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Methodology for determination of radon prone areas combining the definition of a representative building enclosure and measurements of terrestrial gamma radiation

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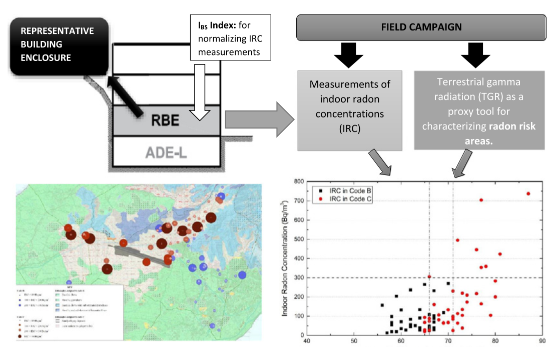
HIGHLIGHTS

- A new methodology for determining radon prone areas has been developed.
- Radon prone areas can be defined by local gamma radiation and building features.
- New definition of a Representative Building Enclosure for indoor radon measurements
- High density of gamma radiation and indoor radon measurements in two study areas
- A new Building Storey Index definition for normalizing indoor radon measurements

GRAPHICAL ABSTRACT



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ABSTRACT

The recommendations of the European Atomic Energy Community (EURATOM) have recently been incorporated into Spanish regulations in the Basic Document of Health Standards of the Technical Building Code (CTE), section HS6, on protection against radon exposure. This further accentuates the need to delimit radon prone areas as a strategy to address measures which minimise the effects of this gas on the population. In this research, measurements of terrestrial gamma radiation and indoor radon of dwellings have been carried out in the same location to delimit these risk areas. A new methodology has been developed including a definition of a Representative Building Enclosure (RBE) and it is proposed a Building Storey Index (I_{BS}) which allows normalizing measurements of indoor radon activity concentration taken in different levels from the ground to the RBE. The results show the need to consider the type of contact that exists between the building and the ground as a determining factor of radon risk. Terrestrial gamma radiation is used as a proxy for radioisotopic composition of soils to characterise the indoor radon risk at different geological formation.

1. Introduction

Under normal conditions, ^{222}Rn from the ^{238}U decay chain constitutes the largest source of natural exposure for human beings (50% of total natural radiation). The danger of exposure to high concentrations

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of radon does not come from the gas itself, but from its short term decay products that are alpha particles emitting elements in solid phase (^{218}Po, ^{214}Pb, ^{214}Bi and ^{214}Po). These products, whether free or attached to atmospheric aerosols, have a high probability of disintegrating in the human lungs before being eliminated, increasing the risk of cancer (Health Protection Agency, 2009). Exposure to radon occurs mainly by inhalation in poorly ventilated rooms, but radon and its products can also be assimilated by ingestion, either dissolved in water or through the consumption of vegetables and most notably tobacco. In consequence, long-term exposure to high concentrations of indoor radon (^{222}Rn) constitutes a public health problem. Radon has been categorised as a Class 1 carcinogenic agent by the International Agency for Research on Cancer (IARC) because it is the second most important cause of lung cancer after tobacco. The World Health Organization (WHO) asserts that long-term exposure to high levels of radon concentration is closely correlated to lung cancer according to several epidemiological studies. They urge institutions to take measures in order to limit radon concentration levels in buildings to minimise the exposure of the population (World Health Organization, 2015).

To mitigate the negative impacts of public exposure to high levels of radon concentration, Directive 2013/59/EURATOM of the Basic Safety Standard (BSS) of the European Union (EU) established that 'Member States shall establish a national action plan that addresses long-term risks from exposure to radon in homes, buildings with public access and workplaces for any source of radon entry, whether from soil, building materials or water'. This directive suggests corrective measures which minimise indoor radon gas concentration and a reference level of 300 Bq/m^3 of radon concentration is established. In December 2019, RD 732/2019 which modifies the Technical Building Code (CTE) was passed in Spain. It was a transposition of the European directive and it supposes a new Section HS6 in the Basic Document of Health Standards of the CTE (Ministerio de Fomento, 2019) on Protection from Radon Exposure. This section, apart from confirming the reference level of 300 Bq/m^3, includes a national map establishing municipal risk zones. However, it is pending approval through the 'Real Decreto por el que se aprueba el Reglamento sobre protección de la salud contra los riesgos derivados de la exposición a las radiaciones ionizantes' (Ministerio de Presidencia y Administraciones Territoriales, 2018), which establishes control criteria in and a maximum level of radon concentrations.

To apply this legislation in a territory, it is necessary to identify radon prone areas defined as those where the probability of finding high activity concentration of indoor radon is above a determined limit. This would allow a good assessment of the radon risk for a population in order to establish priority action areas. It has been necessary to establish maps where radon-prone areas are defined in order to achieve the goals established by the European Directive and national regulations. Different approaches are being used to establish the radon-prone areas of determined geographical zones (García-Talavera and López-Acevedo, 2019):

- Direct measurement of Indoor Radon Concentration (IRC) by placing alpha track detectors in buildings.
- Indirect methods based on variables correlated to radon like radon gas from soils (Kemski et al., 2001; Barnet and Fojtíková, 2008), gamma radiation (García-Talavera et al., 2013) or the uranium content of rocks (Arnedo et al., 2017; Ielsch et al., 2010).
- Hybrid methods which combine direct measurements with geological or radiological variables (Kemski et al., 2009; Cinelli et al., 2011).

In Spain, the Nuclear Safety Council (CSN) has employed two defined strategies to develop an indoor radon risk map of the Canary Islands (García-Talavera and López-Acevedo, 2019). However, they did not adopt the strategy realised for the mainland map, not taking into account the analysis based on gamma radiation data from Project MARNA (Suárez-Mahou et al., 2000). This was possible given the higher density of indoor radon measurements available in the islands. Six of the eight islands (with lower populations) were identified as non-priority action areas because the available measurements were sufficiently below the reference level. Nevertheless, it is important to highlight that the western islands have significantly lower number indoor radon measurements than the eastern islands. The main islands (Tenerife and Gran Canaria) have been analysed by grouping areas according to the chemical composition of lithostratigraphic units of islands (García-Talavera and López-Acevedo, 2019) and applying statistical criteria developed by Garcia-Talavera San Miguel et al. (2013).

Section HS6 of the Basic Document of Health Standards of CTE is based on the methodology and results shown in the work of García-Talavera and López-Acevedo (2019). It classifies Spanish municipalities into two levels according to their radon risk potential: zone I (moderate risk) and zone II (high risk). In Tenerife and Gran Canaria (Canary Islands, Spain), 50 of 52 municipalities are in zone II. Thus 99.4% of the whole population of both islands (83.8% of the population of the Canary Islands) would be affected by the maximum risk level established by regulation, so that, among other requirements, an additional system like a ventilated containment space or ground depressurisation system will be required for new buildings from the approval date.

This paper presents the methodology followed to determine the radon-prone areas in two municipalities of the Tenerife and Gran Canaria islands, belonging to the Spanish archipelago of the Canary Islands.

This work is structured as follows. In Section 2, we will describe the factors which our research is based on. In Section 3, the geology of the area of study will be presented in detail. The instrumentation and methods adopted during the measurement campaigns are reported in Section 4. Section 5 is devoted to the results of our study, including the treatment of statistical uncertainties. Finally, conclusions are drawn in Section 6.

2. Factors influencing IRC

Indoor radon levels are a function of several factors: geology of the area, taking into account the activity concentration of ^{226}Ra of different types of rocks which is the immediate origin of ^{222}Rn in the disintegration chain; the permeability of the ground which is closely correlated with transport and entry of radon into buildings (Font, 1997); building typology, including building material and the permeability of the envelope of the building in contact with the ground (Frutos Vázquez, 2009); the presence and type of basement or chamber below the house; factors like atmospheric pressure or temperature which allow a dominant driving force for radon entry according to differential pressures (Hintenlang and Al-ahmady, 1992); and domestic lifestyles. These factors can be divided into intrinsic permanent (a) and extrinsic variable (b) factors:

a) *Intrinsic permanent factors*:

- Natural: geology (the radon strength of the soil).
- Permeability of the envelope of the building in contact with the ground.

b) *Extrinsic variable factors*:

- Natural: weather and climate factors.
- Artificial: ventilation of the building enclosure depending on the habits of the occupants.

