Influence of architectural style on indoor radon concentration in a radon prone area: A case study

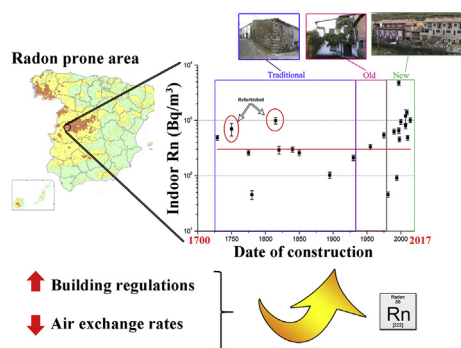A. Baeza [a], J. García-Paniagua [a], J. Guillén [a,*], B. Montalbán [b]

[a] LARUEX, Environmental Radioactivity Laboratory, Dpt. Applied Physics, Faculty of Veterinary Sciences, University of Extremadura, Avda. Universidad, s/n, 10003 Cáceres, Spain
[b] Dpt. Construction, Polytechnics School, University of Extremadura, Avda. Universidad, s/n, 10003 Cáceres, Spain

HIGHLIGHTS

- Surface radon exhalation rate from soil in itself was unable to explain indoor radon.
- Architectural style had a significant influence in the selected radon prone area.
- New dwellings have higher indoor radon concentrations than Traditional ones.
- Refurbished Traditional dwellings have higher indoor radon than Non-Refurbished.

GRAPHICAL ABSTRACT

ABSTRACT

Indoor radon is a major health concern as it is a known carcinogenic. Nowadays there is a trend towards a greater energy conservation in buildings, which is reflected in an increasing number of regulations. But, can this trend increase the indoor radon concentration? In this paper, we selected a radon prone area in Spain and focused on single-family dwellings constructed in a variety of architectural styles. These styles ranged from 1729 up to 2014, with varying construction techniques (from local resources to almost universally standard building materials) and regulations in force (from none to the Spanish regulation in force). The ^{226}Ra concentrations in soil and surface radon exhalation rates were rather similar in this area, mean values ranging 70–126 Bq/kg and 49–100 mBq/m^2·s, respectively. Indoor radon concentration was generally greater than the contribution from soil exhalation (surface exhalation rates), especially in New dwellings (1980–2014). Its concentration in dwellings built in the Traditional style (1729–1940) was significantly lower than in the new houses. This can be consequence of the air tightness of the dwellings as a consequence of the different regulations in force. In the period covered by the Traditional style, there was no regulation in force, and dwelling had loose air tight. Whereas in recent times, there are mandatory regulations assuring a better air tightness of the buildings. Refurbishment of Traditional dwellings also seems to increase the indoor radon concentration, as they must also comply with the regulations in force.

© 2017 Elsevier B.V. All rights reserved.

1. Introduction

Radon (^{222}Rn) is a radioactive gas, recognized by the World Health Organization as the second cause of lung cancer after that produced

* Corresponding author.
  E-mail address: fguillen@unex.es (J. Guillén).

by tobacco smoke (WHO, 2009). Its presence in the environment depends fundamentally on the geochemical nature of the soil, which determines the ^{226}Ra soil content (Baeza et al., 1994) and the permeability of them (Neznal and Neznal, 2005). Although radon concentration in atmospheric air do not usually reach significant concentrations (Arnold et al., 2009), radon can be accumulated in dwellings and its concentration can be high. Since people usually remain a high percentage of their lives indoors, it can become a serious problem from the point of view of radiological protection (Barros et al., 2015; Torres-Durán et al., 2014). Therefore, it is extremely important to know, or better yet, to predict accurately radon levels indoors. Indoor radon concentrations depend on a large number of factors (climate, building materials and construction methods, ventilation, etc.), in addition to the characteristics of the soils where they were built, which can produce a great dispersion of results. This is why a large number of studies have been carried out, from very different perspectives, in order to know the risk posed by high concentrations of radon in the interior of homes and to design solutions to reduce it (Frutos-Vázquez et al., 2011; Bartzis et al., 2012).

In fact, many countries have developed maps identifying areas more or less prone to high levels of indoor radon. Specifically, in Spain (García-Talavera et al., 2013a, 2013b) a predictive radon map has been elaborated based on the geological characteristics of the soils and on the measurements of the gamma dose rate that by external irradiation is received from these. Other radon maps have been constructed from direct measurements of the concentrations of radon within buildings (Miles, 1997; Andersen et al., 2001; Martin, 2004; Friedmann, 2012). In some of them, dwellings have been classified according to their potential exposure to radon in different categories: low

(<150 Bq/m^3); medium (between 150 and 300 Bq/m^3); and high (>300 Bq/m^3) (Martin, 2004).

Other studies have focused on whether it is possible to reduce the build-up of radon in homes. Among them, there is the European Union's RADPAR project (Bartzis et al., 2012), which concludes that in certain newly constructed and energy-efficient buildings, efforts to achieve greater energy savings are also effective in reducing the entry of radon into them. Similar results have been obtained in another study developed in Germany (Kemski et al., 2009), which confirms that the most modern buildings, built after 1995, have more moderate radon concentrations inside. The use of depressurization techniques based on underground sumps was also effective to reduce the indoor radon concentration (Frutos-Vázquez et al., 2011). However, concern about the consequences of high concentrations of indoor radon is relatively recent. In fact, in many countries such as Spain, the conclusions obtained from among others in those studies have not yet been transferred to the rules governing the construction of houses, to achieve an effective reduction of these levels in the areas that can exhale important radon activities from the soils.

