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Assessment of radon risk areas in the Eastern Canary Islands using soil radon gas concentration and gas permeability of soils

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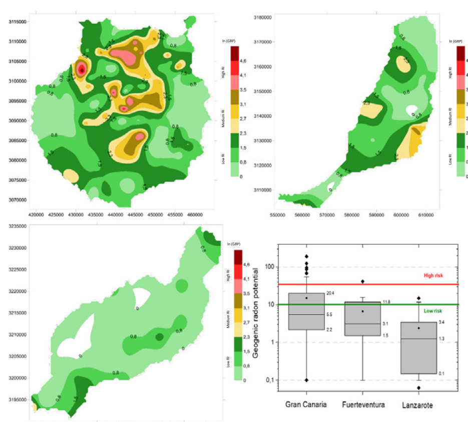
HIGHLIGHTS

- EU Member states have to establish action plans to mitigate radon exposure risks.
- A study of Soil radon gas concentration and gas permeability of soils were performed.
- Geogenic Radon Potential (GRP) distribution in the territory have been assessed.
- Maps of radon prone areas in the Eastern Canary Islands are displayed based on GRP.
- The subsequent zonal classification agrees with those provided by the authorities.

GRAPHICAL ABSTRACT



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ABSTRACT

The Basic Safety Standard (BSS) Directive 2013/59/EURATOM of the European Union (EU) has stated the need for member states to establish national action plans to mitigate their general population's long-term risks of exposure to radon gas. Maps of radon-prone areas provide a useful tool for the development of such plans. This paper presents the maps of radon-prone areas in the Eastern Canary Islands (Gran Canaria, Fuerteventura and Lanzarote) obtained from assessment of Geogenic Radon Potential (GRP) distribution in the territory. GRP constitutes a magnitude that is contingent on both radon activity concentration and gas permeability of soils. An extensive campaign covering all geological formations of the Eastern Canary Islands was undertaken to locally sample these parameters. Geostatistical analysis of the spatial distribution of radon concentration in soils, permeability and GRP was performed on each of the islands, and the relationship between these magnitudes and the characteristic geological formations of the volcanic islands was investigated. Areas dominated by basic volcanic and plutonic rocks (originated by both recent and ancient volcanism) exhibit relatively low levels of radon in soils, and with the exception of specific cases of very high permeability, these areas are not classified as prone to radon risk according to international criteria. Areas in which intermediate or acidic volcanic and plutonic rocks predominate are characterised by greater radon activity concentration in soils, rendering them radon-prone. Given these results, Lanzarote is classified as an island with low radon risk all over its surface; Fuerteventura presents low-medium risk; and Gran Canaria contains extensive areas in the centre and north where the risk is medium or high. This classification is consistent with the risk maps obtained by National and European agencies from indoor radon measurements conducted on these islands.

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1. Introduction

The environmental incidence of radon gas has been matter of study over the last decades due to its adverse effects on human health. Elevated concentrations of indoor radon in homes and workplaces represent a public health concern, as this gas is responsible for approximately 50% of natural radiation dose due to ionising radiations. The International Agency for Research on Cancer (IARC) deems radon a Class 1 Human carcinogen due it being the second most significant cause of lung cancer after tobacco (Elío et al., 2018). In order to mitigate the negative impacts of the population's exposure to high levels of radon concentration, the Basic Safety Standard (BSS) Directive 2013/59/EURATOM of the European Union (EU) stated that “Member States shall establish a national action plan addressing long-term risks from radon exposures in dwellings, buildings with public access and workplaces for any source of radon ingress, whether from soil, building materials or water”. Such national action plans are supposed to include identification of radon-prone areas across the entire territory.

Indoor radon levels represent a complicated function of several factors, such as the geology of the area, building structure (for instance, building materials, presence and type of basement or cellar below the house) and domestic lifestyles. Amongst these factors, the radon concentration in the subsurface in the vicinity of the building, which is contingent on the composition and types of rocks and soils, constitutes the main source of indoor radon (Nero, 1989), while building material is considered a secondary source (Cosma et al., 2013; Nazaroff and Nero, 1988; Righi and Bruzzi, 2006; Szabó et al., 2013). Thus, in order to prevent the radon hazard in a particular region, identification of those areas that (given the local characteristics of the soils) may have the potential to generate high indoor radon levels is essential. In general, radon-prone areas can be identified via two strategies: (a) direct measurements of indoor radon concentration, or (b) in the absence of an adequate number of high-quality indoor radon measurements, assessment of proxy indicators such as radon soil gas concentration, terrestrial gamma dose rate or uranium/radium content in the soil and rocks (Dubois and Bossew, 2006; García-Talavera et al., 2013). When designing a procedure to determine the radon-prone areas, hybrid approaches tend to be developed, i.e., methodologies that integrate all available information for several variables to create optimal tools that facilitate the classification of the territory under study. For instance, in UK (Miles and Appleton, 2005) and Belgium (Cinelli et al., 2011), radon risk maps have been obtained that combine indoor radon measurements with geologic information, whereas in Germany (Kemski et al., 2009) and the Czech Republic (Barnet et al., 2008) radon risk maps have been obtained based on measurements of radon activity concentration and the gas permeability of soils.

The main parameters that affect radon concentration in the soil and its transport towards the external air comprise the concentration of radon precursors (which are dependent on the nature of the geological substrate), the emanation coefficient (such as particle size, porosity, water content and temperature), the transport process (advection and diffusion) and exhalation. In practice, the direct measurement of radon exhalation towards indoor air appears to represent one of the best indicators of the radon potential of the soil. However, direct measurement of this parameter requires long time intervals, stimulating us to instead use the activity concentration of radon in soil for this study.

In this context, soil gas permeability is a fundamental parameter for determination of radon gas mobility (Neznal and Neznal, 2005). This parameter is contingent on porosity and terrain grain size, and can increase with the presence of fractures, structural discontinuities or karst phenomena. For equal quantities of emanated radon (radon moved from within the grain towards the intergranular space or pores) in a terrain, radon flux can vary considerably with permeability, and hence the radon concentration can vary as well. Permeability is significantly conditioned by soil moisture (infiltrated in the pores), which at the same time depends on different factors such as pluviometry and phreatic level variations. A rise in water content in the soil generally stimulates an increase in the radon concentration, but as it approaches saturation level, a sharp drop in measured levels of radon can be identified (Menetrez et al., 1996) due to the abrupt reduction in soil gas permeability. The presence of a superficial soil layer with low permeability can imply an increase in the accumulation of radon below (Johner and Surbeck, 2001), and may also lead to a considerable decrease in the radon's soil-atmosphere flux. This can occur owing to different causes, including the presence of asphalt, water content close to the saturation level, or intrinsic causes of the material such as when the soil contains clay (Wiegand, 2001). Other environmental factors such as barometric pressure or humidity also exist, but due to their variability they cannot be included as indicators for the determination of a terrain's radon potential.

The relationship between radon concentration in soil and indoors has been widely studied (Appleton and Miles, 2010; Barnet and Pacherová, 2011; Barnet et al., 2010; Chen et al., 2009; Kemski et al., 2005). The Geogenic Radon Potential (GRP) is a magnitude that is dependent on both the radon activity concentration at a definite depth in the soil and the soil gas permeability, and is defined as an indicator of the potential of the soil to constitute a source of indoor radon (Neznal et al., 2004). This magnitude operates independently of the influence of any factors related to buildings or living habits. In this paper we present the results of several measurement campaigns of radon in the soil and its permeability, conducted in the Eastern Canary Islands with the aim of obtaining the Geogenic Radon Potential map of this Spanish province, this being a useful tool for the estimation of radon risk across the three islands. This investigation's methodology could also be applied to territories with similar geological characteristics, such as the Western Canary Islands, Islas de Cabo Verde or the Hawaiian archipelago.

