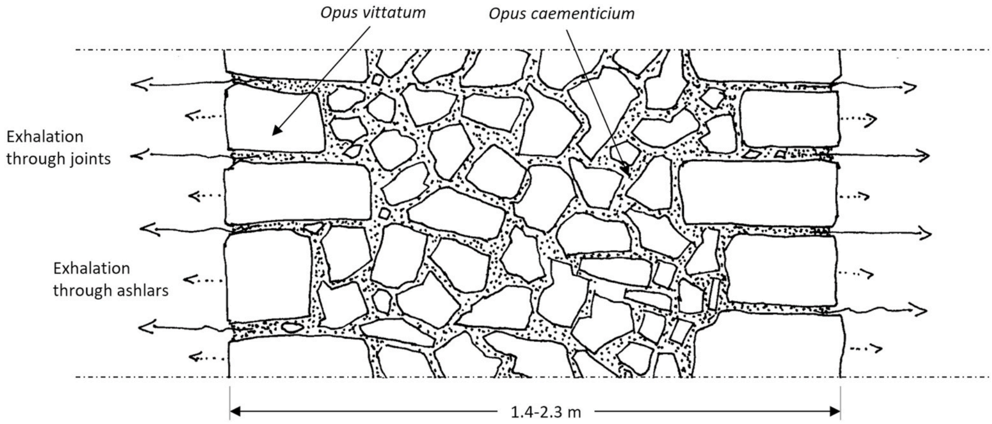

Fig. 5. Cross-section of double wythe wall (*opus vittatum*) showing hypothetical radon exhalation pathways from the inner filler material (*opus caementicium*).

Czech Republic ([77]) is 16 mBq/m^2·s, whilst in Spain the mean ranges from 10 mBq/m^2·s to 250 mBq/m^2·s, depending on the region [35].

The high values observed for floor surfaces were also more typical of soils than of materials. Although both tests were conducted on surfaces bearing joints, the exhalation rates observed varied, perhaps due to differences in their structural particulars. Test 4, with a value of 32.5 mBq/m^2·s, was run on a ground storey granite-tiled floor laid over the tower foundations and natural soil. Test 5, at 149.7 mBq/m^2·s, was conducted on a first storey, likewise granite-tiled floor, laid over a very thick *opus caementicium* vault, where the greater permeability relative to natural soil might have favoured exhalation, explaining the higher rate.

4.3. *Indoor airflows and air changes per hour*

Tests were conducted to identify and quantify draughts inside tower rooms from which to estimate air changes per hour with the outdoors for the intents and purposes of the accumulation and dilution model discussed in section 5.

As noted in sub-section 3.1, the building has three entrances connected to outdoors (Fig. 1) and partially openable windows and all the storeys are inter-connected. The air velocity and direction readings taken in a number of representative points in the tower on two separate days with the three doors opening and closing as usual are summarised in Table 3.

Further to the studies discussed below, strong, sometimes upward and sometimes downward, draughts were recorded. A direct relationship was observed between indoor draughts and the outdoor wind velocity on the two days.

The physics of airflows in a room describe two situations: the stack effect, which would explain upward air movements due to elevation and temperature differences [78]; and the effect of the wind and pressures affecting façades and openings [79]. The tower is sited in an area with strong winds, whose direction and intensity induce pressure or depressurisation depending on the façade. Wind orientation and its effect on openings, in turn, determine whether indoor draughts course upward or downward. The prevalence of wind velocity and direction over the stack effect in governing airflows in the building studied here would explain the differences in the patterns of draughts observed on different days.

Table 3
Airflow velocities and directions in the Tower of Hercules on two days.

Control point		22/11/2018		16/5/2019	
	Location	Vel. (m/s)	Direction	Vel. (m/s)	Direction
0	Outdoors	3.1–11.4	North	6.1–17.2	Northwest
1	Small window (basement)	1.0–3.5	↑	0.3–0.6	↑
2	Connection between basement and ground storey	0.1–0.4	↑	2.3–3.2	↓
3	Door 2 (ground storey)	0.8–2.1	↑	0.6–1.1	↑
4	Door 3 (upper storey)	0.5–0.8	↑	–	–

In pursuit of a fuller understanding of this complex natural ventilation system, the indoor draught velocities and directions were measured with different entrance opening/closure setups. The monitoring point for velocity readings was the stairwell connecting the basement and the ground storey (point 2), given its status as a controlled airflow pathway. The ventilation setups envisaged opening or closing of doors 1 (basement entrance), 2 (ground storey entrance) and 3 (upper storey entrance) described and depicted in sub-section 3.1.