One of the radon prone areas in Spain where there is a high risk that in the, measured annual mean value concentrations of indoor radon may exceed 300 Bq/m^3 (García-Talavera et al., 2013b) is located in the north of the province of Cáceres (Spain) (see Fig. 1), mainly due to the radioactive characteristics of their soils (Baeza et al., 1994). The most widespread type of housing in these regions, eminently rural, is single-family dwellings. These have been constructed, initially ignoring the existence of radon gas or its effects and therefore the need to take counter-measurements into account. At present, although there is
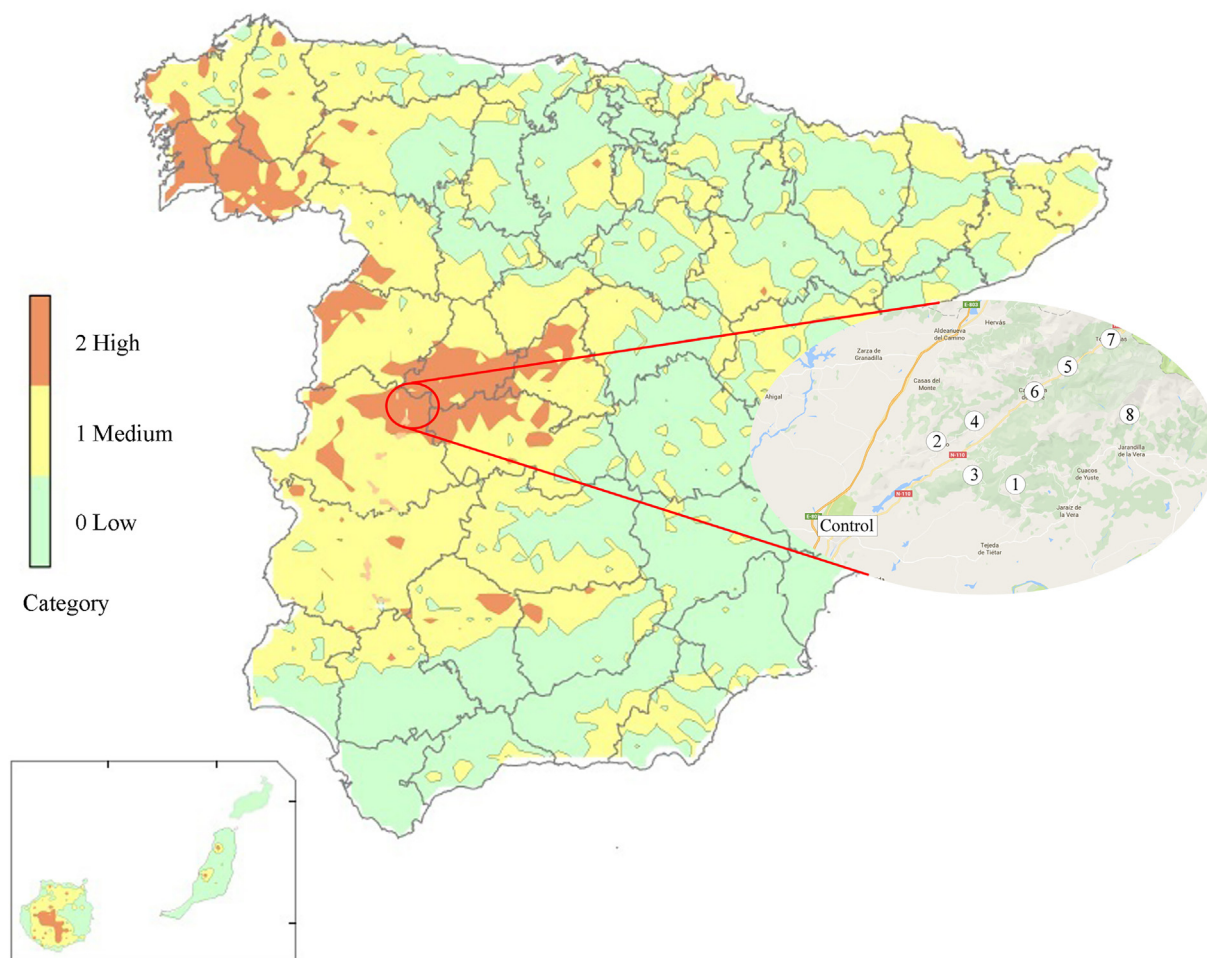

Fig. 1. Map of the selected locations. Modified from the Predictive Map of Radon Exposure in Spain (García-Talavera et al., 2013a). (Low exposure: Rn < 150 Bq/m^3; medium exposure: 150 < Rn < 300 Bq/m^3; and high exposure: Rn > 300 Bq/m^3.)

some concern for existence of radon inside dwellings, there are no measures currently in the building regulations in Spain to mitigate their effects.

The main goal of this study is to analyze whether the different constructive techniques for single-family dwellings, from about 1700 to the present day, in this radon prone area have any influence on indoor radon concentrations. Construction techniques and materials used have undergone remarkable changes along time, always adapted to the climatological conditions of these zones, but without any consideration about the possible occurrence of high concentrations of indoor radon, nor how to reduce them. Each single-family dwelling was constructed according to the regulations in force at that time, which was none for dwellings built in the Traditional style (before 1940) to the latest version of the Technical Building Code in 2006 (CTE, 2006). Although in no regulation in force in Spain the radon is considered, some other requirements such as ventilation and energy efficiency are, which can influence indoor radon concentration. Inside this radon prone area, nine locations were selected, eight in the Jerte and Vera valleys and a control location in the surroundings. In each of these locations, three single-family dwellings were selected and the whole set classified according to different architectural and construction techniques: a) Traditional (before 1940); b) Old (from 1940 to 1980); and New (after 1980).

2. Material and methods

2.1. Temporal evolution of single-family buildings in the north of Cáceres

Single-family dwellings in north of the province of Cáceres (Spain), see Fig. 1, can be classified into three large groups according to the

architectural style and construction techniques: a) Traditional, with a construction date prior to 1940; b) Old, construction took place between 1940 and 1980, approximately; and c) New, construction date after 1980.

The Traditional single-family dwelling (prior to 1940) is characterized by using only local materials in its construction, such as stone, mainly granite, and wood. Two types of Traditional dwellings can be identified: Half-timbered houses and Serrana houses (see Fig. 2a). In both of them, ground floor is in direct contact with the soil, or sometimes it was paved with boulders. Half-timbered houses occupy plots of up to 120 m^2, with thick walls and load walls that provide them reasonable thermal insulation (Pizarro, 1983). They have exterior masonry walls on the ground floor, which isolates them from soil moisture and walls with half-timbered and adobe filling in the remaining plants. Small holes can be generally observed in the building facade. Slabs are made of wood that rest on load walls and pavements are formed by boards nailed to the beams. The Serrana houses have smaller dimensions, up to 40 m^2 (Pizarro, 1983) and square shapes. They have thick masonry walls, generally of granite, in all floors. The rest of the construction is similar to the half-timbered, using local wood for slabs and its deck is covered again with terracotta tiles. Doors and windows were made of wood with no glass and low air tightness. Dwellings from this period did not follow any regulations, as none existed.

The Old dwellings (1940–1980) are characterized by the use of local materials, but also other more universal and industrialized materials, such as concrete (see Fig. 2b). It is a consequence of the generalized introduction of two types of techniques: i) the replacement of structures supported by heavy walls (>700 kg/m^2), by the use from 1940 of lighter reticular structures of pillars and beams of reinforced concrete; and ii)

a) Traditional Style (before 1940)

Traditional wooden half-timbered dwelling

Traditional "serrana" dwelling

b) Old Style (1940 - 1980)

c) New Style (after 1980)