2. Study area: Eastern Canary Islands

The Canary Islands are a 500-km-long linear chain consisting of seven volcanic islands and several minor islets (Fig. 1A). The archipelago has developed in a geodynamic setting characterised by a Jurassic (circa 160–180 Ma) oceanic lithosphere at the African passive continental margin. The archipelago developed over the past 30 million years as a result of the west-to-east movement of the African plate over a mantle hotspot presently beneath the western end of the archipelago (Carracedo et al., 2002). Three types of geological units can be found across the islands (Carracedo et al., 2002): 1) basal complexes (or the pre-shield stage), including turbiditic sediments belonging to oceanic crust intruded by sheeted dike swarms and plutonic rocks of a broad compositional spectrum; 2) shield or juvenile volcanism; and 3) post-shield or rejuvenated volcanism. From a geochemical perspective, the volcanic rocks of the Canary Islands belong to the alkaline igneous series, typical of intraplate volcanism. This igneous series is formed by a sequence of rocks whose composition evolves from ultrabasic and basic terms (including volcanic rocks as foidites, basalts, basanites, trachybasalts, tephrites, phono-tephrites and its plutonic equivalents) to intermediate and acid terms (represented by tephra-phonolites, trachy-andesites, trachytes, phonolites, rhyolites and its plutonic equivalents) The area of study is restricted to the eastern and older Canary Islands, i.e. the islands of Gran Canaria, Fuerteventura and Lanzarote, the three islands are at present day in their volcanic rejuvenated stage.

Gran Canaria (Fig. 1B) is a quasi-circular island with a diameter of about 45 km and an area of 1532 km^2. The highest point of the island is located in the centre at an altitude of 1949 m. From a geological point of view the island can be divided into two differentiated areas: the southwestern or Paleocanaria and the northeastern or Neocanaria halves (e.g., Carracedo et al., 2002; Guillou et al., 2004). The southwestern half (Paleocanaria) corresponds to the island's oldest volcanism (ca. 14.5–8 Ma) belonging to its juvenile stage. This area includes a

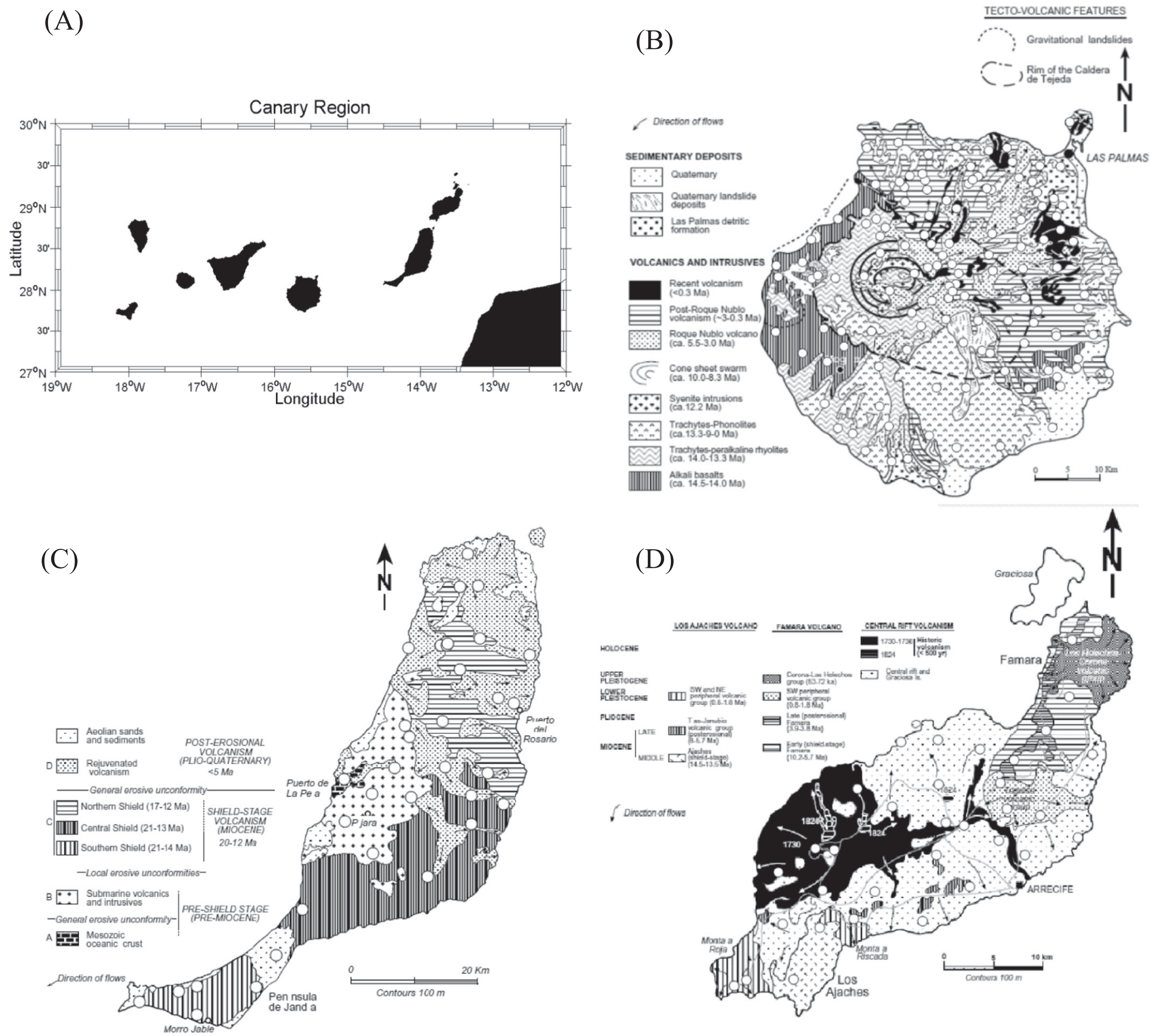


Fig. 1. Geographic localisation of the Canary Islands (A). Simplified geological maps and sampling point distribution for Gran Canaria. (B), Fuerteventura (C) and Lanzarote (D) (Extracted from Carracedo et al., 2002). The white circles correspond to the sampling points. All geological ages provided in Mega-annum (Ma).

broad compositional spectrum of both volcanic and plutonic rocks, ranging from ultrabasic to intermediate and acid terms. In contrast, the northeastern half (Neocanaria) is mainly covered by volcanic rocks belonging to the rejuvenated stage (ca. 5.3 Ma to actual) with a more limited compositional spectrum, mainly dominated by basic volcanic terms (e.g., Guillou et al., 2004; Rodriguez-Gonzalez et al., 2009).

Fuerteventura (Fig. 1C) with an area of 1660 km^2 is the second largest island of the archipelago and the oldest one. The topography of Fuerteventura is characteristic of a post-erosional island with Quaternary sedimentary deposits extensively covering the basal complex (pre-Miocene) and the juvenile volcanism (ca. 21–12 Ma), with minor rejuvenated volcanism (<5 Ma) outcropping in the northern areas (e.g., Coello et al., 1992; Ancochea et al., 1996; Carracedo et al., 2002). Most of the volcanic rocks, both juvenile and rejuvenated stages, are basic in composition. In contrast, plutonic rocks belonging to the basal complex exhibit a broad compositional spectrum, from ultrabasic and basic terms (different kinds of peridotites and gabbros) to intermediate terms (syenites, foid syenites) and carbonatite dykes enriched in REE elements (Mangas et al., 1997).

Lanzarote is the northernmost and easternmost island of the Canary archipelago, with a surface of 846 km^2. Lanzarote, as Fuerteventura, present a flat morphology (maximum elevation of 670 m asl) typical of a post-erosive island. After Fuerteventura is the second oldest island, with juvenile stage ranging from 14.5 to 5.7 Ma. But, opposite to Fuerteventura, rejuvenated volcanism (including historical eruptions) covers large areas of the island. Both, juvenile and rejuvenated volcanism, are dominated by basic terms, with minor intermediate volcanic rocks. Quaternary terrestrial and coastal sedimentary deposits are also present in large areas (e.g., Coello et al., 1992; Carracedo and Rodríguez Badiola, 1993; Carracedo et al., 2002).

In order to analyse the relationship between the geology and concentration of radon activity in the soils, a simplified classification of the geological rocks outcropping in the Eastern Canary Islands has been elaborated (Table 1).