This work will be focused on studying the influence of intrinsic permanent factors in the presence of indoor radon, such as the characteristics of soils and rocks that buildings sit on and the typology of the building and level of contact with the ground.

3. Geology of the area of study

The Canary archipelago has a volcanic origin with geological formations over 30 million years old and it extends to approximately 500 km.

The volcanic rocks of the Canary Islands belong to the alkaline igneous series, in this case, associated intraplate volcanism (Carracedo et al., 2002). This igneous series is formed by a sequence of rocks whose composition evolves from undifferentiated terms represented by basalts, intermediate terms represented by trachybasalts, and finally differentiated terms represented by trachytes and phonolites. In order to analyse the relationship between the geology and concentration of radon activity in the soils, a simplified classification, based on the geochemistry characteristics and radiological behaviour of rocks in the Canary Islands has been elaborated, and is detailed in Table 1.

This classification is based on that established by (Arnedo et al., 2017) for the Canary Islands. In their paper, Arnedo et al. study the content of natural radioisotopes (^{226}Ra, ^{232}Th and ^{40}K) and they determine that the intermediate or acidic rocks (Code A in the present classification) are those that present a higher concentration of natural radioisotopes and, in particular, of ^{226}Ra which is the parent nucleus of ^{220}Rn. In contrast, the work of Arnedo et al. (2017) shows that the basic and ultrabasic rocks (Code B in the present classification), which make up most of the territory of the Canary Islands, present reduced concentrations of natural radioisotopes and of ^{226}Ra in particular. The difference between the radiological behaviour of both codes is observed regardless of whether the rocks are plutonic or volcanic and, for this reason, in the simplified classification of this paper, they are treated jointly in each code. The classification is completed with category C dedicated to terrestrial sediments that mainly encompasses the clays and silts and category D assigned to marine sediments, mainly beach sands.

It is important to note that in the case of the Canary Islands, soils are poorly developed and are fundamentally composed of the type of rocks abundant in their local environment. For this reason, when we study an area and classify it as A and B, it means that the dominant rocks and soils derived from their weathering correspond to these geochemical/radiological categories that we have established. Clays will also be different depending on whether they are acidic or basic in composition, but due to their characteristics of accumulating elements of large ionic radius, they always present a higher concentration of natural radioisotopes than the rocks of their surrounding environment from which they come (which can be either acidic or basic) and therefore have a different code.

The lithological map of the Canary Islands is characterised by having great heterogeneity in a relatively small space. In addition, it is sometimes made up of layers of reduced thickness, with underlying layers of different composition. There is an added difficulty in identifying the soil due to the proximity of the edges of different lithologies or the carrying out of excavations that allow reaching a layer with characteristics different from the outer layer. In order to minimise the aforementioned edge effect, soils have been identified by measuring terrestrial gamma radiation (TGR), which allows us to assign the analysed points to a specific code.

This paper studies the radon exposure risk in San Cristóbal de La Laguna and Telde (second municipalities in order of population for the main islands of the archipelago, Tenerife and Gran Canaria respectively). These municipalities were selected because both are classified as zone II for radon risk (Ministerio de Fomento, 2019) and have similar characteristics (Table 2). Furthermore, both municipalities have most of their

Table 1
Simplified classification of the soils of the Canary Islands based on radiological and geochemical criteria (Arnedo et al., 2017).

Code A	*Intermediate and acidic rock* (trachytes, phonolites, rhyolites, trachybasalts, benmoreites, syenites, sieno-diorite, nefelític syenite)
Code B	*Ultrabasic and basic rock* (basanites, basalt olivine, basalt pyroxenic, basalt plagioclasic, nephelinites, hawaitas, tephrites, mugearitas, phonolitic tephrites, tephrites phonolites, peridotite: dunites, werlites, lerzolites, piroxenites, olivine gabbro, feldspar gabbros, noritas, ijolites)
Code C	*Terrestrial detrital sediments* (clay, silt)
Code D	*Marine detrital sediments* (silt, sand and gravel-edge-blocks in submerged areas, beaches and dunes)

Table 2
Geographical characteristics of study areas.

	San Cristóbal de La Laguna	Telde
Location	NE Tenerife	E Gran Canaria
Area (km^2)	102.60	102.43
Elevation (masl)	1020	1546
Population density (people per km^2)	1551.07	1024.52

geology (volcanic rocks) involve by the TAS diagram (Total Alkali Silica) (R. W. Le Maitre, 2002) which classifies volcanic rocks based on the relation between alkaline minerals content (Na_2O and K_2O) and silicates content (SiO2).

It is necessary to highlight the geological singularity of the municipality of San Cristóbal de La Laguna. Much of its geology is composed of basalts, and we can distinguish three different geological areas in the municipality of La Laguna. The old Anaga Massif Structure located in the East – Northeast zone with predominate basaltic flows and basanitic basaltic flows; the South and Northwest zone which is a younger geological zone composed of basaltic flows; and a Central and Western zone with little difference in elevation made up of soils formed by sandy clay deposits and lake soils. The last one has a particular geology due to the existence of a lagoon (already disappeared in the mid-19th century), in addition to being an area that, due to its rainy climatology, has led to the formation of fertile land of considerable size. The urban area is sited on basic flows (Code B) and clayey soils (Code C).

Most of the area of the municipality consists of basic volcanic rocks and soil deposits (detrital sediments) from these rocks in the area with lower elevation. These types of soils can be classified as Code B. However, there are small located areas of intermediate/acid volcanic rocks (Code A), especially on the northern side, but there are no urban areas there.

4. Instrumentation and methods

4.1. Gamma radiation rate measurement equipment

Gamma radiation is considered in the literature as a proxy variable to estimate radon risk areas. The Spanish maps of radon prone areas uses it as a categorical variable to exclude some areas (García-Talavera and López-Acevedo, 2019). In this work, terrestrial gamma radiation is also explored as a determining factor. The instrumentation used for measuring the external gamma dose rate was:

Saphimo MiniTRACE CSDF is a contamination monitor and multifunctional survey meter. It is provided with an energy compensated Geiger-Mueller pancake detector with an active counter area of 15.5 cm^2 and active diameter of 44.5 mm. For the measurement of ambient gamma radiation, the Rate Mean Mode is used, which performs a temporal average of the measurements that allows reducing the error by increasing the counting time. The uncertainty in the measurements was established to be lower than 10%, which required a counting time of around 20 min. The detector provides the result of the energy compensated to the ambient dose equivalent (H *(10)) in microSv/h. The gamma sensitivity is 4.3 cps/μSv/h and the energy range runs from 26 keV to 1.25 MeV. The lower limit of detection is 0.01 μSv/h. A certificate of calibration was provided by the supplier from October 2018 and verified periodically.

Ludlum μR model 12S is a radiometer equipped with 1″ × 1″ sodium iodide (NaI)Tl scintillation detectors. This equipment is designed for environmental low-level gamma surveys and its typical sensitivity is 175 cpm/μR/h. The inherent detector background was determined from the measurements taken within a 15 cm thick iron shield. It was calibrated in June 2020, before the measurement campaign, at the Spanish Center for Energy, Environmental and Technological Research (Ciemat).