Fig. 2. Examples of the different Architectural styles in the selected region: a) Traditional wooden half-timbered and 'Serrana' dwellings; b) Old; and New styles.
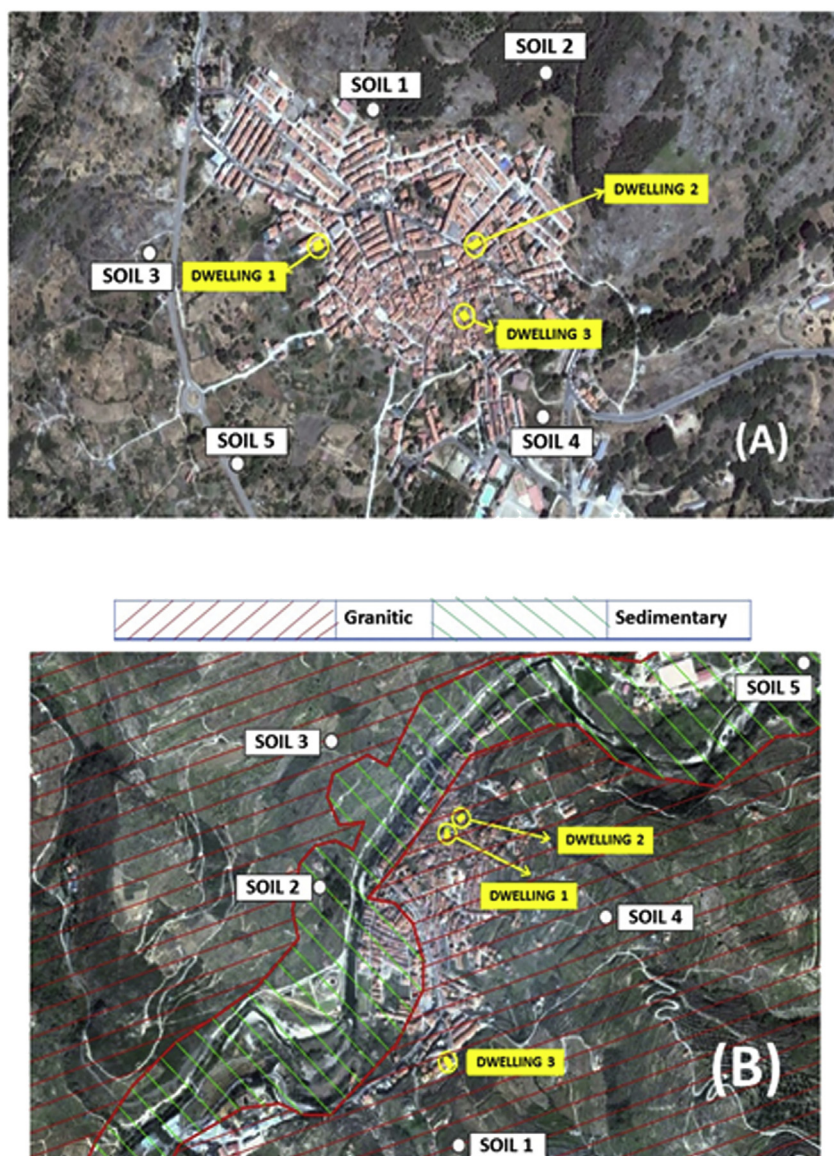
Fig. 3. Location of soil sampling points in the surroundings of two locations of the present study: A) location 1, with homogeneous granitic soil and B) location 6, with mixed of two geological components, granitic and sedimentary.

the change of passive conditioning systems by other electromechanical ones (Monjo, 2005). Facade is usually built of brick. Initially, between the 1940s and 1950s it acted as a loading wall of one foot thick, more recently it was formed by single hollow bricks. Soil was paved with tiles over cement screed. Roof systems are inclined, built on inclined or horizontal concrete slabs and covered by single hollow air brick, cement mortar and tile. Doors and windows were usually made of aluminum, but generally, not air tight. In 1973, the Technological Norms for Building (BOE, 1973) was in force and regulated different actions in the building process. There were also some regulations concerning sanitation and ventilation, but only applicable to blocks of flats, not single-house dwellings. In 1977, the Basic Building Norms (BOE, 1977) were in force, regulating the minimal conditions of livability in the houses. Later, regulations about thermal and acoustic conditions in the houses were included (BOE, 1979, 1981), increasing their tightness level.

Single-family dwellings built after 1980 are identified as New, see Fig. 2c. Construction techniques and materials are equal to those used in any other part of Spain. The ground floor lies on a boulder bed of variable thickness (depending on the terrain), then a waterproof polythene sheet, and a concrete layer reinforced with an electrically welded net. Regarding the building envelope, the most frequent vertical structure

system is reinforced by concrete pillars or loading walls with perforated ceramic brick with air chamber, thermal insulation and cement mortar lining. Horizontal structures are formed by reinforced concrete slabs. Roof is usually formed using a thermal insulator, bricks forming the corresponding pitch, a compression layer of cement mortar and finally tiles. All materials used for its construction pass a quality regulatory control, and techniques of execution are perfectly defined, both in the construction regulations and in the execution project. Basically, the main construction regulations in force in Spain are the Technical Building Code (CTE2006), which establishes the basic requirements for the quality of buildings and their facilities.

Refurbishment of houses is carried out taking into account the regulation in force at that time.

2.2. Sampling points

Indoor radon concentrations were determined in three single-family dwellings in each selected location, distant each other 10–20 km (see Fig. 1). In all of them, rooms located on ground floor were selected. In the surroundings of each location, surface soil (0–5 cm) was sampled in five different points in order to determine its radium concentration,

and radon in situ surface exhalation rates were also determined in the same sampling campaign. Fig. 3 shows the location of the selected dwellings and sampling points in two locations with different geological composition. As control dwelling, one corresponding to the time period identified as New was selected, and used to assess the seasonal variation of indoor radon by monthly determining it in a room located on ground floor. In order to minimize the influence of seasonal variation, in each sampling campaign, i.e. measurement in a dwelling in a location, the radon concentration of the dwelling and the control dwelling were determined in the same period. Comparison between short and long term indoor radon measurements was carried out in the control dwelling.

2.3. Radionuclide determination

The ^{226}Ra concentration in soil sample was determined by γ-spectrometry. An aliquot of 150 g was placed into 191-cm^3 Petri-type capsules and sealed to avoid loss of any ^{222}Rn emanations. After 28 days to allow ^{226}Ra to reach secular equilibrium with its descendants (^{214}Bi and ^{214}Pb), the samples were assayed by gamma spectrometry using a Ge(Hp) detector of 43% relative efficiency. IAEA Soil 6 was used as reference material to check the quality of the measurements. The overall quality control of these determinations is also guaranteed by the accreditation of the laboratory to carry out radioactivity assays in environmental samples according to UNE-EN ISO/IEC 17025 (ISO, 2005).

Radon concentration was determined using charcoal canisters according to the EPA procedure (EPA, 1987). Two canisters were placed in each location and exposed for about 2 days, recording start and end times. At the same time, a canister was placed in the control dwelling and exposed for the same period. This sequence (two canisters in location and one in control dwelling) was carried out during all the study. After exposure, canisters were sealed and transported to the laboratory (3 h after end of exposure) to be measured by γ-spectrometry using the equipment previously described. The ^{214}Pb (295.22 and 351.93 keV) and ^{214}Bi (609.31 keV) were systematically analyzed. The calibration of canister geometry was carried of spiking three blank canisters with a known amount of ^{226}Ra in each of them. Then, they were sealed, and efficiency determined one month later. The validity of the results was checked using a reference canister, reproducing its reference value. In order to check the quality of the radon determination by canister, in five dwellings the indoor radon concentration was determined simultaneously by an active detector, AlphaGuard (Saphymo GmbH). Its accuracy was checked in a proficiency test in field conditions and results were satisfactory (Gutiérrez-Villanueva et al., 2016). Radon exposure (concentration times exposure time) and mean value of radon concentration determined by these two methods were compared. The mean values and standard deviation of the ratio between canister and AlphaGuard were (0.96 ± 0.16 (S.D.)) and (0.90 ± 0.16 (S.D.)) for radon exposure and radon concentration, respectively.