3. Material and methods

3.1. Sampling points

Several campaigns were conducted between 2012 and 2018 to perform a screening of both the radon concentration in the soil and the soil gas permeability across the region. The selection of the sampling points was conceived in order to cover all of the geological units of the islands, as well as areas with differentiated behaviour of several radiological magnitudes such as terrestrial gamma exposure rate or activity concentration of ^{226}Ra, as determined in a previous work (Arnedo et al., 2017). Thus, 131 locations in Gran Canaria, 36 in Fuerteventura and 35 in Lanzarote were sampled to obtain a statistically consistent distribution of Geogenic Radon Potential representative of the islands. In Fig. 1B, C and D, the spatial distributions of sampling points are shown. Thus, the island with the lowest density of measured points is Fuerteventura, which together with Lanzarote presents a lower variability of radiometric data than observed in Gran Canaria (Fig. 1B); moreover, due to their smooth orography, these islands are much more easily characterised. In contrast, a higher number of points were measured in Gran Canaria because it presents a very abrupt orography and far higher variations in radon levels were found. Another factor that was considered when establishing the number of sampling points was that Gran Canaria represents the most populous island, containing approximately 77.3% of the population of the province in comparison with the 9.9% who live on Fuerteventura and 12.8% on Lanzarote, justifying its deeper study.

The climatic variability of the area of study was also taken into account. The Canary Islands are located in the transition zone between areas of temperate and tropical climate, characterised by low and erratic rainfall (300 mm), mostly in low-lying areas due to the presence of the Azores anticyclone for most of the year. The northernmost parts of the islands are exposed to constant humid trade winds, and rainfall here can reach 1000 mm. In the south, rainfall is considerably lower, sunny days are numerous, and periods of rain mainly occur in the late-autumn and winter. Measurements of radon activity concentration and permeability were primarily undertaken in periods corresponding with low or no rainfall.

3.2. Sampling and measuring procedure of radon gas in the soil

In order to measure the radon gas concentration in the soil, a probe designed by Radon v.o.s. was used. This probe represents a cylindrical, hollow probe, measuring 12 mm and 8 mm in outer and inner diameter respectively and 1 m in length, and equipped with a free, sharpened lower end (a lost tip) of 12 mm diameter. Following the procedure, the sharp tip is inserted into the bottom of the probe, which is in turn nailed to the selected depth. Finally, the tip is pushed down to create a hollow in the lower end of the probe, permitting the collection of soil-gas samples, which are drawn up the tube towards the radon measurement equipment. Given that the Canary Islands have poorly developed soils, a sampling depth of 50 cm was chosen. The soil around the probe was tamped to prevent the entry of fresh air when acquiring samples.

Table 1
Simplified classification of the geological materials of the Eastern Canary Islands.

Code 1	Ultrabasic to basic volcanic rocks (foidites, basanites, basalts, tephrites, trachybasalts, phono-tephrites)
Code 2	Intermediate to acid volcanic rocks (tephra-phonolites, trachy-andesites, trachytes, phonolites, rhyolites)
Code 3	Ultrabasic to basic plutonic and subvolcanic rocks (peridotites, gabbros)
Code 4	Intermediate to acid plutonic and subvolcanic rocks (syenites, foid syenites) and carbonatite subvolcanic rocks
Code 5	Sedimentary littoral deposits (sand and gravel beaches and sand dunes)
Code 6	Sedimentary terrestrial deposits (clay, silt, sand and gravel alluvial and colluvial deposits, and soils)

The radon monitor used was the Durridge RAD7, a silicon ion-implanted solid-state detector that measures the concentration of ^{222}Rn by alpha spectrometry of the radon daughter ^{218}Po, which reaches equilibrium with its parent in about 15 min. It has a very low intrinsic background (0.2 Bq/m^3) and an extremely low detection threshold, below 4 Bq/m^3. In order to measure the radon gas concentration in soils, RAD7 includes an automatic GRAB-sampling protocol (Durridge Radon Instrumentation, 2015). Soil gas is pumped for 5 min to the internal chamber until the radon is uniformly mixed with air, and the sample is analysed thereafter to find the radon concentration. While pumping, the air flow rate is about 0.7 L/min and therefore 3.5 L of soil gas are extracted from the soil. If a soil porosity of 0.5 is assumed, the sample will be drawn from a sphere of approximately 12 cm of radius around the sampling point. Thus, the selected depth of 50 cm guarantees that there is no dilution with the air from the surface. The counting time in the grab-sampling protocol is 30 min and the RAD7 has to be purged for about half an hour previously. Therefore, each measurement takes around an hour.

The methodology for the determination of activity concentration of radon gas in soil was successfully validated by participating in three different international comparison measurement exercises of radon in soil gas. The first one took place during the International Intercomparison Exercise on Natural Radiation Measurements under Field Conditions held at the ENUSA facilities in Saelices el Chico (Spain) (Gutierrez-Villanueva et al., 2012) organized by the Environmental Radioactivity Laboratory of the University of Cantabria, (LaRUC), and the state company ENUSA Industrias Avanzadas and financed by the Nuclear Safety Council of Spain (CSN). The next two exercises took place at reference sites of Cetyne and Buk in the Czech Republic in the framework of the Radon Intercomparison Measurements at Radon Reference Sites (RIM) (Matolin, 2017) held on September 2014 and September 2018 organized by radon v.o.s and the Charles University in Prague.

3.3. Measurement procedure of soil gas permeability

Parallel with the radon measurement campaign, we obtained the soil gas permeability at each point using a permeameter RADON-JOK. The permeability measurements were carried out almost simultaneously with the measurements of radon in the soil using the same hollow rod hammered into the ground to a measured depth. The principle of the RADON-JOK equipment consists of air withdrawal by means of negative pressure. The air is pumped from the soil under constant pressure through a specially designed probe with a constant contact area between the probe head and the soil. The constant active area is created in the head of the probe by pushing the tip by means of the punch wire inside the probe a small distance. The special rubber sack, via gravity and the help of one or two weights, pumps air from the soil and facilitates measurements at very low pressures. The soil gas permeability is calculated using the time spent by the rubber sack to be completely filled of air and employing a nomograph provided by the company RADON v.o.s with the relationship between such time and permeability. The RADON-JOK permeameter has a detection limit of 6 s that corresponds to a permeability of 1.8×10^{-11} m^2. The manufacturer's specifications indicate that for very impermeable floors, with a time value above 1200 s, a permeability of 5×10^{-14} m^2 must be assigned.

4. Results and discussion

4.1. Radon gas activity concentration in soils

4.1.1. Gran Canaria

Fig. 2 shows the histogram of the data distribution of radon activity concentration in the soil measured in Gran Canaria, the corresponding statistical indicators and box plot.

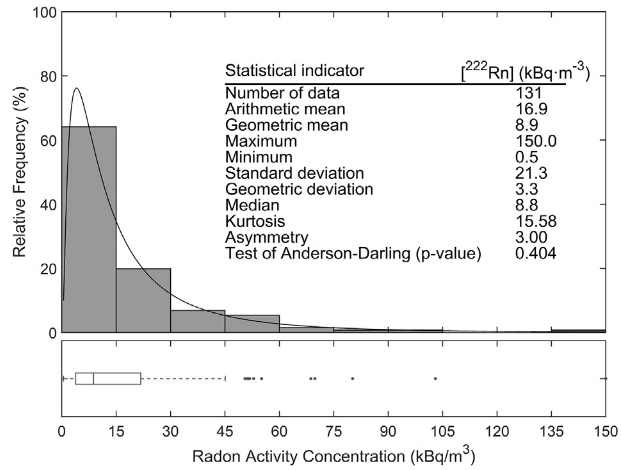


Fig. 2. Histograms and box plot of the data distribution of radon activity concentration in the soil and statistical indicators for Gran Canaria.

As can be seen in Fig. 2, the distribution of data is asymmetric with a higher frequency of low values, ranging from 0.5 to 150 kBq/m^3. The distribution has a median of 8.8 kBq/m^3, an arithmetic mean of 16.9 kBq/m^3 and a standard deviation of 21.3 kBq/m^3. The box plot quantitatively shows that most values correspond to relatively low concentrations (75% of the data are below 20 kBq/m^3), with few values of high concentration (8% of data above 50 kBq/m^3). The kurtosis and asymmetry values (a.k.a. skewness) confirm that the data follow a non-normal and non-symmetric distribution. The theoretical distribution that better approximates to the experimental distribution is the log-normal distribution, attaining a coefficient of determination R^2 of 0.9948. The Anderson-Darling test was applied, and the result did not reject the null hypothesis at a 5% significance level, that the distribution of data follows a log-normal distribution, with a p-value of 0.404. This theoretical distribution is widely utilised in Geostatistics for the study of the spatial distribution of minerals and formation of soils (Rendu, 1979).