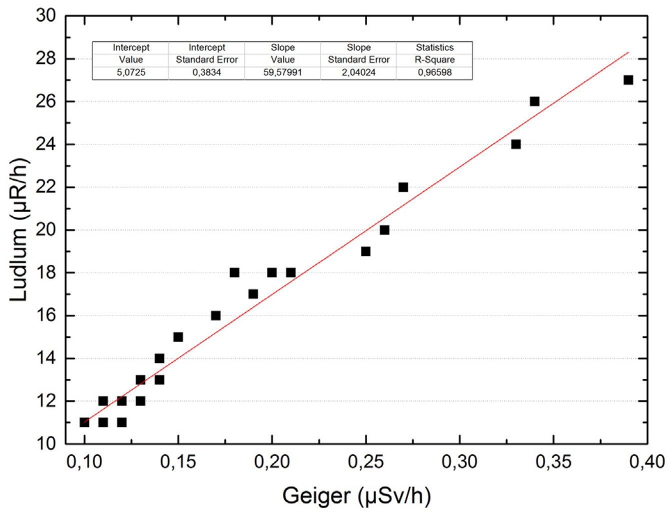


Fig. 1. Intercomparison curve with the distance from a standard point source of both radiometers.

To relate the measurements of both detectors, an intercomparison curve with the distance from a standard point source has been carried out according to the usual process. Using this curve (see Fig. 1), measurements from the Geiger minitrace detector in μSv/h can be directly converted to the readings from the Ludlum 12S radiometer in μR/h. The final results are expressed in nGy/h using a factor 8.76 as equivalence between μR/h and nGy/h.

4.2. IRC measurement equipment

As it is said above ^{222}Rn is a radioisotope in the decay chain of ^{238}U which decays into ^{218}Po by alpha emission of 5.59 MeV. Nuclear tracks detectors are a widely used method to obtain radon activity concentration in air. In this work, Radosys RSKS Type alpha-track detectors have been used to measure IRC. Each dosimeter is made up of a cylindrical plastic diffusion chamber containing a 100 mm^2 CR-39 chip. Typical detector equilibrium time is 3 h. They have a sensitivity of 2.0 tracks·cm^2·kBq^{-1}·h^{-1}·m^3 and a saturation limit greater than 12,000 kBqh/m^3. The typical starting background of the detector is 0.3 tracks·mm^{-2} and its detection limit is 6 Bq/m^3 for 90 days of exposure. Etching is performed using a 25%/6.25 M sodium-hydroxide solution at an Etching temperature of 90C with an Etching time of 4.5 h. Two systems were used for processing the dosimeters: the Radosys NanoReader/NanoBath system located at the Laboratory and Construction Quality Service of the Ministry of Public Works and Transport of the Canary Islands Government on the Island of Tenerife; and the Radosys Radometer 2000/RadoBath system located at the Laboratory of Environmental Radioactivity of the Physics Department of the University of Las Palmas de Gran Canaria on the Island of Gran Canaria. Each batch of detectors used has individualised calibration parameters provided by the supplying company and updated in the control/reading software.

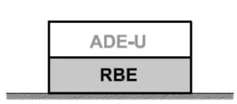
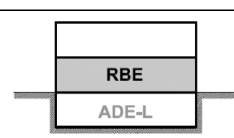
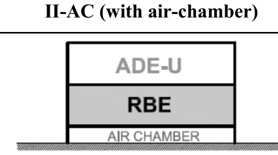
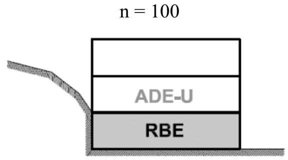
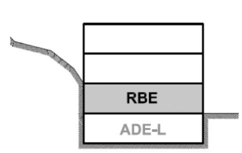
4.3. Classification of building enclosures according to their level of contact with the ground

Given the fact that radon flux penetrates into the building through the envelope from subsoil, it is reasonable to highlight the importance of considering the contact level of the envelope of enclosures as a determinant influence in IRC. For this purpose, a classification of buildings based on the contact level to the ground of the lowest storeys has been designed distinguishing among five simplified types from A to E as shown in Table 3.

Several researchers agree on the decision to take long-term measurements of radon on the ground floor (floor of a building at ground level) or in those lowest floors of the building with a certain level of occupation, but without giving a special relevance to establishing some uniformity in the choice of the floor in which measurements will be realised (Robayna Duque, 2002; Sainz-Fernandez et al., 2014; Collignan et al., 2016). However, the most common tendency in Europe is taking indoor radon measurements on the ground floor (Bossew, 2015; Elío et al., 2017). The Pan-European Map (Elío et al., 2019) proposes an interesting specification to homogenise the way of treating data from all the participating countries in order to elaborate a common indoor radon map. This map covers 50% of Europe with a 10 km × 10 km grid containing the mean of the measurements from each cell from participating countries realised exclusively on the ground floor of the studied buildings, regardless of whether it is in direct contact with the ground or has one or more basement floors on the lower level.

One of the objectives of this work is to try to quantify in some way the significance of the results in the way of choosing the enclosures in each building where such measurements should be taken. This is necessary to propose a clear definition of a representative building enclosure and its behaviour against radon concentration, where measurements have to be taken, that could be extended to the protocol of radon prone areas research in other regions. We define as the **Representative Building Enclosure** (**RBE**), the enclosure located on the lowest floor at ground level which offers sufficient natural lighting and ventilation conditions in order to be inhabited. Complementarily, in order to analyse

Table 3
Distribution of RBE and ADE in the studied buildings and categorisation of RBE.

RBE Category I	RBE Category II II-B (with basement)	RBE Category II II-AC (with air-chamber)
 Type A) Building without basement n = 100	 Type B) Building with basement n = 32	 Type C) Building with air-chamber n = 6
 Type D) Building without basement built on hillside n = 16	 Type E) Building with basement built on hillside n = 0	

the data obtained in this work, we define the Upper and Lower **Adjacent Building Enclosure** (**ADE-U** and **ADE-L** respectively), as the enclosures located immediately above RBE when there is no basement in the building, or the enclosure immediately below RBE in other cases (see Table 3).

Although the criterion followed by Elío et al. (2019) involves an important improvement to achieve a procedural homogenisation, if the objective is characterising radon risk in different territories in a homogeneous way, it is still possible to establish greater control over the variability of the boundary conditions that should be considered. By considering only ground floor results in order to map radon prone areas in Germany, Bossew realised an interesting comparison between results from ground floors of buildings with and without a basement (Bossew, 2015).

In this work, we wanted to take a further step in studying this effect by proposing a more specific categorisation of ground floor enclosures in buildings. For this, two complementary categories of RBE have been considered based on the contact with the ground, so that Category I enclosures have a direct contact, whereas Category II enclosures have an interspace between the measured enclosure and the ground. Within Category II, if this interspace is constituted by a basement, the RBE is classified as Sub-category II-B, while to classify those RBE which have an air-chamber below the ground floor Sub-category II-AC is used (see Table 3.)

4.4. IRC measurements campaign

According to the described methodology, a total of 154 locations (85 from La Laguna and 69 from Telde) have been investigated in this work (see Figs. 2 and 3). These buildings have been randomly selected to perform long-term measurements of IRC using passive detectors. The aim is to achieve a distribution of these points in the territory as uniformly as possible, as well as of a sufficient density to carry out an adequate statistical study. This process provided a list of buildings with residential buildings predominating, most of which are inhabited by a single family, although a certain number of buildings in the service sector are also included.

In order to achieve the purposes of this work, it is necessary to analyse and try to quantify the capacity of radon gas to distribute vertically between adjacent floors. For this reason, alpha track detectors were placed on the two lowest floors of the buildings studied, where it was possible to measure on several floors simultaneously. The protocol for the distribution of detectors in each building is as follows:

- In order to guarantee the quality of the measurement performed, duplicated detectors were placed in RBE.
- Only one detector was placed on the upper (ADE-U) or lower (ADE-L) floor depending on the case.

Next, we kept a check on the placement protocol, choosing the best situation and avoiding spaces with draughts. Also, additional information was collected by using ad hoc sheets (identifiers for dosimeters, type of building, year of construction, location, time of use, periods of occupation and perception of ventilation). After a minimum period of three months, for the removal of detectors, a dosimeter wrapping protocol was carried out to minimise possible contamination of the sample from the point of exposure to the laboratory.

