



Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

Spanish experience on the design of radon surveys based on the use of geogenic information

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ARTICLE INFO

Article history:

Received 6 November 2015

Received in revised form

24 June 2016

Accepted 5 July 2016

Available online xxx

Keywords:

Radon mapping

Spain

Geology

Lithostratigraphy

Lithology

ABSTRACT

One of the requirements of the recently approved EU-BSS (European Basic Safety Standards Directive, EURATOM, 2013) is the design and implementation of national radon action plans in the member states (Annex XVIII). Such plans require radon surveys. The analysis of indoor radon data is supported by the existing knowledge about geogenic radiation. With this aim, we used the terrestrial gamma dose rate data from the MARNA project. In addition, we considered other criterion regarding the surface of Spain, population, permeability of rocks, uranium and radium contain in soils because currently no data are available related to soil radon gas concentration and permeability in Spain. Given that, a Spanish radon map was produced which will be part of the European Indoor Radon Map and a component of the European Atlas of Natural Radiation. The map indicates geographical areas with high probability of finding high indoor radon concentrations. This information will support legislation regarding prevention of radon entry both in dwellings and workplaces. In addition, the map will serve as a tool for the development of strategies at all levels: individual dwellings, local, regional and national administration.

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1. Introduction

The design of the Spanish indoor radon map is part of the framework of the Joint Research Centre (European Commission) plan for the elaboration of the European Indoor Radon map, as a part of the Atlas of Natural Radiation (Dubois et al., 2010; Tollefsen et al., 2011; Bossew et al., 2015; De Cort et al., 2011). This atlas will support policies in the field of public health, and it will contribute to increase the general public's awareness of the annual dose due to natural radioactivity.

A number of studies in various countries have proved that there is a clear correlation between exposure to radon inside buildings and the risk of developing lung cancer (ICRP, 2010, 2011). Radon gas is responsible for between 3 and 4 per cent of deaths caused by this illness in the first world (IAEA, 2011; WHO, 2009), being the main source of ionizing radiation (EURATOM, 1990, 1996, 2013; ICRP, 1994). Therefore, it is crucial to determine the areas where there

is a greater probability of finding buildings with higher radon concentrations, as well as to analyse the variables which affect radon concentrations inside buildings.

Indoor radon concentration varies geographically. This is due to the large number of factors that affect radon appearance in buildings, such as the geology of the areas upon which buildings are constructed, soil permeability, specific rock characteristics, the meteorology and topography of the region, the proximity of active fault lines, the materials employed in construction, the design features of the buildings and the lifestyle habits of the occupants (García Talavera et al., 2013a). In our work we used a geographic information system (GIS) which enabled us to capture, store, create searches, analyse and visualize the statistical data we obtained. In order to adapt to the design of the European Indoor Radon map, Spain followed national (CSN, 2012a; CSN, 2012b) and international legislation (ICRP, 1993, 2009, 2014).

This paper aims to produce a radon map of the Spanish territory that shows the probability of finding areas with levels of radon indoors, and is related to the European legislation that has to be implemented in the member states before the end of 2018

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(EURATOM, 2013). This map will be a very useful instrument for applying the requirements of European legislation on the radon issue at all administrative levels: national, regional and local.

2. Methodology

2.1. Definition of the grid

The grid was generated using the programme ArcGIS where the extreme corners are defined by established coordinates in the European Datum 1950 UTM Zone 30N (ED50) projection system: Peninsular Spain NW (433193,87; 4834808,73) SE (692906,43; 3961069,60) Canary Islands NW (692915,11; 3320469,29) SE (698454,23; 2988008,14). These coordinates were converted to longitude-latitude ED50 (decimal degrees) to generate a 10×10 km² grid of the Spanish surface in Google Earth format (.kml). The ED50 system is an old geodetic reference system used in Europe and has been used in coexistence with the ETRS89 in Spain until 2015. The parameters of this system are defined in the ArcGIS.