Fig. 3A shows the map of radon activity concentration in the soil of Gran Canaria, computed by ordinary kriging interpolation of the measured data. The empirical semivariogram used for Krigging is shown in Fig. 3B. It shows a clear structure in which no relevant nugget effect (intercept of the empirical semivariogram) can be identified. The empirical semivariogram was fit to a theoretical exponential model, also shown in this Fig. 3B, from which it can be inferred that the values are correlated up to a distance of approximately 5 km. This may be considered the range over which geology tends to considerably vary.

This map partly reflects the geological structures explained in Section 2. Higher values of radon in the soil were found in the centre and northeast of the island (the maximum with a value of 150 kBq·m^{-3} is situated in the Tamadaba-Altavista Massif), approximately within the limits of the zone named "Caldera de Tejeda" (Tejeda Basin) in Fig. 1B. The lithology associated with these zones principally corresponds to materials from the first and second magmatic cycle of the island (Ancient cycle and Roque Nublo cycle). A proportion of these materials comprises plutonic rocks (such as dykes and plutons), volcanic magmatic rocks (for instance ignimbrites) and differentiated magmatic rocks (trachytes, phonolites and syenites), where radioactive trace elements such as uranium or thorium, elements from rare earths and major minerals such as potassium are accumulated. Given that a higher concentration of radon precursors can be found in the bedrock in these areas, radon activity concentration in the soils was expected to be high, as was duly observed. Points with intermediate levels (between 30 and 50 kBq·m^{-3}) were also found in the southern zone, in the extra-basin zone (corresponding to rhyolitic structures of the Miocene edifice), and in the north, associated to post-Roque Nublo volcanic cones.

The zones with low radon activity concentration in the soil are localised, as expected, primarily in the northeastern and eastern parts of the island, corresponding to the most recent volcanism (post-Roque Nublo), which as previously mentioned is characterised by the preponderance of basic rocks. The zones of fluvial and aeolian deposits present very low concentrations of both radon and natural radionuclides.

Fig. 4 shows the box plots of the data of radon activity concentration in the soil corresponding to the various geological categories defined for Gran Canaria according to the classification in Table 1. On each box, the central line indicates the median (second quartile or Q_2), and the bottom and top edges of the box indicate the first and third quartiles (Q_1 and Q_3), respectively. The ends of the whiskers represent the lowest datum within 1.5 IQR (Interquartile range: Q_3–Q_1) of the lower quartile, and the highest datum within 1.5 IQR of the upper quartile, and therefore not considered outliers. The outliers are plotted individually as

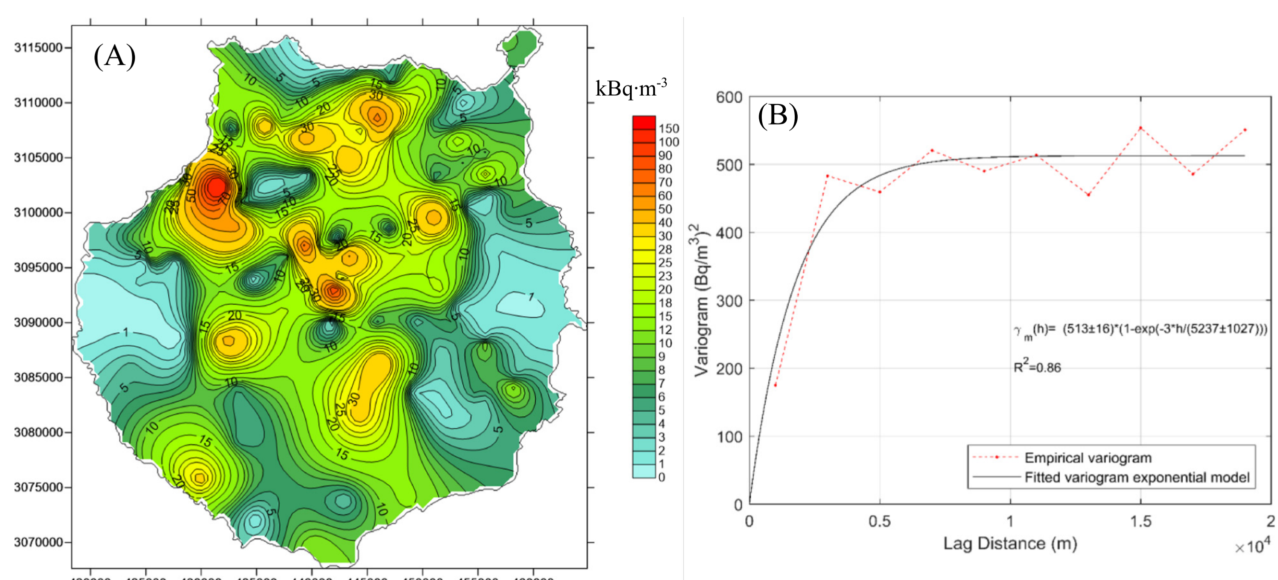


Fig. 3. (A) Map of radon activity concentration in the soil at 50 cm depth (UTM coordinates are given in m) in Gran Canaria. (B) Corresponding semivariogram. Lag distance (h) is the distance that separates the location of two measuring points.

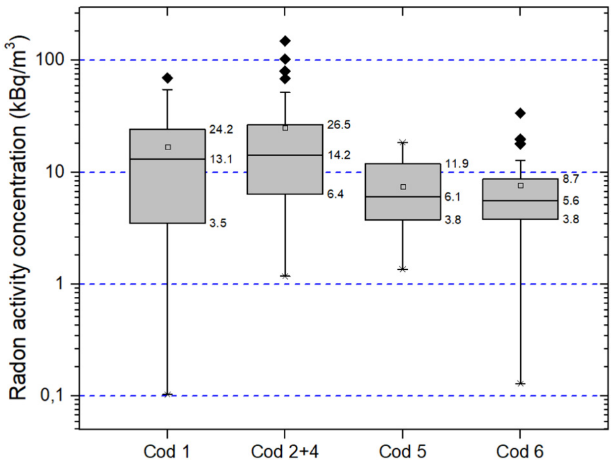


Fig. 4. Box plot of radon activity concentration in different geological formations of Gran Canaria.

dots out of such an interval. Additionally, the point within the boxes represents the arithmetic mean. The allocation of each sampling point to a geological code has been performed utilising the digital geological maps provided by the cartography of SDI- GRAFCAN S.A. As can be seen, the largest values are found to be associated to the geological codes 2 and 4, corresponding to the soils in which intermediate and acid rocks predominate (volcanic and plutonic). Given that 75% of the obtained values for codes 2 and 4 are below 20 $kBq \cdot m^{-3}$, the arithmetic mean of the distribution (25.2 $kBq \cdot m^{-3}$) is clearly affected by the extremal values encountered, presenting a distribution with a very high degree of dispersion. Therefore, the geometric mean, a better indicator in case of lognormal distributions, is considerably lower (13.4 $kBq \cdot m^{-3}$). Nevertheless, this value is considerably higher than the geometric means corresponding to the geological codes 1 (ultrabasic and basic volcanic rock, 7.9 $kBq \cdot m^{-3}$), 5 (Sedimentary littoral deposits, 5.6 $kBq \cdot m^{-3}$) and 6 (Sedimentary terrestrial deposits, 4.9 $kBq \cdot m^{-3}$), as expected due to the geology of the different zones.

However, the complex geography of the island, its varied geological composition, the intrusions of different types of rocks due to the island volcanic genesis and subsequent rejuvenation cycles generally imply considerable local inhomogeneity, which accounts for the fact that intermediate values (i.e. values between 20 and 60 $kBq \cdot m^{-3}$) appear to be associated to points with geological codes 1 (basic rocks), 2 and 4 (acid rocks).

4.1.2. Fuerteventura and Lanzarote

Fuerteventura and Lanzarote are part of the same insular volcanic edifice (Carracedo, 2011; Zazo et al., 2002) and exhibit similar geological characteristics, with low levels and little variability of environmental radioactivity (Arnedo et al., 2017). Therefore, a less comprehensive sampling was performed for these islands. The measurement campaigns took place between the years 2012 and 2015, sampling a total of 71 points covering all geological zones.