4.5. Radiation gamma measurements

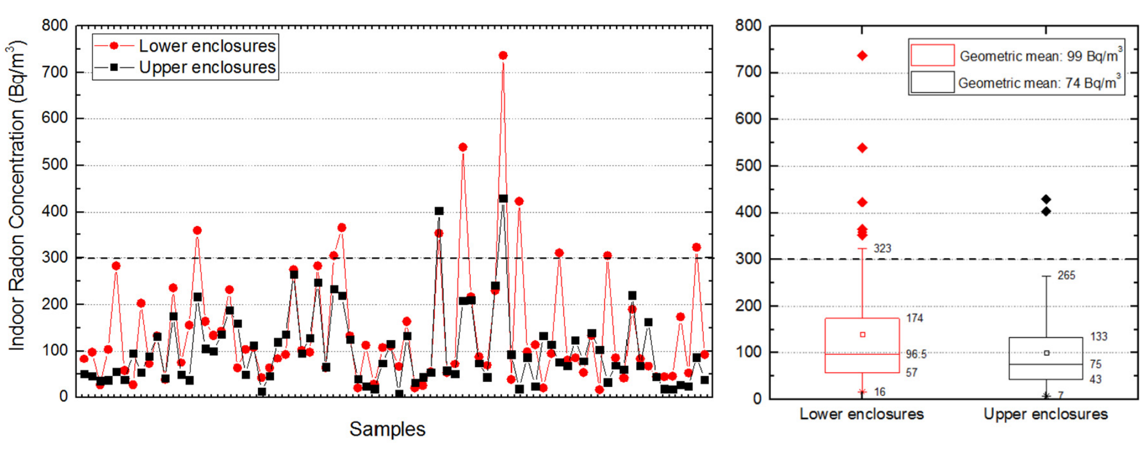
Measurements of the rate of exposure to gamma radiation were taken in areas of unbuilt terrain as close as possible (not upper than 150 m) to the buildings where IRC were obtained in the previous campaign, as reported in Fig. 6a and b. Measurements were made one meter from the ground, with the objective to determine the influence of the different lithologies of the ground in the different study areas. It has been necessary to take various measurements in each location due to the statistical nature of the radiological phenomenon. MARNA (Suárez-Mahou et al., 2000) recommends taking about five measurements at 5-minute intervals, considering the average obtained from them as the definitive measurement. Even more measurements were taken close to the boundaries of lithology change according to the geological map.

For this work, only external sources of gamma radiation were considered which come from the presence of radioisotopes in the ground, building materials, water and air. The cosmic radiation component at the latitude of Canary Islands was estimated using the equation considered in the work of Arnedo et al. (2017).

4.6. Geoprocessing of the results

An important tool to organise the results and additional data is the Geographic Information System (GIS). It provides the possibility to introduce geological and geotechnical maps (lithological areas), and it facilitates the identification of abnormal behaviour of some results. The free and open-source cross-platform of GIS Quantum GIS (QGIS) was used for this goal.

The maps used in this work have been obtained from Cartografía de Canarias S. A. (GRAFCAN) (Canary Islands special data infrastructure of Canary Islands Government): IDECanarias Topographical Map 1:20.000, IDECanarias Geological Map, IDECanarias Geotechnical Map and IDECanarias Integrated Topographical Map 1:5000.



(a) (b)

Fig. 2. IRC results in lower and upper enclosures.

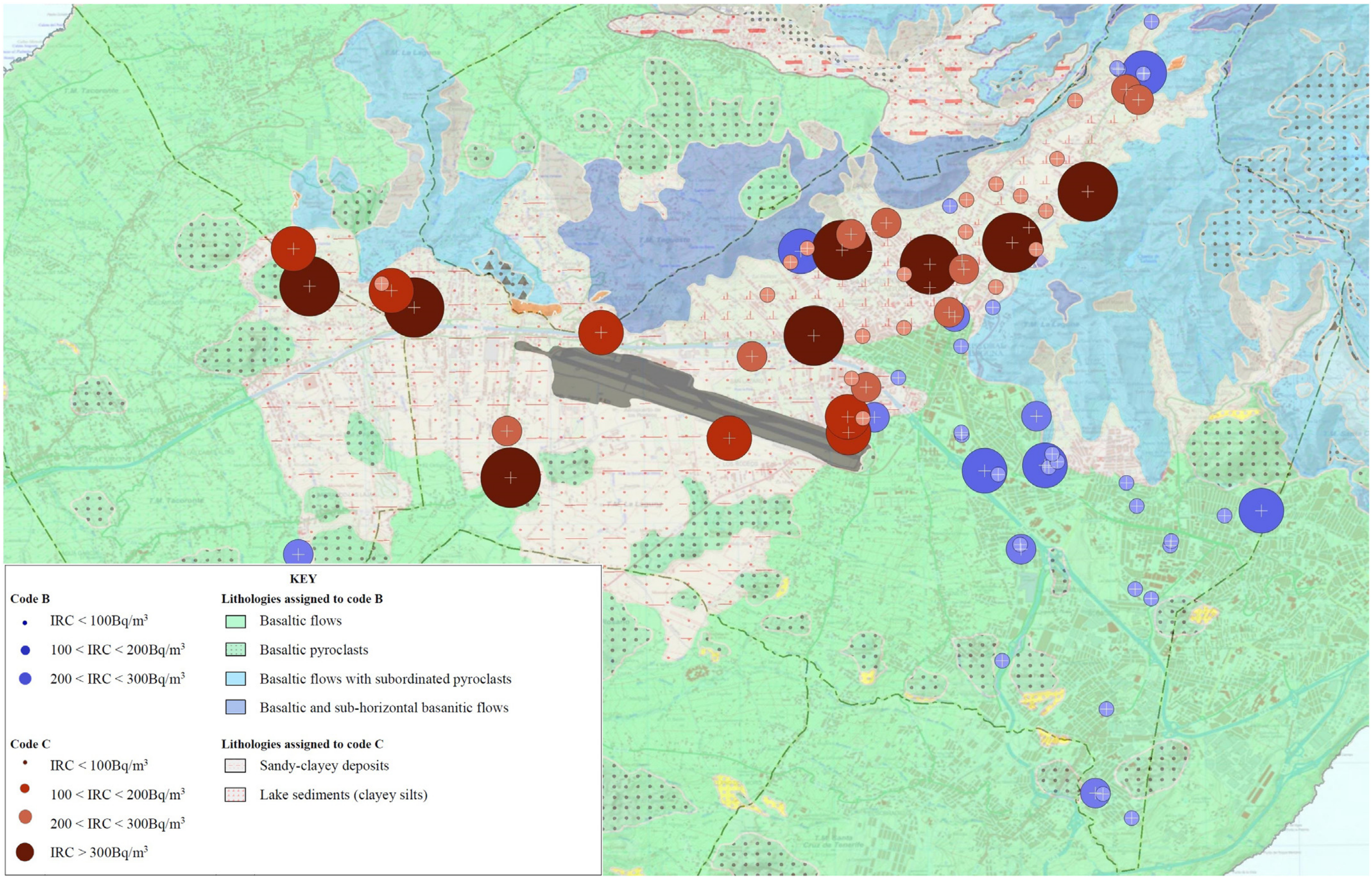


Fig. 3. Map of IRC results in La Laguna coloured by geological code.