In order to follow a similar scheme as other EU member countries, we began working with a continental level projection system (GISCO-Lambert Acimutal Equal Area) suggested by the Joint Research Centre of the European Commission (Dubois et al., 2010; Tollefsen et al., 2014). The Lambert Azimuthal Equal Area projection is a planar projection, which means that map data are projected onto a flat surface.

In this way, measurements between countries can be homogenized and the so-called border effect avoided. We took into account those parameters set by the Joint Research Centre to convert the coordinates ED50 to GISCO-LAEA. Therefore, we obtain the following limits for 10×10 km² grid: Peninsular Spain NW (–1500000; –301000) SE (–351000; –1350000) Canary Islands NW (–2700000; –1900000) SE (–2150000; –1650000).

To define the working area, we used the administrative boundaries provided by the National Geographic Institute (IGN, 2014). Thus, we generated a total number of 5478 cells of 10×10 km² surface. For each cell, an identifying code was created and its centroid in meters (“x” and “y” coordinates) calculated. The statistical data obtained from the measurements were georeferenced to the centroid.

2.2. The measurement campaigns

The Spanish indoor radon map comprises, to date, 9211 measurements, obtained over successive sampling campaigns (Fig. 1). In each campaign, a series of measurements for each cell was defined, taking into account superficial, population, external gamma dose (MARNA Project) (Sainz Fernández et al., 2014; Suarez Mahou and Fernandez, 1997, Suárez Mahou et al., 2000; Quindós et al., 2004; Quindós et al., 2008) and lithostratigraphic criteria. To decide the number of measurements per cell, it was essential to prioritise objectives and establish criteria. The decision on which

criteria to use was made taking into account the objectives behind the European Radon map.

1. Surface criterion: The whole Spanish territory had to be covered by, at least one measurement per 10×10 km grid cell. 478 cells out of 5478 total number of cells are inhabited and no data were collected. Thus, the lowest number of possible measurements was done over 5000 cells.
2. Population criterion: In the first measurement campaign, an extra measurement was done for each town with a population exceeding 50,000 inhabitants, based on the Spanish National Statistics Institute (INE, 2014). Hence, an additional 1000 measurements had to be made according to this criterion. In the second campaign this criterion was expanded to require a minimum of 6 measurements in cells including towns with populations larger than 200,000. A further 123 measurements were taken to meet this criterion.
3. MARNA criterion: Considering the importance of the geological factor it was decided to increase the number of measurements in areas with high radon potential. The starting point was the MARNA project (Suárez Mahou et al., 2000). MARNA determines potential radon emissions by taking into account the correlation between ²²⁶Ra concentration in the soil and outdoor gamma dose levels.

This criterion is based from 7400 locations where measurements were performed to determine the potential for radon emission. These locations were linked to the 10×10 km² cells, so-called MARNA cells. The different levels of exposure assigned to each cell correspond to the town with the higher dose. During the first campaign, additional measurements were taken in each cell identified in the MARNA project with a gamma exposure level over 35 nGy/h [4 µR/h]. 2 additional measurements were added for each location that exceeded 35 nGy/h [4 µR/h].

By doing so, 2000 additional measurements were made (Sainz Fernández et al., 2014). Throughout the second campaign, measurements focused on towns with a gamma exposure level between 65 and 122 nGy/h [7.5 and 14 µR/h] (median risk) and

Table 1

Description of the spatial units of interest as used in this work and their lithostratigraphical definition.