Fig. 5 shows the histogram of the distribution of radon activity concentration in the soil, as well as the corresponding statistical indicators for Fuerteventura and Lanzarote. The distribution of data for Fuerteventura ranges from 0.5 to 35.1 kBq/m^3, whereas for Lanzarote it ranges from 0.1 to 12.1 kBq/m^3. Thus, Fuerteventura exhibits overall higher values than Lanzarote, with medians of 6.3 and 2.9 kBq/m^3, respectively. In both cases these values are considerably lower than those from Gran Canaria. Fuerteventura also presents higher dispersion than Lanzarote, with a standard deviation of 8.5 kBq/m^3 as opposed to 3.1 kBq/m^3. In this case, the theoretical distribution that approximates best to the experimental distributions is again the log-normal distribution for Fuerteventura (attaining a coefficient of determination R^2 of 0.7659) and the exponential distribution for Lanzarote ($R^2 = 0.8498$), assertions which were confirmed again by the test of Anderson-Darling.

Fig. 6 displays the map of radon activity concentration in the soil for Fuerteventura and Lanzarote, computed by interpolation by means of the kriging method. For these islands, the exponential model for the empirical semivariogram reaches the maximum variance (sill) at approximately 10 km and 6.4 km, respectively. This fact confirms that both islands are more homogeneous than Gran Canaria as far as radon in the soil is concerned.

Overall, the distribution of radon on Fuerteventura according to the soil map is congruent with the geology of the island as explained in Section 2. Low values of radon activity concentration in the soil are found across most of the island, which is dominated by igneous basic rocks (such as basalts) and ultrabasic (including pyroxenites and peridotites) from the basal complex and the subaerial volcanism unity (Miocene and recent volcanism). Two local maxima are located in the basal complex in zones of differentiated rocks (especially syenites, trachytes and carbonatites), in the massif of Betancuria and at the mountain of Tindaya (trachy-rhyolites). A third local maximum was found outside the basal complex, located over deposits (gravitational slides) of slopes in the ravine of Pozo Negro, and belonging to subaerial volcanic edifices from the cycle of Miocene volcanism (central edifice) (Ancochea et al., 1996), whose soils are principally conformed by basalts, as well as to a lesser extent by series of differentiated formations, middle-alkaline and peralkaline-trachytes.

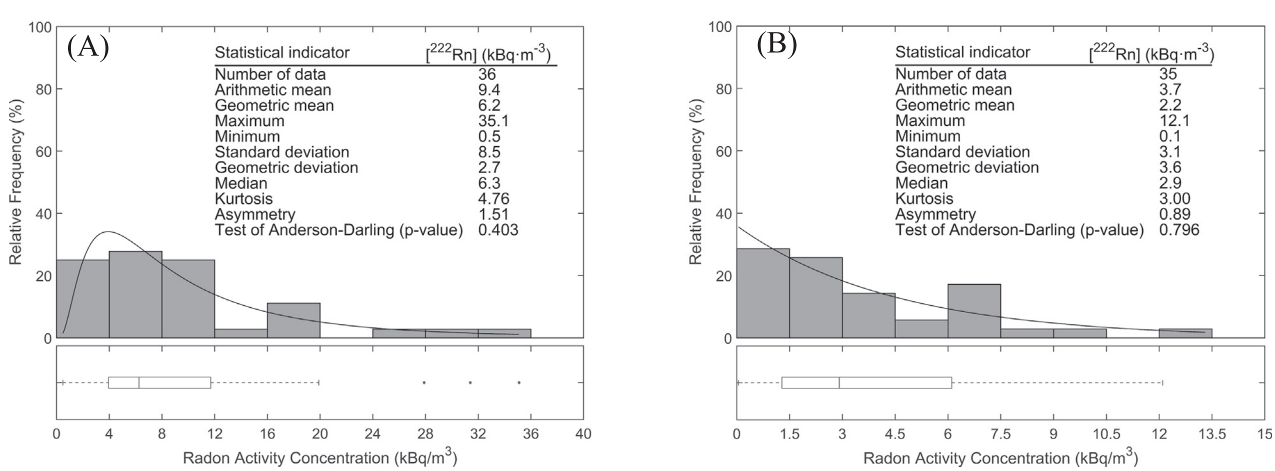


Fig. 5. Histograms and box plot of radon activity concentration in the soil and statistical indicators for Fuerteventura (A) and Lanzarote (B).

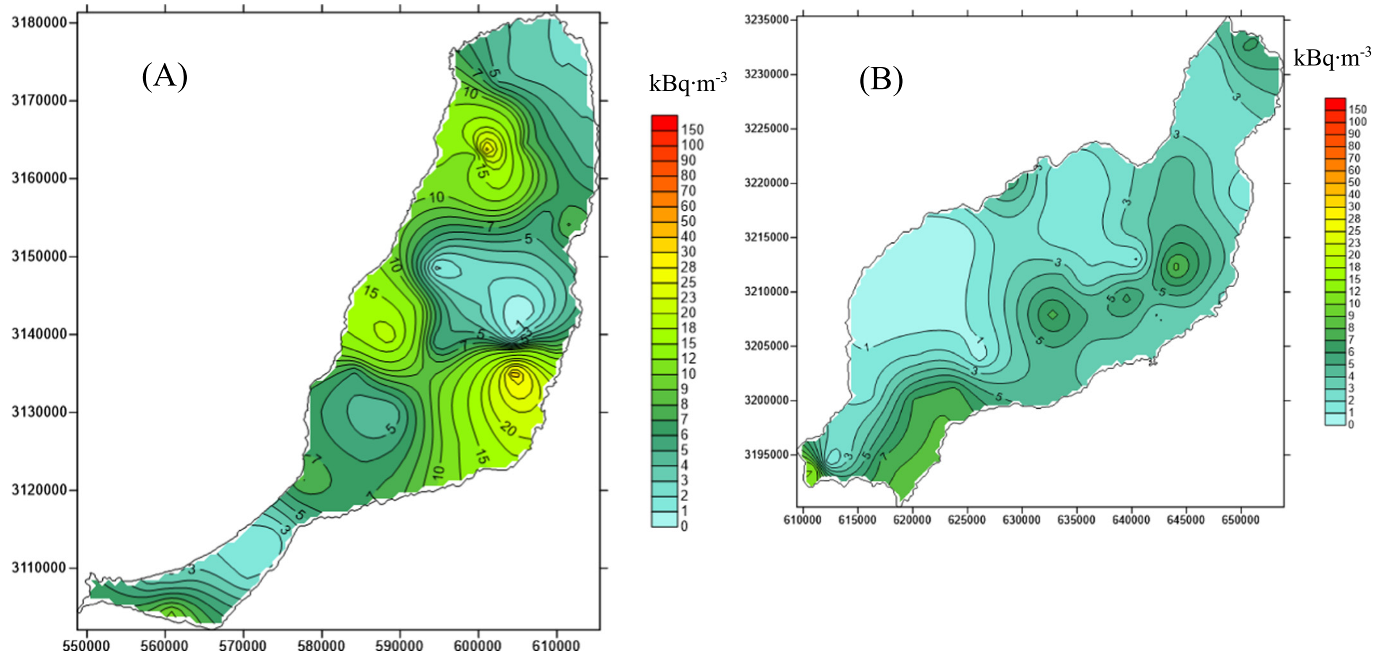


Fig. 6. Maps of radon activity concentration in the soil at 50 cm depth in Fuerteventuta (A) and Lanzarote (B) (UTM coordinates are given in m).

Lanzarote presents the lowest soil radon activity concentration and variability of the islands studied. The values obtained accord with the geology of the island, in which 98% of the rocks are basaltic, with very little presence of radioactive minerals (olivine and pyroxene), there being no natural radionuclides in the volcanic glass. The maximum values are found in the south of the island, in the vicinity of Montaña Roja, a 1.2 million-year-old volcano located above the emerged part of the Miocene building of Los Ajaches (14 Ma) and largely constituted of alkaline basalt and trachybasalt (Zazo et al., 2002), and related to the group of volcanoes belonging to the quaternised fissural volcanism (group of volcanoes in the area of Teguise) (Carracedo and Rodríguez Badiola, 1993). Suárez Mahou and Fernández Amigot (1996) have suggested that a proportion of thorium of 0.1 ppm and 0.02 ppm of uranium can be found in ultrabasic igneous rocks.