4.7. Upper tolerance bounds

The criteria established by García-Talavera and López-Acevedo (2019) for the determination of the different radon-prone areas have been followed, as developed in the work of García-Talavera et al. (2013). One of the most outstanding characteristics of indoor radon gas concentration is its great variability and asymmetry. It is characteristic of a data set affected by random variables that follow a log-normal statistical distribution, where x_g is the geometric mean and s_g is the geometric standard deviation (IGFAE, 2019). Therefore, it can be affirmed that its logarithm ($y_i = ln\,[x_i]$) follows a normal distribution. Accordingly, the unilateral upper bound $\widetilde{Y_p}$ with a confidence level $100(1-\alpha)\%$ that $100p\%$of population where n samples come from at least for a normal distribution is given by Eq. (2) Odeh and Owen (1980):

$$\widetilde{Y_p} = \overline{y} + g'_{(1-\alpha),p,n} \cdot S_y \tag{2}$$

where, $\overline{y}$ is the arithmetic mean, $g_{(1-\alpha),\ p,\ n}{}'$ a factor tabulated by Odeh and Owen (1980) for the calculation of the upper tolerance bound in a normal distribution and S_y is the estimate of the population standard deviation given by Eq. (3):

$$S_y = \sqrt{\frac{n}{n-1}} \cdot s_y, \tag{3}$$

where s_y is the standard deviation of the samples.

Finally, taking into account the equations derived from considering ($y_i = ln\,[x_i]$), $x_g = e^{\overline{y}}$ y $s_g = e^{S_y}$ the unilateral upper tolerance bound $\widetilde{X_p}$ with a confidence level of 100 (1-α)% that 100p% of the population which the n data come from at least, for a log-normal distribution is given by Eq. (4):

$$\widetilde{X_p} = X_g \cdot (S_g)^{g'_{(1-\alpha),p,n}} \tag{4}$$

Therefore, the upper tolerance bound considered in this work (UTB), as indicated in the first paragraph, a value of $\alpha = 0.10$ and a value of $p = 0.90$, are considered, being able to obtain the values of $g_{(1-\alpha),\ p,\ n}{}'$from tables included in Odeh and Owen (1980) and Meeker et al. (1991) for different values of n, (1-α) and p. To handle a type II error, a sample size of $n \geq 27$ must be set (García-Talavera and López-Acevedo, 2019).

It is important to note that this statistical methodology applied to assess the risk of indoor radon in enclosures is sufficiently sensitive for the size of the sample and the deviation of the set. Given the high level of certainty required (90%), it happens that in the cases in which a population group with a small sample size is subjected to analysis, the characterisation value obtained is strongly penalised. In this sense, for example, a sample of 27 values with a geometric mean of 70 Bq/m^3 and a geometric deviation of 2.36 Bq/m^3 corresponds to an estimate of its UTB of 300 Bq/m^3 and keeping the same geometric mean and the same deviation but with only 10 samples the UTB value is 413 Bq/m^3. This means that a smaller sample size reports higher UTB and small increases in its size cause significant reductions in the UTB obtained. Nevertheless, when sample size starts to be enough, UTB trends to stabilise asymptotically. According to this, García-Talavera et al. (2013) established in $n = 27$ a compromise sample size for which the effort spent to increase the sample size is greater than its effect on the accuracy of the results. In those cases analysed in this paper where the sample size is clearly lower than this reference value, as the comparison of UTB among groups may be distorted by this circumstance, it has been preferred to use the geometric mean rather than this parameter in the analysis of the results.

5. Results and discussion

As already indicated above, this work aims to analyse the influence of *intrinsic permanent factors* when radon prone areas are identified. Firstly, the typology of building has been studied, especially the level of contact with the ground of enclosures studied, as a first attempt to highlight the importance of its weight on the behaviour of IRC. Secondly, an analysis of the geology of both municipalities was carried out, using radiometric tools in order to evaluate possible different indoor radon levels based on the composition of soils where buildings sit.

5.1. Lower and upper enclosures in the building

Buildings for residential use, mostly single-family type, and service sector buildings were studied. These buildings have been classified according to their typology following the five general types based on the way that the building makes contact with the ground (See Table 3).

Measurements were recorded in the two lowest adjacent floors of the building if possible, provided that one of them is in contact with the ground, according to the methodology. The lower building enclosure can be located on the basement or ground floor depending on the existence of a floor below ground level. These pairs of measurements per building can allow to distinguish a different indoor radon behaviour between both adjacent enclosures.

Fig. 2 shows results for lower floors and upper floors displayed in pairs in locations where both measurements could be realised (78 buildings represented in abscissas). It enables the comparison of the observed indoor radon results (Bq/m^3).

As can be seen in Fig. 2, lower enclosures have a higher radon indoor level in most cases because of the greater direct contact with the ground, being 64% of all cases. This supposes a significant difference between geometric means being above 33% in the case of upper enclosures compared to lower enclosures with the same geometric standard deviation ($s_g = 2.3$). Considering the upper type of enclosure in order to characterise a building to estimate radon prone areas gave a UTB of 255 Bq/m^3 (45 Bq/m^3 below 300 Bq/m^3) while if lower type results are considered UTB rises to 346 Bq/m^3 (46 Bq/m^3 over reference level 300 Bq/m^3).

Radon measurements for the two types of enclosures can be described by a lognormal distribution (Kolmogorov-Smirnov test with Lilliefors correction, p-value = 0.20 for both lower and upper enclosures) and Levene's test showed heterogeneous variances (p-value = 0.01). Thus, the application of the Mann-Whitney *U* test shows a p-value = 0.05, which is in the limit of rejecting the null hypothesis. Thus, it is not possible to conclude that there is a difference between the radon measurements of both lower and upper enclosures. Therefore, it seems appropriate to bear this circumstance in mind when selecting the enclosures in which the measurement is carried out, considering the difference that is observed between lower and upper enclosures in the building given the influence they may have when estimating the values of UTB and the geometric means of IRC in the characterisation of radon prone areas as it happens in our case.

On the other hand, there are few references to systematic studies that deal with the relationship between radon concentration levels that can be obtained between building enclosures located on adjoining floors of the same building. Only one document (Cinelli et al., 2019) refers to the estimation of ratios between floors without specifying the methodology followed to obtain them. These ratios establish the relationship between the different floors of the building and the ground floor making use of IRC studies developed in some European regions. From one of these studies (Bochicchio et al., 2005), the ratio is calculated using the quotient between the arithmetic mean of the results obtained on each floor of the buildings considered in the field campaign and that of those obtained on the total of the ground floors. The contribution of this work to this line of research is that, having the pair of values obtained in the two lower adjacent floors of each building studied, allows comparing pairs of data obtained under exactly the same boundary conditions or, as we propose in this article, under the same intrinsic and extrinsic factors.

Table 4 shows a comparison between the ratios in different regions, verifying that the results obtained in this work are close to the ranges of values obtained in other regions. In addition, this table includes the basement/ground floor ratio, which has only been compared with the available data from Bochicchio et al. (2005).

Considering this behaviour, this paper has determined an index with the aim to correct the lack of homogeneity due to the fact of choosing building enclosures with different levels of ground contact to characterise a determined area. This index allows to normalise values obtained in building enclosures located in different storeys with respect to the RBE in order to be considered in the process of elaboration of radon prone maps in determined study areas. For this purpose, a **Building Storey Index (I_{BS})** has been established, which is defined according to Eq. (5).

$$I_{BS}(i) = \frac{\langle IRC_{RBE}\rangle_{GM}}{\langle IRC_i\rangle_{GM}} \tag{5}$$

where IRC_{RBE} is the IRC in the representative building enclosure and IRC_i is the IRC in a building enclosure located in a determined storey different to RBE where I_{BS} is to be calculated.

In order to obtain this expression, the distribution of the set of IRC_{RBE} has been studied, which is adapted to a log-normal distribution, considering the normal distribution of its transformed values (natural logarithm of IRC_{RBE}). In the same way, we have taken the values of IRC in upper and lower adjacent building enclosures separately (IRC_{ADE-U} and IRC_{ADE-L}) and it has been proved that its transformed values also conform to a normal distribution. A Shapiro-Wilk test was conducted to the transformed data with a level at decision of 5% of IRC_{ADE-L} ($p = 0.67$), and Kolmogorov-Smirnov test for transformed values of IRC_{ADE-U} ($p = 0.56$) and IRC_{RBE} ($p = 0.93$). Considering the log-normal distribution of the values of IRC, it is inferred that the geometric mean is the statistical mean which adjusts better to this kind of distribution. The I_{BS} in Eq. (5) is obtained from the quotient of geometric means of IRC.