According to the data in Table 4, and as expected, the lowest velocities were recorded in setup A, with all the doors closed, whilst the highest were observed in setup H, with doors 1 and 3 open and door 2, the ground storey entrance, closed.

Making allowance for the prevailing conditions, the findings were consistent with the open/close setups. Differences in outdoor wind velocity and direction were found to have a heavy impact on indoor airflows, and even change the direction from upward to downward or vice-versa in a matter of minutes. Those findings, in addition to the fact that door opening/closing varies with the number of visits and the weather on any given day, prevented establishing an exact pattern of indoor draughts on which to base air change per hour calculations.

Consequently, the velocity value used for the intents and purposes of the model was the median of the 16 readings listed in the table, i.e., 0.4 m/s. Ventilation airflows, in turn, at 0.6 m^3/s, were obtained by multiplying velocity by the area of the section of the pathway between the basement and ground storey, 1.6 m^2. That value was divided by 2 for the model, in the understanding that the air did not flow in a constant direction, but with alternating upward and downward movements, and that air changes cannot be defined in one direction only. The resulting airflow, 0.3 m^3/s, when expressed in air changes per hour (ACH) for the tower's total 2800 m^3 volume yielded a value of 0.38 h^{-1}.

That value is on the order of airflow rates reported in other studies conducted in Europe [80–82]. It also appears to be reasonable in the context of international standard ISO 13790 [83], which classifies building airtightness at a pressure of 50 Pa as measured in the blower door test. Applying those criteria to the conditions prevailing yielded a value ($>$0.25 h^{-1}) defined as low in the standard, which seems to be in keeping with tower realities.

Table 4
Maximum and minimum velocities at control point 2, by doorway opening/closure setup with outdoor NW wind speed 6.1 m/s to 17.2 m/s (https://opendata.aemet.es/).

Door	Set up (x = door closed)							
	A	B	C	D	E	F	G	H
1	x				x	x	x	
2	x	x				x		x
3	x	x	X				x	
Vel. (m/s)	0.1–0.2	0.3–0.4	0.5–0.8	0.6–1.6	0.1–0.2	0.3–0.4	0.2–0.4	1.7–2.0

5. INDOOR ACCUMULATION MODEL. Effect of inner wall mass

The test results set out in section 4 were entered in a radon accumulation and dilution model to gain a fuller understanding of the percentage contribution attributable to the sources identified, in particular walls and their inner fillers. A comparison of the predictions to the empirical data would provide grounds for drawing conclusions on radon exhalation in this type of historic buildings.

Further to the explanations in sub-section 3.5 the model, based on Equation [3], addresses the following items: exhalation rates in the areas measured on walls, floors and vaults; accumulation volume in the space at issue; and estimated infiltration or air-tightness expressed as air changes per hour.

The exhalation rates and areas of the surfaces measured on the various construction elements are given in Table 5. The value applied to the bare soil in the basement was the 120 $mBq/m^2 \cdot s$ reported in the literature for natural soils in radon-prone areas such as Corunna.

The total exhalation rate used in the model (281 Bq/s) was the sum of the values for all the components, assuming gas to accumulate across the total volume. The decision to apply that value was based on the understanding that the tower behaves as a single enclosure in which air circulates freely among all its parts. Application of those criteria delivered the results given in Table 6.

The concentration predicted by the model amounted to close to 84.4% of the actual readings. That discrepancy was associated with the ventilation measurements, as discussed in the preceding sub-section. Empirical exhalation rates may also have been underestimated due to back diffusion. That combination of factors might explain the deviation between the model and the empirical findings. The estimate is nonetheless fairly close to the actual measurements and higher than obtained if standard rates for construction materials are applied instead of the values measured in situ in areas with inter-ashlar joints. That was verified by replacing the present data with the rates reported in the literature for high exhalation materials (1 $mBq/m^2 \cdot s$). The result, 390 Bq/m^3, was much lower and much more discrepant with the data recorded.

That, together with the uniformity of concentrations across all the elevations, would appear to denote high exhalation rates in walls and floors, much greater than found in the literature for those members and closer to the values observed in soils.