Fig. 7 shows the box plot of the data of radon activity concentration in the soil corresponding to the various geological categories that exist on Fuerteventura and Lanzarote, defined according to the classification in Table 1. Again, in Fuerteventura local maxima correspond to codes 2 and 4. The uniformity of Lanzarote can be appreciated in Fig. 7B, where only soils corresponding to codes 1 "Ultrabasic and basic rocks" and 5 "Sedimentary littoral deposits" can be identified.

4.2. Soil gas permeability

As previously mentioned, soil gas permeability is a fundamental parameter that can prove decisive for the release of radon to the atmosphere or its accumulation in the soil (Castelluccio et al., 2010). A greater degree of permeability permits the upward migration of radon, facilitating its exhalation to the atmosphere, and consequently a smaller amount will remain in the ground. In this work, the range of permeability values was divided into three categories following the thresholds established by Neznal and Neznal (2005) on the basis of their experience, as displayed in Table 2.

Fig. 8A uses box plots to summarise the distribution of soil gas permeability data obtained for the three islands. It should be noted that the measurement campaigns of Gran Canaria, Fuerteventura and Lanzarote were carried out in periods of very low rainfall in the summer and autumn, in an effort to make the measurement conditions as homogeneous as possible. As can be observed, the soils of the three islands are

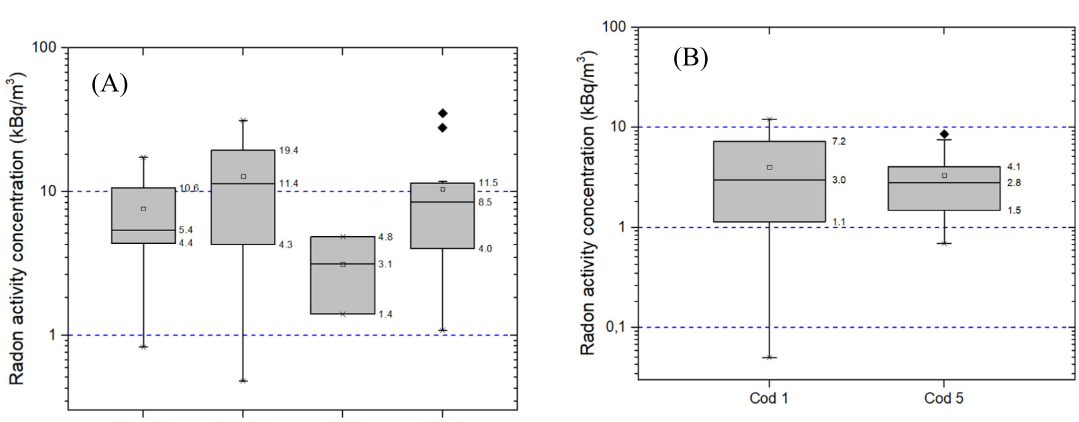


Fig. 7. Box plots of radon activity concentration on different geological formations of Fuerteventuta (A) and Lanzarote (B). (UTM coordinates are given in m).

Table 2
Classification of the soil gas permeability (Neznal and Neznal, 2005).

Class	Permeability (m^2)
High	$k > 4.0\ 10^{-12}$
Medium	$4.0\ 10^{-12}\ m^2 \geq k \geq 4.0\ 10^{-13}$
Low	$k < 4.0\ 10^{-13}$

mostly located in the middle-high permeability range. Gran Canaria presents 52% high-permeability soils, relative to Fuerteventura with 39% and Lanzarote with only 35%. Only Gran Canaria and Fuerteventura have low-permeability soils, with percentages of 18% and 25%, respectively. Again, Lanzarote is the most homogeneous island as far as permeability is concerned, with 65% of its soils exhibiting medium permeability.

Fig. 8B to D show the maps of soil gas permeability for the three islands. In Gran Canaria, the soils with low permeability are mainly located in the central and northwestern zones, corresponding to the most eroded environments on the island, which in many cases exhibits very little soil. In the north and east of the island, soils with high permeability predominate. In the southern zone of the island, the sampled soils have medium and high permeabilities. However, regarding the interpretation of these results, it is necessary to be cautious, for it is customary on the island of Gran Canaria to move soils from one area to another for crops. Even though attempts were made to conduct measurements on lands unaltered by humans, some densely populated areas exist in which the totality of measurements cannot be guaranteed to have fulfilled such a condition. A significant proportion of the territory of Fuerteventura corresponds to basalts, which exhibit a high level of permeability. Low-permeability soils appear within the basal complex, corresponding to areas of heavily eroded soils. The soils formed by sands and marine sediments have medium and high permeability. Overall, and as mentioned above, in Lanzarote the soils have medium permeability, and as is true of Fuerteventura, there are areas of high permeability associated with old basalts.

4.3. Geogenic Radon Potential and radon-prone areas

The Geogenic Radon Potential (GRP) is a magnitude defined as providing a quantitative measurement regarding "what the earth delivers" in terms of radon (Bossew, 2015). Nevertheless, a single definition of GRP does not exist, with diverse parameters used by different authors and national regulators, such as in the Czech Republic (Neznal et al., 2004), Germany (Kemski et al., 2001), France (Ielsch et al., 2010), Switzerland (Piller and Johner, 1998; Johner and Surbeck, 2001) and Sweden (Åkerblom, 1986). Nevertheless, most authors define GRP in terms of a combination of radon activity concentration in soils and soil gas permeability. In this work we adopt a heuristic approach similar to

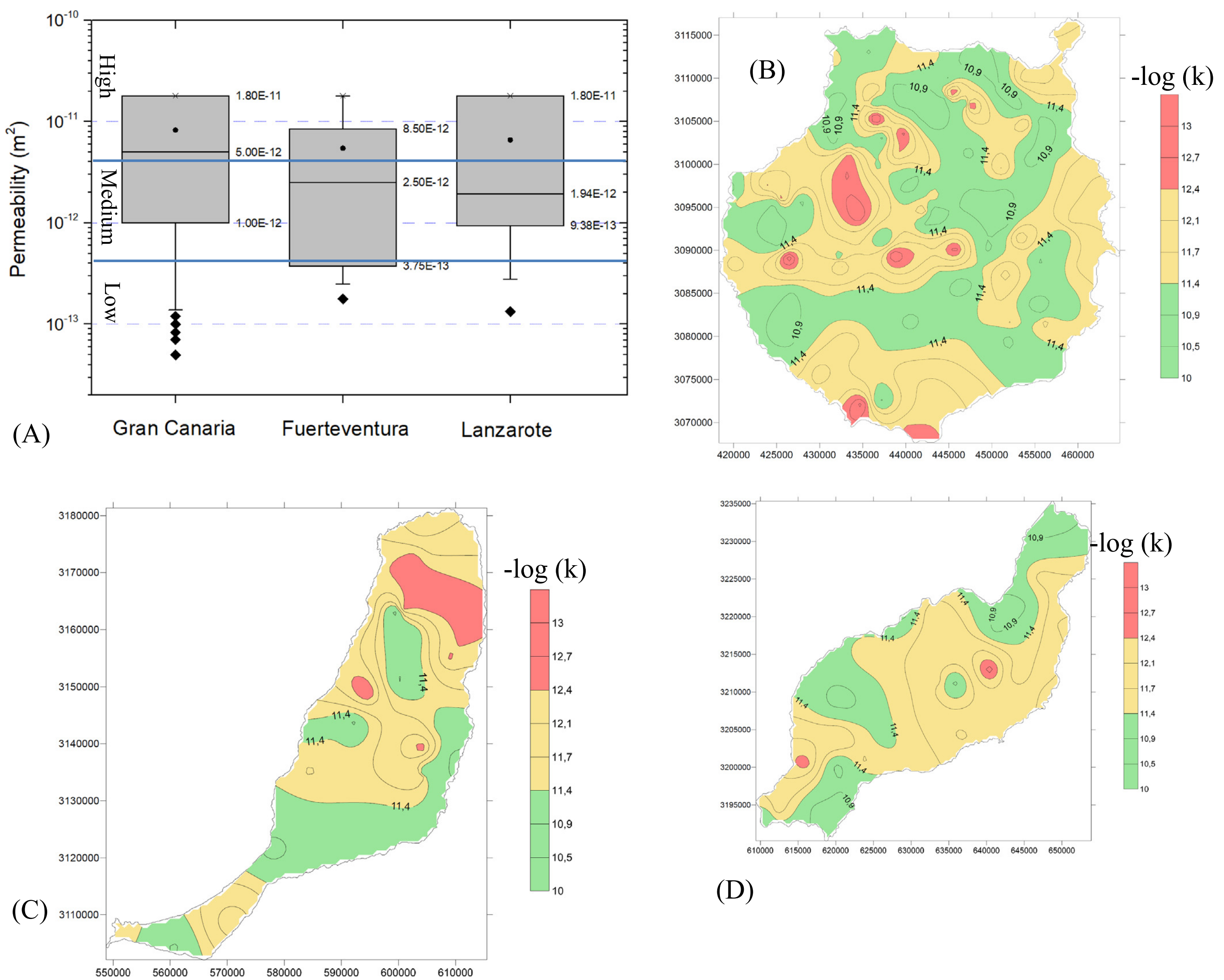


Fig. 8. (A) Box plots of soil gas permeability in the three islands. Maps of permeability of (B) Gran Canaria, (C) Fuerteventura, (D) Lanzarote.