For the calculation of the combined uncertainty of I_{BS} (i) the Eq. (6) is applied according to the procedure suggested by CSN (Romero et al., 2003).

$$u_{I_{BS}(i)} = I_{BS}(i)\sqrt{\frac{u_{RBE}^2}{\langle IRC_{RBE}\rangle_{GM}^2} + \frac{u_i^2}{\langle IRC_i\rangle_{GM}^2}} \tag{6}$$

where u_{RBE} and u_i are the uncertainty of geometric means of IRC_{RBE} and IRC_i respectively.

Table 5 shows the application of I_{BS} to the values obtained in this work in the lower adjacent enclosure ADE-L (basements) and in the upper adjacent enclosure ADE-U (first floors). The I_{BS} index takes values less than unity when IRC corresponds to building enclosures in a storey lower than the RBE storey, while in the case of upper building enclosures the opposite occurs.

Table 4
Comparative among IRC ratios by storey levels from various researches.

Region	Ratio basement/ground floor	Ratio ground floor/first floor
Italy	1.2[a]	1.2
Barcelona (Spain)	–	1.6
Madrid (Spain)	–	1.5
UK	–	1.5
Austria (from 5 provinces)	–	1.5
This work	1.4	1.4

[a] Data obtained from (Bochicchio et al., 2005).

Table 5
I_{BS} of lower adjacent building enclosures (basement) and upper (first floors) in study area.

	ADE-L (basement)	ADE-U (first floor)
Number of samples	17	61
Geometric mean of $IRC_{ADE,}$ (u_{ADE})	121 (2.1) Bq/m^3	74 (2.1) Bq/m^3
Geometric mean of $IRC_{RBE,}$ (u_{RBE})	95 (2.0) Bq/m^3	96 (2.2) Bq/m^3
Building Storey Index, I_{BS} (u_{IBS})	0.79 (0.02)	1.31 (0.05)

Therefore, by applying the I_{BS}, it is proposed to standardise the methodology of data processing for the preparation of maps, being able to compensate, when necessary, for the variability of boundary conditions in which the measurement can be realised when information on the RBE of the building is not available. The I_{BS} index allows to correct the values obtained in adjacent building enclosures to the one in which the proposed pan-European map indicates that measurement should be carried out.

Most of the buildings considered are exclusively single-family homes and those with a basement are generally used as a garage. Buildings with a collective garage below ground floor with a forced ventilation system should not be extrapolated the value $I_{BS\text{-}L}$ without having a specific study which analyses the appropriateness of its application to these cases.

5.2. Ground floor in contact or without direct contact with the ground

To better assess the influence of the building typology, especially the contact between the envelope and the ground, the results of IRC levels obtained for the two categories of RBE defined in Table 3 of this article in both municipalities are analysed separately. In Table 6 it is observed that, although both categories can be considered ground floor according to their definition in the pan-European map (Elío et al., 2019), there are certain discrepancies in the results that can characterise their behaviour. A difference of 14% between the geometric means of both categories I and II can be observed, increasing to 28% when UTB results are considered.

Related to this result, it is important to consider that among the buildings in category II, a Sub-category identified as II-AC in Table 3 is included, consisting in those buildings with an air-chamber system between the ground floor slab and the ground. This kind of building is common in the higher area of La Laguna where single family homes predominate. This air-chamber solution is adopted in buildings trying to avoid damp on the walls as a consequence of a high humidity in the ground due to the rainy climate of this area. It is relevant to note that CTE recommends the application of an air-chamber system as a solution in order to reduce radon levels in new buildings (Ministerio de Fomento, 2019).

It appears logical to study the effects of this kind of system in the statistical analysis which is necessary to determine the risk levels caused by indoor radon gas. In the samples studied, there are 6 cases of building enclosures with air-chamber systems which report a geometric mean of 58 Bq/m^3. This fact supposes that the difference between geometric means of Category I and the set of Sub-Category II-B (Category II discounting results from Sub-category II-AC) is reduced to less than half. It could be concluded a certain coherence between the results of categories I and II, so that it can be concluded that this differentiation is not necessary. Moreover, the important decline of geometric mean of the set Sub-category II-AC confirms the advisability of the remediation prescribed by the CTE. Going further, it could be concluded that measurements realised in air-chambered buildings can be considered anomalous from a statistical point of view when the radon exposure level of a building is being determined given its effectiveness against IRC. Air-chambered enclosures can obtain low results despite being on radon prone ground, and this fact distorts the statistical analysis.

Table 6
Statistical results of categories I and II RBE of buildings in study areas.

	Category I	Category II	Sub-category II-B
Number of samples	110	28	22
Geometric mean (geometric standard deviation)	88 (2.4) Bq/m^3	77 (2.0) Bq/m^3	83 (1.9) Bq/m^3
Upper tolerance bound (UTB)	309 Bq/m^3	241 Bq/m^3	264 Bq/m^3

As has been discussed above, in order to obtain better quality information in the field, data filtering has been carried out. This consists of excluding the cases in which the buildings have air-chambered enclosures from the analysis to avoid the mentioned distortion caused by this architectural solution.

5.3. Analysis of influence in IRC of geological code and TGR

The second variable analysed in this work is the geology of the ground where the buildings are sited related to its radiological and geochemical characteristics. In this work, an identification of different codes of lithologies has been realised with the help of IDECanarias Geological and Geotechnical Maps, as well as the work of Losada et al. (2013) concerning the geochemical characteristics of Canary rocks. In a complementary way, TGR measurements have been realised as an indicator tool of radon gas presence in soils. García-Talavera and López-Acevedo (2019) assert that the correlation between TGR and IRC in dwellings is certainly weak but qualitatively significant. Quindós et al. (2008) concluded that gamma radiation can only be used as a qualitative indicator of high IRC levels rather than a quantitative IRC predictor, and then it can be used to obtain a first approximation for the identification of radon prone areas.

5.4. Geological code

Figs. 2 and 3 show the locations of buildings with the IRC results in RBE obtained chromatically differentiated by codes over a lithological map. The results have been graphically divided into different ranges (below 100 Bq/m^3, between 100 and 200 Bq/m^3, between 200 and 300 Bq/m^3, and over 300 Bq/m^3). There is a total of 79 results in RBE of buildings located in La Laguna discounting those with air-chamber systems, 37 of them located on basic geology (Code B) and the other 42 located on clayey soils (Code C). On the other hand, Telde have 53 results in RBE of buildings sited on basic geology, so that a unique Code B is considered.

Fig. 3 shows that the highest results are predominantly in Code C areas of the municipality of La Laguna. In fact, all results over 300 Bq/m^3 belong to Code C. In the case of the municipality of Telde (Fig. 4) it is not distinguished as an area where the highest results are predominant, but these points are distributed along the map without a specific area with higher density. However, it can be observed that lower ranges of IRC are predominant in Code B areas from both municipalities.

Fig. 5a and b show IRC results from Telde and La Laguna respectively, identifying geological codes B and C. It can be confirmed that geological codes B (basic rocks) from both municipalities have a very similar behaviour with low values of IRC, observing only four results slightly above 300 Bq/m^3 in the case of Telde, while higher results are found in geological code C from La Laguna, where clayey soils are extended.

The application of Mann-Whitney *U* Test reveals that there are differences between IRC measured in Code B and Code C (p-value $= 0.00$). Values of IRC obtained in each code B and C describe a log-normal distribution (Kolmogorov-Smirnov Test with Lilliefors correction, p-value $=$ 0.20 for Code B and Shapiro-Wilks test, p-value $= 0.65$ for Code C) with heterogeneous variances (Levene's test, p-value $= 0.00$).