Radon concentration in the control dwelling was also determined using long term exposure techniques, in particular electret ion chambers (E-Perm®) with different combinations electret-chamber adequate for several exposure times. Two electrets were monthly exposed during a year, simultaneously with electrets exposed for 3-months, 4-months and 1 year (1-month: electretLT and chamber S; 3-month: electretST and chamber L-OO; 4-month and 1-year: electret LT and chamber L-OO). Indoor radon concentration was calculated using the E-Perm® software, and the quality control was carried out using the reference blank and voltage electrets.

2.4. Surface radon exhalation

Surface radon exhalation was determined by means of the accumulation method described in the ISO/FDIS 11665-7 (ISO, 2012). The accumulation container has cylindrical shape with (5.88 ± 1%) L volume and 530.9 cm^2 open surface. The container had two orifices which were

available for continuous air circulation, and was coupled to the measurement device, AlphaGuard. The air circulation was maintained using an AlphaPump (Saphymo GmbH), which was kept at a low intensity flow-rate to avoid actively sucking radon from the sample.

3. Results and discussion

3.1. Soil parameters

Table 1 shows the mean values of ^{226}Ra concentration in surface soil (0–5 cm) and surface radon exhalation rate determined in each location. The ^{226}Ra concentration in soil is quite uniform in the area, being the mean value of (106 ± 21 (S.D.)) Bq/kg, within the range 70–126 Bq/kg. Therefore, the ^{226}Ra can be considered as homogeneous in this area. These activity levels are consistent with those previously reported in the map of natural radioactivity of Spain (MARNA) (CSN, 2000) and are also within the range 13–165 Bq/kg, reported for ^{226}Ra for soils of the whole province of Cáceres (Baeza et al., 1992). The ^{226}Ra soil concentration was also found to be constant with depth in this province (Baeza et al., 1994). The standard deviation of the ^{226}Ra concentrations reflected the geological variability of the soils in each location. They were mainly granitic, but in three locations there was also a sedimentary contribution due to the Jerte River. Surface radon exhalation rate was also similar in each location, being the mean value for all locations (66 ± 18 (S.D.)) mBq/m^2·s, within the range 9–160 mBq/m^2·s. This range is partially within the range 20–2100 mBq/m^2·s reported for surface soil exhalation rates in Syrian soils with ^{226}Ra concentration in the range 9–77 Bq/kg (Shweikani and Hushari, 2005), and above those reported for other possible sources of indoor radon: building materials, 0.0025–0.012 mBq/m^2·s (Immé et al., 2014) and polished granite, 0.0036–2.9 mBq/m^2·s (Guillén et al., 2014). The mean value of the surface exhalation rates is located in the upper part of the distribution functions for radon exhalation or radon flux derived from geological and meteorological databases for the elaboration of national and international predictive maps (López-Coto et al., 2013; Manohar et al., 2013; Karstens et al., 2015). In those studies the ^{226}Ra activity concentration in soil was estimated from total uranium concentration maps assuming secular equilibrium, in the range 26–55 Bq/kg, lower than those determined in our study area (see Table 1).

3.2. Annual variation of indoor radon concentration

As indoor radon concentrations show seasonal variations related to climatic conditions, it is necessary to correct the radon concentrations detected by charcoal canister, which had a very limited exposure time, about 2 days. Therefore, and simultaneously with the measurement of radon concentration in the selected locations, the radon concentration in the control dwelling was also determined using the same methodology. Fig. 4 shows the seasonal variation of indoor radon in the control dwelling during the duration of the measurements: using short term measurements (canister, 2 days, see Fig. 4a), and long term

Table 1
Mean value and standard deviation (1σ) of ^{226}Ra concentration in soil, expressed in Bq/kg, and surface radon exhalation rate, E, expressed in mBq/m^2·s, determined in the surroundings of each location. Geological background of soils in each location is also indicated.

Location code	Geological background	^{226}Ra (Bq/kg)	E (mBq/m^2·s)
1	Granitic	124 ± 16 (107–150)	65 ± 16 (42–87)
2	Granitic	123 ± 30 (93–165)	66 ± 27 (33–97)
3	Granitic	121 ± 15 (104–131)	49 ± 23 (20–83)
4	Granitic	126 ± 28 (93–160)	82 ± 53 (42–160)
5	Granitic/sedimentary	91 ± 26 (61–130)	52 ± 39 (9–93)
6	Granitic/sedimentary	110 ± 37 (77–166)	60 ± 59 (14–160)
7	Granitic/sedimentary	84 ± 16 (60–104)	100 ± 15 (88–120)
8	Granitic	70 ± 7 (61–77)	55 ± 23 (25–85)

measurements (electret, 1 month, see Fig. 4b). It can be observed that the indoor radon concentration varied within the range 110–730 Bq/m³ for short term measurements, and within the range 71–585 Bq/m³ for long term ones, showing the well-known seasonal variation, with maximum in winter and minimum in summer (Xie et al., 2015). The annual indoor radon concentration was determined by fitting the experimental data to Eq. (1).

$$Rn(t) = Rn(annual) + A \cdot \sin\frac{2\pi}{T}(t-t_0) \qquad (1)$$

where $Rn(t)$ is the indoor radon concentration at time t, expressed in Bq/m³; $Rn(annual)$ is annual indoor radon concentration, expressed in Bq/m³; A is the amplitude of the seasonal variations, expressed in Bq/m³; t is the time, expressed in days; T is the period, expressed in days; and t_0 is the initial phase, also expressed in days. For fitting purposes, the origin of time was fixed at the beginning of 2016. Table 2 shows the results for the fitted parameters for the two sets of measurements. The annual indoor radon concentration in the control dwelling was (332 ± 24) Bq/m³ using canisters, and (246 ± 27) Bq/m³ for 1-month electrets. The short term/long term ratio (canister/electret) was 1.35 ± 0.18, which suggested that the annual mean value derived using

Table 2
Fitting parameters seasonal variation of indoor radon concentration in the control dwelling (see Eq. (1)) using short term (canister: 2 days) and long term (electret: 1 month) measurements. Rn(annual), annual radon concentration; A, amplitude; T, period; and t_0, initial phase. N: number of experimental data.

Fitting parameters		
Exposure time	Short term (2 days)	Long term (1 month)
Rn(annual) (Bq/m³)	332 ± 24	246 ± 27
A (Bq/m³)	230 ± 31	209 ± 39
T (day)	352 ± 26	351 ± 32
t_0 (day)	268 ± 9	217 ± 10
R^2	0.7556	0.7077
N	18	12

canister values was slightly greater than the one using electrets. But, the two values were around the 300 Bq/m³ annual mean reference level used in the radon predictive map (see Fig. 1). Table 3 shows the indoor radon concentration determined in the control dwelling using 3-months, 4-months and 1-year electrets. The annual indoor radon mean values were 267 ± 86 (S.D.) Bq/m³ for 3-month electrect, 250 ± 100 (S.D.) Bq/m³ for 4-month electrect, and 245 ± 15 for 1-year electret. As it can be seen, the annual mean value was fairly consistent using different long term exposures.