This study showed that most of the radon accumulating in the tower is attributable not to the surface material itself (granite), but to the material filling the space between the two wythes forming its very thick walls. In this type of historic construction systems, inner filler exhalations would flow into indoor spaces across inter-ashlar joints. In light of the characteristics of the local soil and other materials used (crumbled stone bound with lime mortar), the filler may have a high radioactive isotope content, especially in radon-prone areas. Moreover, as an altered and manipulated material, the filler is more porous and permeable than its natural components. Those two factors may afford it high emission potential, giving rise to radon exhalation across the joints in the massive walls so typical of this manner of historic buildings.

Table 5
Exhalation rates and areas for five components and total Tower of Hercules exhalation.

Element	Φ = Exhalation rate ($mBq/m^2 \cdot s$)	Area (m^2)	Exhalation (mBq/s)
Natural soil (basement)	120	1039	124 680
Opus vittatum indoor wall (all storeys)	33.3	760	25 308
Opus vittatum façade (all storeys)	50.2	1270	63 754
Floor over natural soil (ground storey)	32.5	40	1300
Floors and vaults (storeys 1,2,3,4)	149.7	440	66 000
TOTAL EXHALATION			281 042

6. Conclusions

This study was undertaken to explore the hypothesis that the absence of any significant change in radon concentrations with elevation observed in some historic buildings might denote the presence of the radionuclides in elements other than the soil. The present paper discusses a case study conducted on a Roman building, the Tower of Hercules, at Corunna, Spain, a UNESCO world heritage monument characterised by the presence of massive structural walls, vaults and floors.

The literature was reviewed to collect data on laboratory-determined exhalation rates in construction materials. Although high values have been reported, particularly for rocks such as granite (at close to 1 $mBq/m^2 \cdot s$), they are not high enough to themselves explain the indoor radon concentrations recorded in the tower, where in situ readings of up to two orders of magnitude greater were recorded. The lowest value found for a jointless granite surface was 3.6 $mBq/m^2 \cdot s$. The jointed surfaces measured yielded values of 33.3 $mBq/m^2 \cdot s$ in 1.4 m indoor walls and 50.2 $mBq/m^2 \cdot s$ in 2.3 m façades. The tower's stone floors exhibited high and uneven values: 32.5 $mBq/m^2 \cdot s$ in a stone floor laid on natural soil and 149.7 $mBq/m^2 \cdot s$ in one laid on a vault with inner filler.

These exhalation data were entered into a pollution accumulation model together with air change per hour values. A comparison between the model predictions and the concentrations actually measured supported the explanation put forward for the radon exhalation rates observed in such buildings. Further to the model results, the concentrations measured are explicable only where high radon exhalation rates are assumed for wall and floor surfaces. Those high rates, in turn, are not attributable to the rock itself, but to radon migration from the inner filler across the joints between the ashlars forming the inner-most of the two constituent wythes.

That opens a new avenue for exploring such historic buildings, where not only the soil and construction materials, but also inner fillers in walls, vaults and floors must be viewed as possible sources of radon. Establishing the method needed to measure exhalation in walls and floors to supplement laboratory readings would entail in situ monitoring of the construction system as a whole, including joints between finishes, which are usually stone-based in heritage buildings.

Approaches such as adopted in this study should be envisaged when analysing centuries-old buildings whose very thick walls contain an

Table 6
Comparison of model and empirical radon concentration values.

	Total exhalation rate	Vol.	ACH (ventilation + infiltration)	Radon decay rate	Estimated radon concentration C_{Rn} (est.)	Measured radon concentration C_{Rn} (meas.)
	Φ (Bq/s)	(m^3)	λ_{ACH} (h^{-1})	λ_{Rn} (h^{-1})	(Bq/m^3)	(Bq/m^3)
Tower of Hercules	281	2800	0.38	0.00756	956.9	1133.8

inner filler. Given, furthermore, that many such buildings are occupied by institutions or government services, staff exposure to radioactivity is an issue not to be overlooked.

A broader study of the physics involved may lead to proposals for protection systems that address the problem holistically. One less invasive remedy than the present practice of treating all the stone to lower radon concentrations that might be explored, for instance, would be to simply seal the inter-ashlar joints with a material more impermeable to the gas (k $<$ 10–12 m^2 and D $<$ 10–11 m^2/s). Other approaches that might be considered include drawing the gas from wall interiors by positioning outward-evacuating depressurisation devices in the ashlars, provisions on heritage assets permitting. Techniques might also be deployed to dilute the gas with outdoor air. While less invasive, that solution would entail installing machinery and insufflation ducting that need to be compatible with building use, image or listed status.

Intervention in such buildings, never an easy task, calls for specific expertise as well as coordination with the professionals entrusted with heritage management.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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