5.5. Terrestrial gamma radiation

As has been indicated above, in this work a methodology has been established consisting of designing a gamma radiation measurements campaign carried out in the proximity of the same locations where

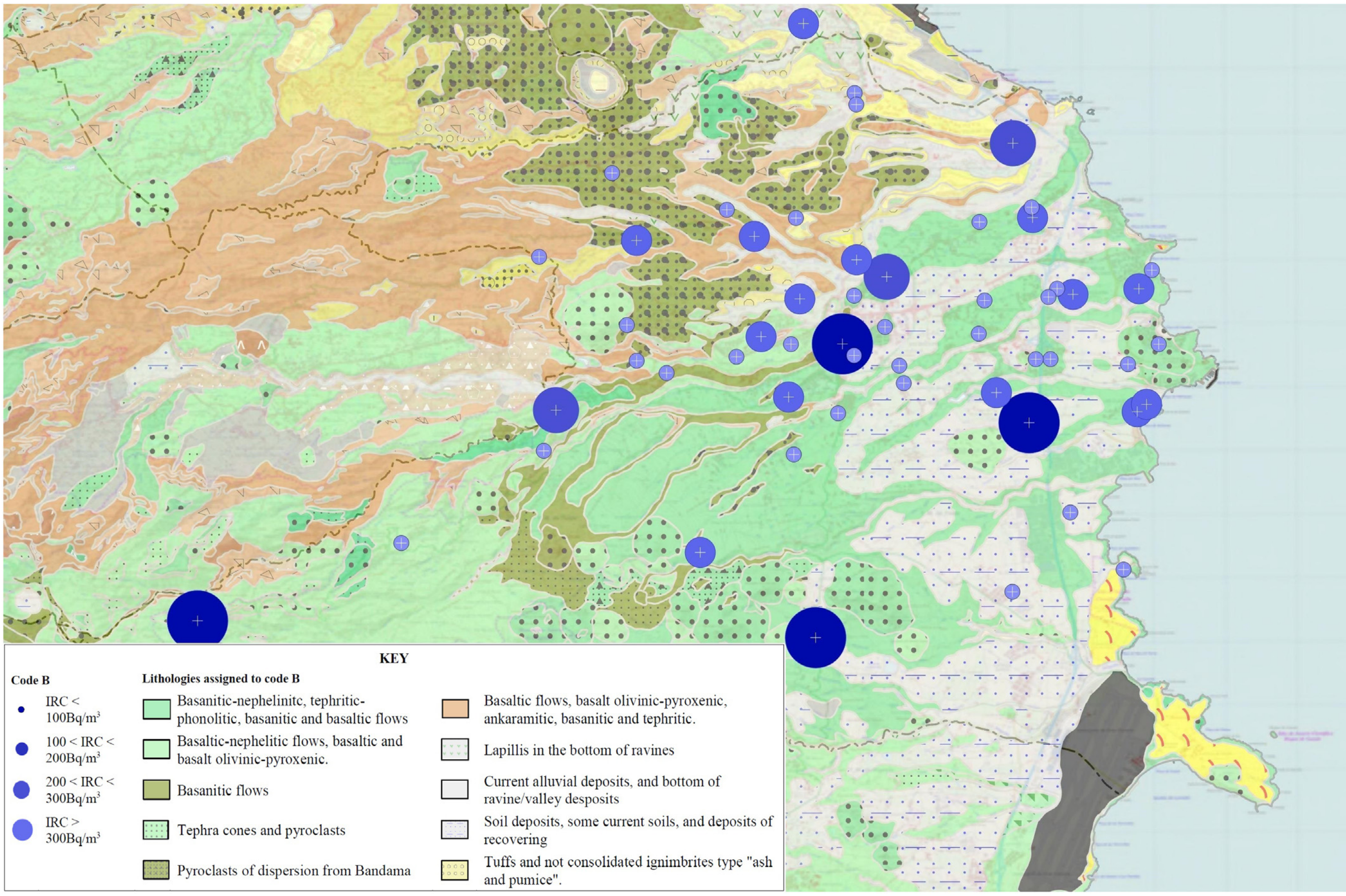
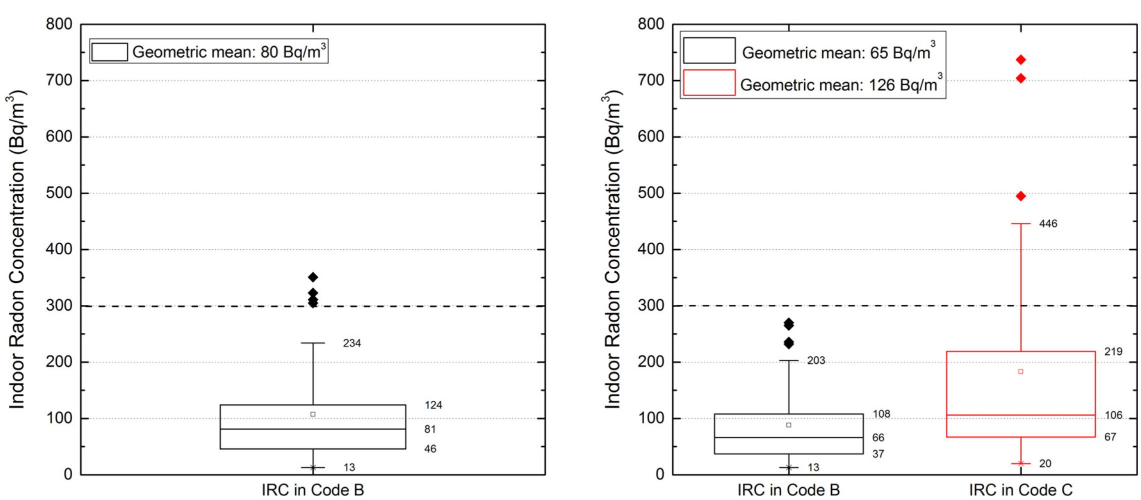


Fig. 4. Map of IRC results in Telde coloured by geological code.

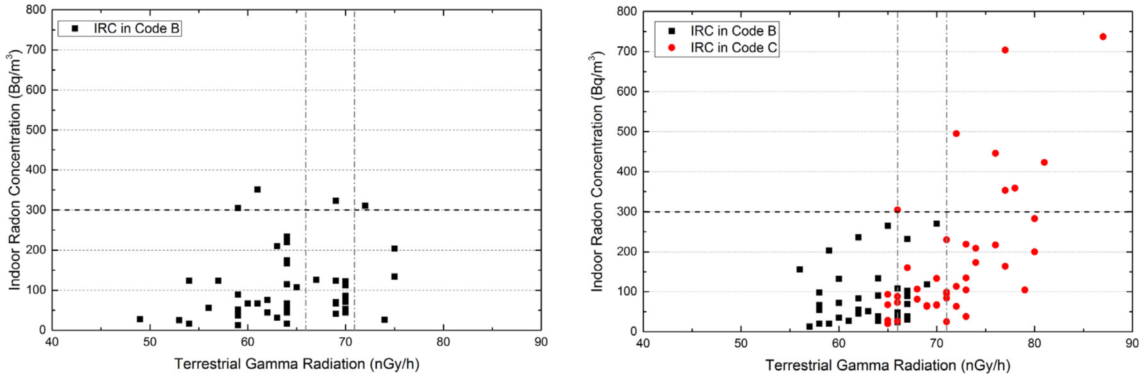
IRC measurements were taken in both municipalities. Applying this methodology, the aim was to obtain a quantifiable information about the influence of geological characteristics of the underlying ground on IRC.