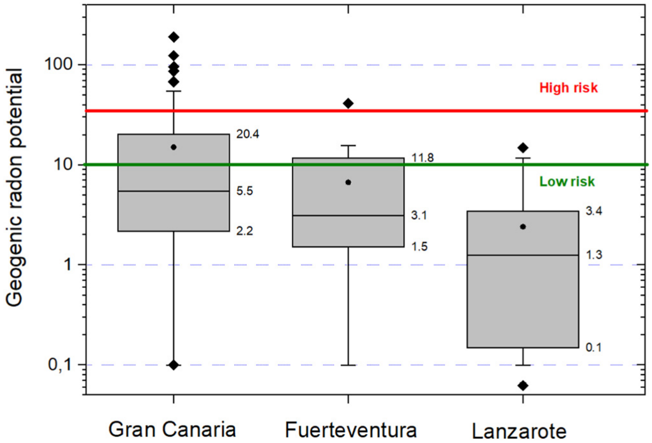


Fig. 9. Box plot of Geogenic Radon Potential in Gran Canaria, Fuerteventura and Lanzarote.

that proposed by Neznal et al. (2004), defined through Eq. (1), where C_{Rn} is the radon activity concentration in the soil (kBq m^{-3}) and k is the soil gas permeability (m^2).

$$GRP = \left(\frac{C_{Rn}}{-\log_{10}(k) - 10}\right) \quad (1)$$

Maps of Geogenic Radon Potential represent useful tools to identify regions where there is a higher probability of significant levels of indoor radon appearing due to natural sources, when few or no previous indoor radon measurements are available. In addition, the GRP maps can be used to prioritise those areas where it is deemed necessary to reinforce indoor radon measurements as high levels of activity concentration in dwellings are expected.

According to the 2014 Basic Safety Standards (EU-BSS) for protection against ionising radiation of the European Union, a Radon Prone Area (RPA) is defined as a geographical zone or administrative region where the radon concentration (as an annual average) in a significant number of buildings is expected to exceed the relevant national reference level. Even though the BSS definition is based on indoor radon concentration, several regulators instead (or in addition) opt to estimate it by means of other radon-related variables. One common approach is to use a Radon Index based on the values of GRP to categorise the radon risk of a territory. Based on numerous years of extensive research, in the Czech Republic three categories for Radon Prone Areas depending on the values of the GRP has been established: R.I. Low (GRP < 10), R.I. Medium (10 < GRP < 35) and R.I. High (35 < GRP).

Following the Czech classification, the soils of Fuerteventura and Lanzarote are characterised on average (arithmetic) by low values of GRP, while the soils of Gran Canaria would be characterised on average (arithmetic) by medium GRP. If the three islands were represented by their geometric mean, it would lower the classification of Gran Canaria towards low GRP as well. This fact was illustrated in Fig. 9, displaying the box plots of radon potential by islands. The geometric means of the three distributions are in the low-risk zone, whereas the high-risk points correspond to the high-value data of the islands of Gran Canaria and Fuerteventura. For this reason, the arithmetic mean of the island of Gran Canaria is at medium risk. It is also interesting to see how virtually the entire island of Lanzarote exists below the low risk line.

Neznal et al. (2004) have proposed a graphic representation for GRP, as displayed in Fig. 10A. This diagram is a modification of that proposed by Barnet (1994). In this diagram, the empirical criteria established by Neznal et al. (2004) are represented using straight lines in order to delimit the low-medium and medium-high radon index (RI) zones. Following these criteria on Gran Canaria, we found that 63.4% of the measured points are in the low RI category, 36.6% belong to the medium RI category, and the remaining 10.7% in the high RI category. This representation clearly explains the fact that points with the same concentration of radon, but different levels of permeability can be associated with different levels of radon risk. It can also be appreciated that for radon activity concentrations above 35–40 kBq m^{-3}, soils cannot be classified as low risk. Hence, this is an outstanding tool that facilitates decision-making and permits exclusion of the possible presence of high levels of indoor radon due to the surrounding soils and the geological environment of a house. In Fig. 10A the GRP values were labelled by their simplified geological code. As can be seen, most of the points located in the high-risk area of the diagram correspond to code 2 (Intermediate or Acidic Volcanic Rocks), as had been expected given the geochemical characteristics of this substrate. Some points belonging to basaltic soils (code 1) with fairly low values of radon in the soil are also located in the high-risk area due to the fact that they are situated in zones of high permeability.

These facts are reflected in Fig. 10B, where the map of GRP (in natural logarithmic scale) for Gran Canaria is shown. This map can be used as an estimator of the Radon Prone Areas for the island. A colour code of nine levels was adopted corresponding to three main categories of R.I defined above. The low-medium and medium-high R.I limits are, respectively,

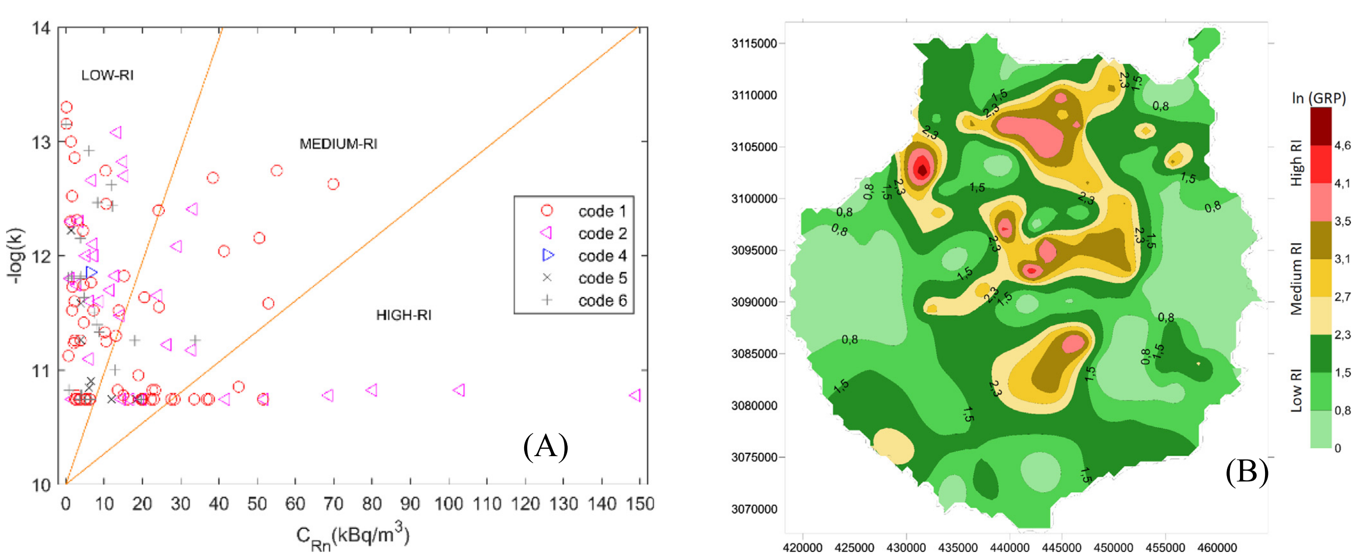
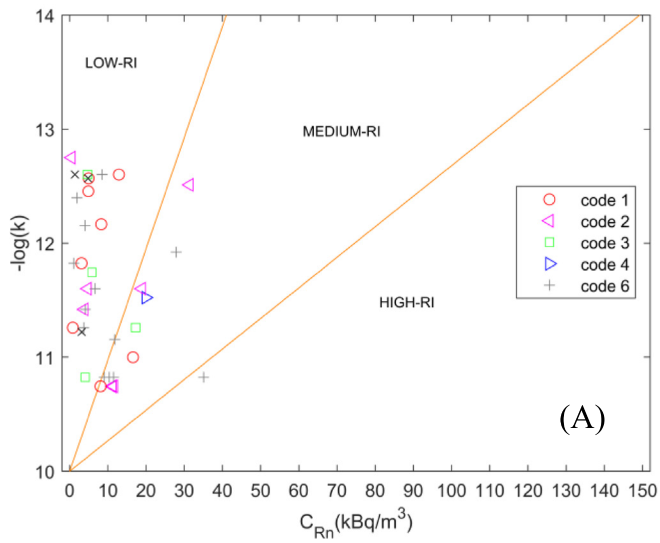


Fig. 10. (A) Diagram of radon risk for Gran Canaria. (B) Map of Radon Prone Areas of Gran Canaria.