The influence of seasonal conditions (ratio between amplitude and annual mean value) can vary the indoor radon in about 69–85%, which is consistent with the observed experimental range. The period was approximately one year for short and long term measurements, reflecting the seasonal climatic conditions over the year.

3.3. Indoor radon concentration and architectural type

Radon exhaled from soil may be a significant source of indoor radon. In order to analyze whether the surface exhalation rate determined in the different location was a key contributor to the indoor radon concentration, it was estimated using the following approach:

$$C_{Exh}\left(\frac{Bq}{m^3}\right) = \frac{E_h \cdot S_r}{V_r \cdot \lambda_V} \qquad (2)$$

where C_{Exh} is the indoor radon concentration due to exhalation from surface soil, expressed in Bq/m³; E_h is the surface exhalation rate, expressed in Bq/m²·s; S_r is the room surface, expressed in m²; V_r is the room volume, expressed in m³; and λ_v is the air exchange rate, expressed in s⁻¹. Soil exhalation contribution to the indoor radon concentration was calculated for each dwelling using the corresponding values of room surface and volume. The mean value of the room surface was 17.54, 28.26 and 44.43 m² for Traditional, Old and New architectural styles; while the height ranged from 2 to 3 m for all styles. The air exchange rate was considered to be 0.63 h⁻¹ (about 1.75 · 10⁻⁴ s⁻¹), as typical for a room with ventilation (Bearg, 1993). In this approach, a constant value for the air exchange rate was considered because the focus was on the influence of the surface exhalation rate from soil. As Table 1 shows the range of variation for soil surface exhalation rates, a range of indoor radon due to this source was estimated for each dwelling. Fig. 5 shows the contribution of soil exhalation to the indoor radon
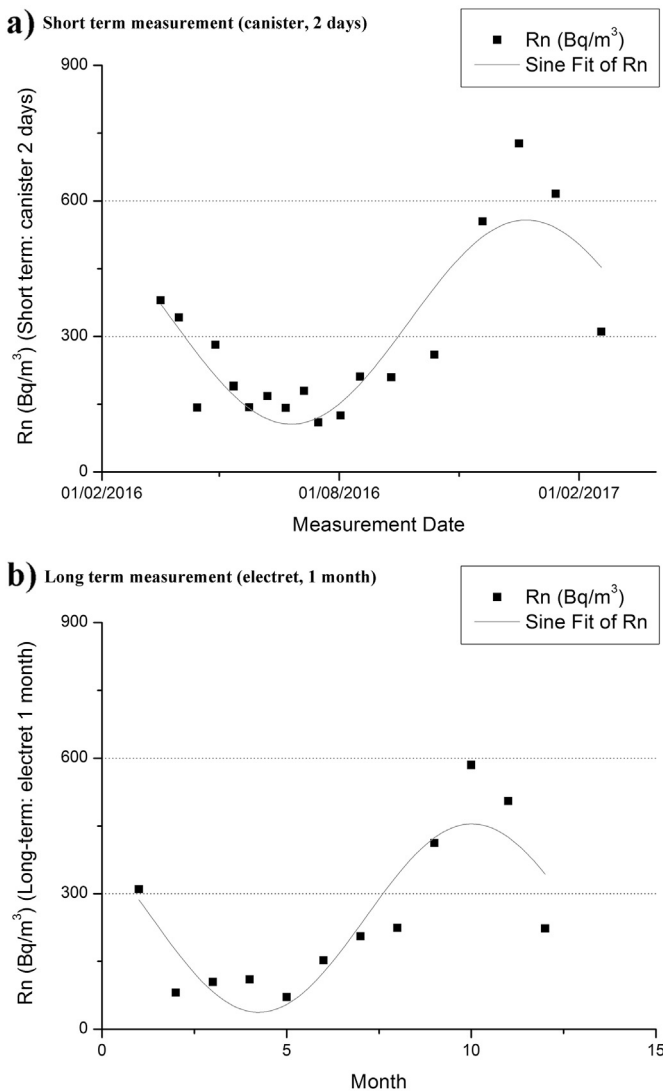

Fig. 4. Annual variation of radon concentration, expressed in Bq/m³, in the control dwelling using a) short term exposure (canister, 2 days), and b) long term exposure (electret, 1 month).

Table 3
Indoor radon concentration in the control dwelling, expressed in Bq/m³, determined using 3-month, 4-month, and 1 year exposure time electrets.

Electret 3-months		Electret 4-months		Electret 1-year	
Exposure	Rn (Bq/m³)	Exposure	Rn (Bq/m³)	Exposure	Rn (Bq/m³)
Mar–May	235 ± 12	Mar–Jun	196 ± 10	Mar–Feb	245 ± 15
Jun–Aug	165 ± 8	Jul–Oct	185 ± 10	–	–
Sep–Nov	305 ± 28	Nov–Feb	369 ± 19	–	–
Dec–Feb	364 ± 18	–	–	–	–

concentration in the selected dwellings. It can be observed that for most dwellings the indoor radon concentration was far greater than the contribution only due to soil exhalation. This implied that the surface exhalation rates did not account for the indoor radon exhalation by itself. The ratio between the mean value of exhaled radon and the indoor radon concentration was 0.82 for Traditional, 0.34 for Old and 0.31 for New dwellings. This suggested an influence of the architectural type, as this ratio for new houses was smaller than for the other types. Ground floor in Traditional and Old architectural types was on directly on soil or on a layer of boulders or cement; while it has a more complex construction in the New type.

The air exchange rate in each dwelling, λ, was also estimated using a slight modification of Eq. (2), just replacing the estimated indoor radon concentration, C_{Exh}, by the actual radon concentration measured in each dwelling. As the latter was determined using the canister method, λ was the average air exchange rate during the same period, 2 days (see Table 4). Fig. 6 shows the relationship between indoor radon concentration and air exchange rates for all studied dwellings in the different location (experimental data in Table 4), which fitted to the following expression, reproducing Eq. (2):

$$\text{Indoor Radon } \left(\frac{Bq}{m^3}\right) = (80 \pm 7) \cdot \lambda^{-(1.04 \pm 0.06)}; R^2 = 0.886 \qquad (3)$$

The mean value of the air exchange rates for the Traditional dwellings without refurbishment was $(0.7 \pm 0.5 \text{ (S.D.))} \text{ h}^{-1}$, which is close to the value 0.63 h^{-1} previously used for ventilated rooms. It should be noted that these air exchange rates were for closed rooms, as canister radon method requires the rooms to be closed 48 h before exposure and during it. The two refurbished dwellings presented lower λ values, 0.16–0.26 h^{-1}, which were similar to the mean value for the New dwellings $(0.32 \pm 0.35 \text{ (S.D.))} \text{ h}^{-1}$.