The CSN in its Technical Report Cartography of radon potential in Spain (García-Talavera and López-Acevedo, 2019) proposes using the level of gamma radiation as a variable that allows a first approximation to be made to identify the geographical areas most exposed to radon gas. In this way, the territory is divided into three categories depending on whether their radiation level is below 66 nGy/h (~7.5 μR/h) for the first category, between that value and 123 nGy/h (~14.0 μR/h) for the second and higher values for the third. García-Talavera and López-Acevedo (2019) concludes that the level of 66 nGy/h is a very reliable



a) Telde b) La Laguna

Fig. 5. Box and whiskers graphs of IRC results in study areas differenced by codes.



a) Telde b) La Laguna

Fig. 6. Results of IRC in RBE of buildings from study areas according to TGR. Black squares marks indicate Code B and red circles marks denote Code C. Vertical lines correspond to the cut-off proposed by García-Talavera et al. (2013) (66 nGy/h) and the cut-off proposed in this work (71 nGy/h).

limit to determine that the regions with lower rates do not constitute priority action areas for radon exposure.

Fig. 6 shows TGR ambient dose results regarding IRC in La Laguna and Telde respectively, identifying geologies chromatically.

Considering these ranges, it can be observed in Fig. 5a and b that both in Telde and in La Laguna those points that show TGR lower than 66 nGy/h have an IRC lower than 300 Bq/m^3, except only two cases in Telde which slightly exceed this limit (305 Bq/m^3 and 351 Bq/m^3). In line with this, in Table 7 it can be observed that the geometric mean of IRC in this range gives a result of 65 Bq/m^3. This geometric mean is lower than that obtained by García-Talavera in 181 municipalities from mainland Spain with TGR lower than 66 nGy/h whose value is 70 Bq/m^3. On the other hand, no point from Telde or La Laguna exceeds the cut-off of 123 nGy/h, so that the whole set of samples evaluated would belong to the two first ranges, obtaining a UTB of 237 Bq/m^3 for those points with TGR lower than 66 nGy/h and 360 Bq/m^3 for the rest. The application of a Mann-Whitney *U* Test shows that there are differences between measurements of IRC belonging to the two ranges considered (p-value = 0.00).

In the paper of Talavera et al. (2013) the TGR cut-off 66 nGy/h is linked to a geometric mean of IRC 70 Bq/m3 in the study area. However, these limits proposed by the CSN respond to a geological reality typical of continental Spain (mainland Spain), which is very different from the Canarian geology markedly characterised by its intraplate volcanic origin (Carracedo et al., 2002). In this work, the value of TGR which gives exactly this geometric mean is 70.5 nGy/h. This leads us to propose a limit of 71 nGy/h for our study area. The observed behaviour in the graphs of Fig. 5a and b around the value 71 nGy/h, makes to infer that using this cut-off allows for establishing ranges of TGR which adjust better to the results obtained in the current study. As can be observed, only 4 of the 97 points in which a TGR lower than the analysed limit is observed slightly exceed 300 Bq/m^3, offering a geometric mean of 71 Bq/m^3, while those points which exceed 7 nGy/h show a more disparate behaviour obtaining a geometric mean of 169 Bq/m^3. As can be seen in Table 8, this fact translates into obtaining an UTB of 212 Bq/m^3 in values from the first range, and UTB of 712 Bq/m^3 in values from the second. For these new ranges, the existence of different ranges of IRC measurements was also verified (Mann-Whitney *U* test, p-value = 0.00).

As explained in Section 4.7 of this paper, the shortage of cases exceeding 71 nGy/h in both analysed municipalities makes it not statistically advisable to establish several different ranges for this population, since the consequent loss of samples in each range would distort the results.

6. Conclusions

Two indoor radon and terrestrial gamma radiation measurement campaigns have been carried out in the study areas considered (municipalities of the Canary Islands: La Laguna and Telde), matching the physical location of the measurement points in both campaigns.

A definition of the RBE has been proposed with the intention of achieving greater precision when selecting the enclosures in which the measurements are carried out in order to provide greater uniformity of criteria in the elaboration of maps.

The IRC field campaign has been designed in such a way that whenever possible two passive detectors have been placed in the RBE and another one in the vertically contiguous enclosure, the upper one when the RBE was in contact with the ground and the lower one when it was not.

To study the influence of the type of soil in which a building is located on the IRC, a simplified coding has been defined based on the geochemical and radiometric characteristics of the soils in the study areas that classify them into acidic rocks (Code A), basic (Code B), clayey (Code C) and marine detrital sediments (Code D).

The main results obtained are the following:

1) It has been observed that the UTB calculated for a study area can give disparate values depending on whether rooms located on the upper or lower storey are predominantly chosen. As a result of the implementation of this methodology, a I_{BS} was obtained that would allow normalizing measurements taken in rooms from storeys which are not ground floor, when this information is available in the initiatives carried out to define radon prone areas.

Table 7
Statistical results of IRC measured in RBE by ranges of TGR values according to CSN.

	Low TGR (<66 nGy/h)	Intermediate TGR (66–123 nGy/h)	High TGR (>123 nGy/h)
Samples	57	73	0
Geometric mean (geometric standard deviation)	65 (2.3) Bq/m^3	108 (2.2) Bq/m^3	–
Upper tolerance bound (UTB)	237	360	–

Table 8
Statistical results of IRC measured in RBE by ranges of TGR values.

	Low TGR (<71 nGy/h)	High TGR (≥71 nGy/h)
Samples	101	29
Geometric mean	71	169
Geometric Standard deviation	2.1	2.4
Upper tolerance bound (UTB)	212	712

2) On the other hand, it has been found that there are constructive solutions that drastically reduce radon gas access to the building, even though it sits on land with a high concentration of radon, such as an air-chamber between the floor and the ground. The results obtained in the air-chambered enclosures can be considered statistically anomalous, and their use in calculating the UTB of the geographical area under study is not recommended.
3) La Laguna is fundamentally divided into two codes (Code B and Code C), while Telde in its entirety is in Code B. In this study, it is shown that buildings that sit on basic rocks or soils derived directly from these rocks present significantly low IRC results. In clayey soils of La Laguna (Code C), due to their formation process in which a higher concentration of natural radioisotopes accumulates than in the rocks from which they come, buildings withstand significantly higher IRC levels.
4) After analysing the TGR campaign carried out in the vicinity of the points where the IRC has been measured, it is observed that in these municipalities the TGR can be used as a categorical variable to define radon risk areas. Considering the limits of TGR established by CSN to delimit radon prone areas it can be observed that values below 66 nGy/h have a UTB of 237 Bq/m^3, so these would not be priority action areas (<300 Bq/m^3). However, CSN links this limit to a geometric mean of IRC of 70 Bq/m^3, so it is also possible to define a TGR cut-off of 71 nGy/h around which, in addition, a certain ordering of radon levels is distinguished. Below this cut-off, the IRC results are concentrated in geological code B (Telde and part of La Laguna), obtaining for this population a UTB of 212 Bq/m^3, and therefore a low risk level for radon gas exposure. Buildings located in code C in La Laguna whose UTB exceeds 700 Bq/m^3 exhibit TGR values above 71 nGy/h.
5) The proposed methodology can be applied in other Canary geographic areas. Increasing the density of samples, the results here presented can be generalized to the entire archipelago and help to define the radon prone areas with more precision.

CRediT authorship contribution statement

C. Briones: Writing – original draft, Investigation, Methodology, Visualization, Validation, Data curation, Writing – review & editing. **J. Jubera:** Investigation, Writing – review & editing, Supervision, Project administration, Methodology, Data curation. **H. Alonso:** Investigation, Resources, Project administration. **J. Olaiz:** Investigation, Resources, Project administration. **J.T. Santana:** Investigation, Resources. **N. Rodríguez-Brito:** Investigation, Resources. **A. Tejera:** Investigation, Resources. **P. Martel:** Investigation. **E. González-Díaz:** Investigation, Formal analysis, Writing – review & editing, Data curation, Supervision. **J.G. Rubiano:** Investigation, Writing – review & editing, Supervision, Project administration, Methodology.

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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