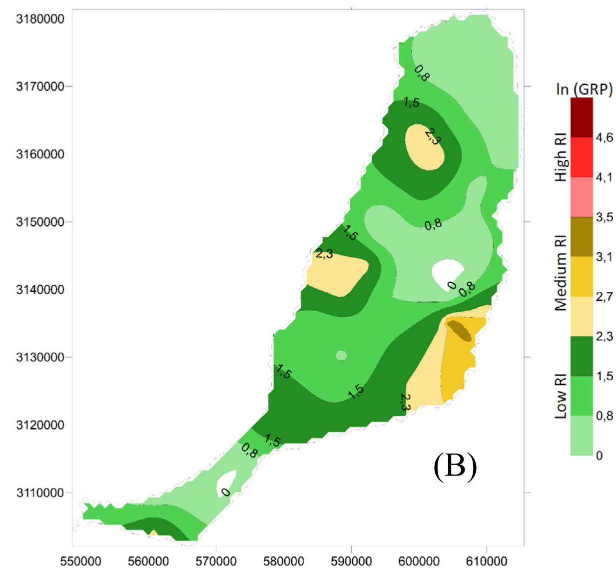


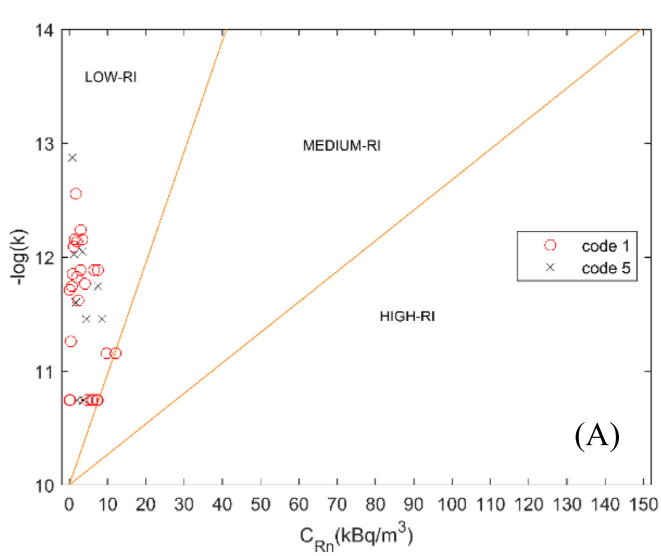
Fig. 11. (A) Diagram of radon risk for Fuerteventura. (B) Map of Radon Prone Areas of Fuerteventura.

2.203 and 3.555, corresponding to the natural logarithm of the aforementioned R.I. limits (10 and 35). This map shows that a substantial part of the surface of the island can be classified as low radon risk. These areas mainly correspond to basaltic soils and are found in the periphery of the island. The medium- and high-risk zones are located in the north-centre of the island, where the presence of soils coming from intermediate or acidic volcanic rocks (trachyites - phonolites) is more significant. These final zones cover parts of the municipalities of Arucas, Firgas, Moya, Santa Brígida, Teror, San Mateo, Valleseco, Tejeda, Artenara, Agaete, Valsequillo, San Bartolomé de Tirajana and Mogán, which contain approximately 125,000 inhabitants (15% of the total population of the island). Dwellings in these areas primarily constitute traditional village single-family land houses and cave houses, buildings that are more prone to the accumulation of indoor radon than the apartment buildings common in large cities in Spain.

Fig. 11A displays the radon risk diagram for Fuerteventura, where all values except one are concentrated in the area of low- or medium-low risk of radon. The only exception observed in the area of high risk (although almost at the medium-high border) is mainly due to the high permeability of the soil at that point, as the radon activity concentration is below 40 kBq/m^3. Fig. 11B shows the RPA map for Fuerteventura. As expected according to its geology, most of the island presents a low-radon risk profile, albeit slightly higher in zones of the basal complex where some rocks from codes 2 and 4 outcrop. Regarding Lanzarote, Fig. 12A demonstrates that nearly all of the values are located in the low radon risk zone, with a few points situated above the straight line delineating the transition between the low-risk and medium-risk zones, not due to high radon concentrations, but to corresponding high-permeability values. Accordingly, the map of RPA of Lanzarote shown in Fig. 12B presents the only low-risk category throughout the territory.

5. Summary and conclusions

The use of a Risk Index based on the GRP for the zonal classification of the Eastern Canary Islands permits classification of the islands of Lanzarote and Fuerteventura as being areas of low radon risk, whereas Gran Canaria has a large area classified as being at potentially high radon risk. The maps developed in this work may prove a highly useful



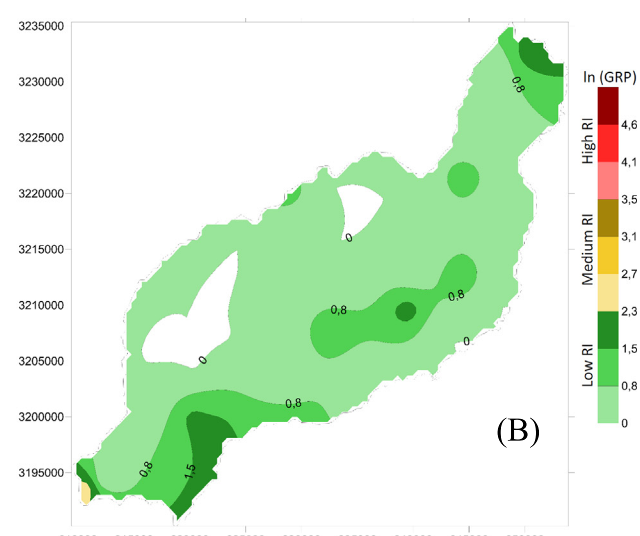


Fig. 12. (A) Diagram of radon risk for Lanzarote. (B) Map of Radon Prone Areas of Lanzarote.

tool for the authorities responsible for the monitoring and control of radon exposure in the territory of the Canary Islands, both when preparing action plans for sampling and preventing the appearance of indoor radon in dwellings and workplaces. The Spanish Building Code is currently under revision in order to include protection against radon by means of classifying the country and building site in question under several radon risk categories, as well as the inclusion of radon-resistant techniques in new buildings. The GPR maps based on soil properties are ideal for this task and are already being used in several countries such as the Czech Republic and Germany. The zonal classification obtained accords with the indoor radon risk map of the Canary Islands included in the European Radon Map developed by the EU-JRC (Gruber et al., 2013). This map has been developed from direct indoor measurements in houses, and although it has poor resolution in the island territory, it coincides with the classification of Lanzarote and Fuerteventura as non-prone areas and the centre of Gran Canaria as a radon-prone area. This fact is also reflected in the map of radon potential in Spain developed by the Nuclear Safety Council of Spain based on terrestrial gamma radiation, geology and indoor radon data (García-Talavera et al., 2013; Sainz Fernández et al., 2017; Consejo de Seguridad Nuclear (CSN), 2018).

It is important to note that the classification of a certain area as being at low risk of radon does not mean that radon concentrations above the reference level of 300 Bq/m^3 will not occur in any building; it simply means that this is unlikely. In the same way, a high-risk area does not imply that any house built in the zone will necessarily exceed the established reference level.

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