The indoor radon concentration in the different dwellings were determined at different times, as their availability and accessibility was also different (determined by the owners). In order to minimize the influence of seasonal variability on the analysis according to the different architectural styles, two approaches were used: i) the ratio of the indoor radon concentration between the different dwelling and the oldest dwelling in a given location, removing in this way the seasonal variation in each location, but information about architectural style and date of construction of the dwellings cannot be considered; and ii) the normalization of their concentration, in which these data can be used after seasonal variability analysis and estimation of the normalized indoor radon concentration.
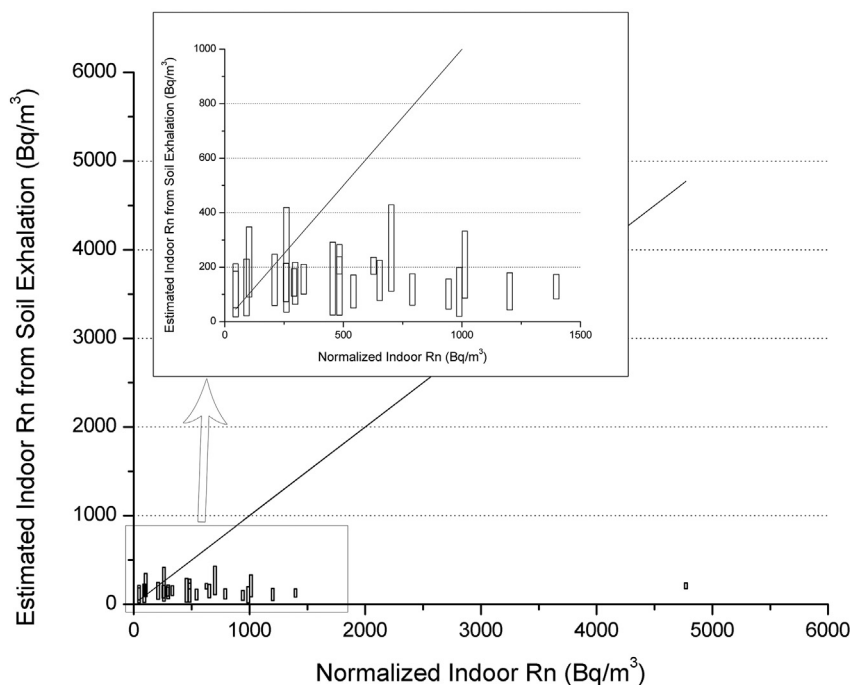
Table 4
Radon concentration, expressed in Bq/m^3, detected in the studied dwellings and in the control one during the same exposure time. Estimation of air exchange rate, expressed in h^{-1}, for each dwelling based on the room dimensions, soil surface exhalation rates and indoor radon concentration. The construction date and architectural style is detailed for each dwelling.

Location	Construction date	Style	Control Rn (Bq/m^3)	Dwelling Rn (Bq/m^3)	λ (h^{-1})
1	1820	Traditional	177 ± 18	156 ± 18	0.59 ± 0.19
	1955	Old	253 ± 14	253 ± 16	0.39 ± 0.10
	2009	New	215 ± 12	910 ± 55	0.090 ± 0.023
2	1850	Traditional	273 ± 16	212 ± 16	0.43 ± 0.18
	1997	New	233 ± 15	457 ± 29	0.21 ± 0.09
	2007	New	127 ± 7	302 ± 19	0.25 ± 0.10
3	1780	Traditional	327 ± 21	45 ± 7	1.75 ± 0.9
	1930	Traditional	992 ± 55	628 ± 44	0.15 ± 0.07
	2007	New	438 ± 28	1590 ± 430	0.042 ± 0.022
4	1750[a]	Traditional	253 ± 17	540 ± 130	0.26 ± 0.18
	1895	Traditional	357 ± 24	110 ± 9	1.0 ± 0.7
	2014	New	293 ± 15	892 ± 46	0.12 ± 0.08
5	1815[b]	Traditional	143 ± 8	426 ± 42	0.16 ± 0.12
	1981	New	342 ± 18	47 ± 3	1.4 ± 1.0
	1994	New	380 ± 19	105 ± 6	0.8 ± 0.6
6	1775	Traditional	281 ± 14	220 ± 13	0.4 ± 0.4
	1998	New	144 ± 8	197 ± 11	0.3 ± 0.3
	2010	New	189 ± 10	175 ± 15	0.24 ± 0.24
7	1729	Traditional	142 ± 8	206 ± 11	0.61 ± 0.10
	1990	New	180 ± 10	340 ± 18	0.37 ± 0.06
	1997	New	168 ± 9	2410 ± 130	0.054 ± 0.008
8	1840	Traditional	125 ± 8	112 ± 7	0.8 ± 0.3
	1975	Old		204 ± 13	0.34 ± 0.14
	2000	New		355 ± 21	0.18 ± 0.08

[a] Refurbished in 1986.
[b] Refurbished in 1990.

Fig. 5. Contribution of soil surface exhalation rate (in range) to the normalized indoor radon concentration in the selected buildings (see Eq. (3)). Solid line marks the situation in which both radon concentrations are equal. A constant value of air exchange rate (0.63 h^{-1}) was considered to derive the estimated indoor radon concentration from soil exhalation rates.
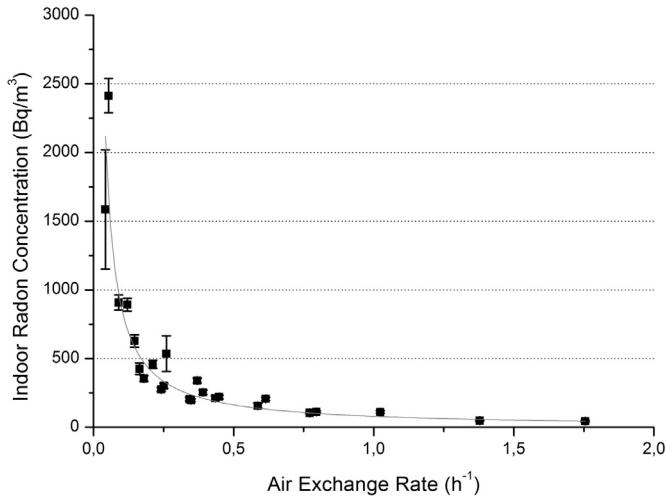
Fig. 6. Relation between air exchange rate, in h^{-1}, and indoor radon concentration, expressed in Bq/m^3 in dwellings for all locations.

Fig. 7 shows the ratio between the indoor radon concentration in the oldest dwelling in each location and the others. As dwellings built in the middle period between the oldest and newest dwellings belonged to different architectural styles in each location, we opted for considering them as medium in this approach. All dwelling in the oldest and newest groups belonged to the Traditional and New architectural styles, respectively. By definition, all ratios in the oldest group were 1. The radon ratio in the newest dwellings presented was higher than 1 in all locations, but in location 5, in which the oldest dwelling was refurbished. Therefore, it implied an increase in the indoor radon concentration in the New dwellings regarding the Traditional ones.

The normalized indoor radon concentration for the location i, Rn$_{norm}$(i), was calculated according to Eq. (4).

$$Rn_{norm}(i) = Rn(t, i) * \frac{Rn(annual, control)}{Rn(t, control)} \quad (4)$$

where Rn(t, i) is the indoor radon concentration at time t and location i, expressed in Bq/m^3; Rn(annual, control) is the annual indoor concentration in the control dwelling, (332 ± 24) Bq/m^3; and Rn(t, control) is the indoor radon concentration in the control dwelling at the same time t, expressed in Bq/m^3. Experimental data from short term canister measurements were used in Eq. (4). The Rn$_{norm}$(i) values would be
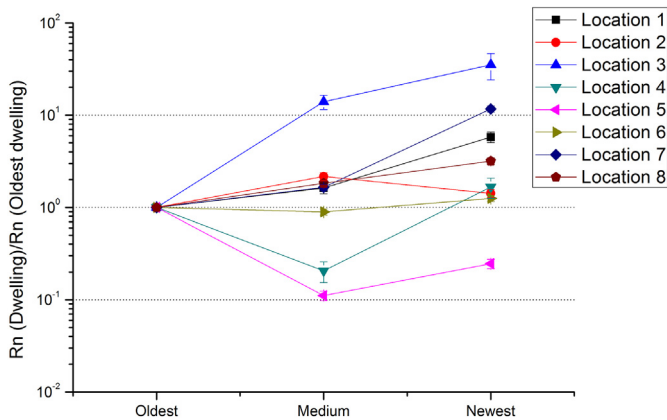

Fig. 7. Ratio between the indoor radon concentration in the selected dwellings and the oldest dwelling in each location. In this approach, the oldest dwellings in each location were considered in the Oldest group, the newest in the Newest group, and the remaining in the Medium group. The oldest dwellings (Traditional style) in locations 4 and 5 were refurbished in the New style.

Table 5
Mean value and range of the normalized indoor radon in dwellings grouped by architectural type. N = number of dwellings considered.

Architectural type	Date of construction	N	Normalized annual indoor Rn (Bq/m^3)
Traditional[a]	1729–1939	10	360 (46–990)
Old	1940–1980	2	440 (330–540)
New	1981–2014	12	1040 (46–4800)

[a] Two dwellings were built in 1750 and 1815 according to Traditional type, but they were refurbished in 1986 and 1990 respectively, according to the current legislation at that time.

equivalent to an estimation of the annual mean value of indoor radon in the studied dwellings. It should be noted that in Eq. (2), a similar seasonal variation in indoor radon in the control dwelling and those in the different locations was assumed, as all of them are located in the same area.

Table 5 shows the mean values and ranges of the normalized indoor radon concentration in the different locations grouped according the architectural type. It can be observed that the mean value of the indoor radon concentration generally increased as the construction date of the dwelling is more recent. In each architectural type, low indoor radon values can be found. However, the percentage of occurrence of concentrations higher than 300 Bq/m^3 also increased in the same way, being 30, 100 and 83% for the Traditional, Old and New architectural types, respectively. Fig. 8 shows the relationship between the construction date of the dwelling and the indoor radon concentration. It can be observed clearly an increase in indoor radon concentration in more recent dwellings. However, in two locations dwellings belonging to the Traditional architecture (built in 1750 in location 4 and in 1815 in location 5) were completely refurbished in 1986 and 1990 respectively, according to the existing construction regulations in force at that time. This seems to imply an increase of the indoor radon concentration due to more restrictive regulations concerning the air tightness of the dwellings, related with noise and energy conservation aspects in the regulations.
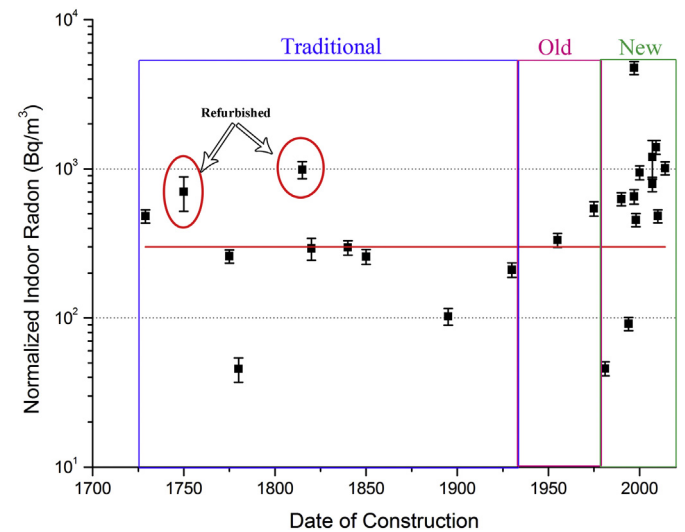

Fig. 8. Relation between the normalized indoor radon concentration, expressed in Bq/m^3, and the date of construction of single-family dwellings in the considered radon prone area. Experimental data are grouped according the dwelling architectural type (Traditional, Old and New). Two Traditional dwellings, built in 1750 and 1815, were refurbished in 1986 and 1990 respectively, according to the current legislation at that time. Solid line shows the reference limit of 300 Bq/m^3.

4. Conclusion

The indoor radon concentration in different single-family dwellings, ranging about 200 years, in a radon prone area in Spain was determined. These dwellings can be classified according to architectural and construction techniques in Traditional (before 1940), Old (1940–1980) and New (after 1980). Indoor radon concentration was determined in them, along with other ^{226}Ra concentration in soil and surface exhalation rate. Seasonal variation of indoor radon concentration was considered and all determinations were normalized to annual estimates. We have shown that:

• The concentration of ^{226}Ra in the selected locations was similar, and surface radon exhalation rates from soil cannot explain in themselves the indoor radon concentration.
• Architectural styles present in this area had an influence on the indoor radon concentration. Its concentration was higher in dwellings of recent construction, as the regulations in force apply criteria for energy conservation and noise reduction that tends to increase the air tightness of the dwellings (and consequently reduction of air exchange rates). Traditional dwellings on the same locations, with poor air tightness, present lower indoor radon concentration.
• Refurbishment of Traditional dwellings increased the indoor radon concentration, as they have to meet the quality criteria of the regulations in force at the moment, being the same as the New style, with similar air exchange rates.

Acknowledgements

References

Andersen, C.E., Ulbak, K., Damkjær, A., Kirkegaard, P., Gravesen, P., 2001. Mapping indoor radon-222 in Denmark: design and test of the statistical model used in the second nationwide survey. Sci. Total Environ. 272, 231–241.

Arnold, D., Vargas, A., Ortega, X., 2009. Analysis of outdoor radon progeny concentration measured at the Spanish radioactive aerosol automatic monitoring network. Appl. Radiat. Isot. 67, 833–838.

Baeza, A., del Rio, M., Miró, C., Paniagua, J., 1992. Natural radioactivity in soils of the province of Cáceres (Spain). Radiat. Prot. Dosim. 45, 261–263.

Baeza, A., del Río, M., Miró, C., Paniagua, J., 1994. Natural radionuclide distribution in soils of Cáceres (Spain): dosimetry implications. J. Environ. Radioact. 23, 19–37.

Barros, N., Field, D.W., Steck, D.J., Field, R.W., 2015. Comparative survey of outdoor, residential and workplace radon concentrations. Radiat. Prot. Dosim. 163 (3), 325–332.

Bartzis, J., Arvela, H., Bochicchio, F., Collignan, B., Fojtikova, I., Gray, A., Grosche, B., Holmgren, O., Hulka, J., Jiranek, M., Kalimeri, K., Kephalopoulos, S., Klerkx, J., Kreuzer, M., McLaughlin, J., Mueller, S., Olsen, B., Reina, V., Ringer, W., Rovenska, K., Schlesinger, D., Standring, W., Strand, T., Venoso, G., Zeeb, H., 2012. Radon Prevention and Remediation, RADPAR, Final Scientific Report. http://web.jrc.ec.europa.eu/radpar/.

Bearg, D.W., 1993. Indoor Air Quality and HVAC Systems. CRC Press, p. 64 (ISBN 0-87371-574-8).

BOE, 1973. Decreto 3565/1972, de 23 de diciembre, por el que se establecen las Normas Tecnológicas de Edificación, NTE. Boletín Oficial del Estado n° 13. España, de 15 enero 1973. pp. 752–756 (in Spanish).

BOE, 1977. Real Decreto 1650/1977, de 10 de junio, sobre normativa de la edificación. Ministerio de vivienda, Boletín Oficial del Estado n° 163. España, de 9 de julio de 1977. pp. 15443–15444 (in Spanish).

BOE, 1979. Real Decreto 2429/1979, de 8 de julio, por el que se aprueba la Norma Básica de Edificación NBE-CT-79, sobre condiciones térmicas del edificio. Boletín Oficial del Estado n.° 253. España, de 22 de octubre de 1979. pp. 24524–24550 (in Spanish).

BOE, 1981. Real Decreto 1909/1981, de 24 de julio, por el que se aprueba la Norma Básica de Edificación NBE-CA-81, sobre condiciones acústicas del edificio. Boletín Oficial del Estado n.° 214. España, de 7 de septiembre de 1981. pp. 20556–20577 (in Spanish).

CSN, Consejo de Seguridad Nuclear, Proyecto Marna, 2000. Mapa de radiación gamma natural. Colección Informes Técnicos 5. Consejo de Seguridad Nuclear, Madrid, Spain (in Spanish).

CTE, 2006. Real Decreto 314/2006, de 17 de marzo, por el que se aprueba el Código Técnico de la Edificación. Boletín Oficial del Estado n.° 74 (Spain. in Spanish).

EPA, U.S. Environmental Protection Agency, 1987. EPA 520/6-87-005. EERF Standard Operating Procedures for Radon-222 Measurement Using Charcoal Canisters (Montgomery, Alabama, U.S.A.).

Friedmann, H., 2012. The Austrian Radon Project (ARP). http://homepage.univie.ac.at/harry.friedmann/Radon/onrape.htm (Retrieved 30/07/2012).

Frutos-Vázquez, B., Olaya-Adán, M., Quindós-Poncela, L.S., Sainz-Fernández, C., Fuente-Merino, I., 2011. Experimental study of effectiveness of four radon mitigation solutions, basedon underground depressurization, tested in prototype housing built in a highradon area in Spain. J. Environ. Radioact. 102, 378–385.

García-Talavera, M., García-Pérez, A., Rey, C., Ramos, L., 2013a. Mapping radon-prone areas using γ-radiation dose rate and geological information. J. Radiol. Prot. 33, 605–620.

García-Talavera, M., Martín, J.L., Gil, R., García, J.P., Suárez, E., 2013b. El mapa predictivo de exposición al radón en España. Colección de Informes Técnicos n° 38.2013. Consejo de Seguridad Nuclear (in Spanish).

Guillén, J., Tejado, J.J., Baeza, A., Corbacho, J.A., Muñoz, J.G., 2014. Assessment of radiological hazard of commercial granites from Extremadura (Spain). J. Environ. Radioact. 132, 81–88.

Gutiérrez-Villanueva, J.L., Sainz, S., Fuente, I., Quindós, J., Quindós, L., Celaya, S., Fernández, A., López, D., Fernández, E., Quindós, L.S., 2016. Intercomparación de monitores de gas radón en condiciones de campo. Saelices el Chico (España). Universidad de Cantabria, Santander, Spain (in Spanish).

Immé, G., Catalano, R., Mangano, G., Morelli, D., 2014. Radon exhalation measurements for environmental and geophysics study. Radiat. Phys. Chem. 95, 349–351.

ISO, 2005. ISO/IEC 17025:2005. Requisitos generales para la competencia de los laboratorios de ensayo calibración (in Spanish).

ISO, 2012. ISO/FDIS 11665-7. Measurement of Radioactivity in the Environment - Air: Radon-222 - Part 7: Accumulation Method for Estimating Surface Exhalation Rate.

Karstens, U., Schwingshackl, C., Schmithüsen, D., Levin, I., 2015. A process-based ^{222}radon flux map for Europe and its comparison to long-term observations. Atmos. Chem. Phys. 15, 12845–12865.

Kemski, J., Klingel, R., Siehl, A., Valdivia, M., 2009. From radon hazard to risk prediction-based on geological maps, soil gas and indoor measurements in Germany. Environ. Geol. 56, 1269–1279.

López-Coto, I., Mas, J.L., Bolivar, J.P., 2013. A 40-year retrospective European radon flux inventory including climatological variability. Atmos. Environ. 73, 22–33.

Manohar, S.N., Meijer, H.A.J., Herber, M.A., 2013. Radon flux maps for the Netherlands and Europe using terrestrial gamma radiation derived from soil radionuclides. Atmos. Environ. 81, 399–412.

Martin, J., 2004. Concentración de radón en viviendas españolas. Otros estudios de radiación natural. Colección de Informes Técnicos 13.2004. Consejo de Seguridad Nuclear (in Spanish).

Miles, J., 1997. Development of maps of radon-prone areas using radon measurements in houses. J. Hazard. Mater. 61, 53–58.

Monjo, J., 2005. La evolución de los sistemas constructivos en la edificación. Procedimiento para su industrialización. Inf. Constr. 57, 499–500 (in spanish).

Neznal, M., Neznal, M., 2005. Permeability as an important parameter for radon risk classification of foundation soils. Ann. Geophys. 48, 175–180.

Pizarro, F.J., 1983. Arquitectura popular y urbanismo en el Valle del Jerte. Caja de ahorros de Plasencia, Los Santos de Maimona, Badajoz (in Spanish).

Shweikani, R., Hushari, M., 2005. The correlations between Radon in soil gas and its exhalation and concentration in air in the southern part of Syria. Radiat. Meas. 40, 699–703.

Torres-Durán, M., Ruano-Ravina, A., Parente-Lamelas, I., Leiro-Fernández, V., Abal-Arca, J., Montero-Martínez, C., Pena-Álvarez, C., González-Barcala, F.J., Castro-Añón, O., Golpe-Gómez, A., Martínez, C., Mejuto-Martí, M.J., Fernández-Villar, A., Barros-Dios, J.M., 2014. Lung cancer in never-smokers: a case–control study in a radon-prone area (Galicia, Spain). Eur. Respir. J. 44, 994–1001.

WHO, World Health Organization, 2009. In: Zeeb, Hajo, Shannoun, Ferid (Eds.), Handbook on Indoor Radon. A Public Health Perspective. 2009 (France).

Xie, D., Liao, M., Kearfott, K.J., 2015. Influence of environmental factors on indoor radon concentrationlevels in the basement and ground floor of a building - a case study. Radiat. Meas. 82, 52–58.