Lithostratigraphic unit code (IGME)	Lithostratigraphy definition
130	Limestones, calcoschists and whiteboards
131	Micaschists, gneises, phyllites, quartzites and plasters
133	Micaschists, quartz and gneises
79	Slates, sandstone and quartzite. Series of los Cabos
85	Slates, sandstones and microconglomerates. Slates from Lancia
86	Quartzites, shales and rocks volcanocl. and volcanised. Quartzite from Barrios and FM Oville
91	Slates and sandstones. Huergas slates
104	Quartzites, slates, sandstones, shales, limestones and dolomites. Paleozoic Iberian Aragon
117	Quartzites and slates
369	Shales with interbedded carbonate and gypsum
7	Plutonic basic Hercynian rocks (gabbros, diorites, tonalite, ultramafic rocks)
2	Acid rocks metamorphic (Otogneises, migmatitas). Gn. gland., metarriolitas (Olla Sapo).
127	Phyllites, schists, quartzites, limestones, slates and corneal (metamorphic)
152	Sandstones, sands, sandy limestones, marls, clay and loamy
153	Sandstones, shales and marls
173	Limestone reef, rudistas, bioclastic limestones, dolomites and marl
174	Marl, limestone, clay and dolomites

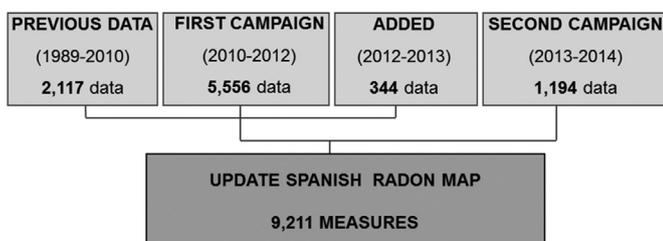


Fig. 1. Description of the sampling campaigns carried out towards the development of the Spanish indoor radon map.

Table 2

Examples of the unit numbers of interest in some Spanish regions and the number of measurements necessary to take. Not all Spanish regions are represented.

Region	Lithostratigraphic unit code of interest (IGME)	Measures to take	Criterion
Andalucía	130-131-133	40	In at least 6 different municipalities (covering 3 units) with a minimum of 6 measures by municipality
Asturias	79-85-86-91	50	In at least 8 different municipalities (covering 4 units) with a minimum of 6 measures by municipality
Aragón	104-117-369	40	In at least 6 different municipalities (covering 3 units) with a minimum of 6 measures by municipality
Extremadura	7	30	In at least 4 different municipalities with a minimum of 6 measures by municipality
Cataluña	2-7-117-127	60	At least 3 municipalities in unit 2, 3 municipalities in unit 7 and 3 to 117 and 127 units. With a minimum of 6 measures by municipality
País Vasco	152-153-173-174	50	In at least 8 different municipalities (covering 4 units) with a minimum of 6 measures by municipality

involved a minimum of 6 measurements for each cell with this dose criterion. In the whole territory, 1655 MARNA cells of this type were found, but the number of measurements taken was reduced to 960 due to limitations in budget.

The towns were classified according to their highest dose starting from 122 nGy/h and arranged in a decreasing order. A selection was made from the highest doses adjusted to the requirement of 6 measurements per cell, obtaining 960 measures. The result was the implementation of the measures in cells between 104 and 122 nGy/h [12 and 14 μ R/h].

4. Lithostratigraphic criterion: Numerous studies support the importance of soil permeability factor in determining the radon potential in buildings (Kemski et al., 2001). Permeability is part of the lithological nature of the rock. The proper way to map this feature is by means of the lithostratigraphic map. It represents the lithostratigraphy homogeneously grouped by similar levels mapping permeability values (García Talavera et al., 2013b).

Following the proposals of the Joint Research Centre (Bossew et al., 2015; De Cort et al., 2011) towards designing a European

wide geological radon map, a series of areas of interest were identified in accordance with lithostratigraphic units for different regions and with that a specification of the number of measurements to be taken per unit and town (see Tables 1 and 2). The permeability is one of the parameters that could be used as proxy of indoor radon (Tollefsen et al., 2014).

By using the lithostratigraphic map from Geological and Mining Institute of Spain (IGME, 2014a; IGME, 2014b) at 1:200,000 scale, which includes the permeability factor of lithological units, units of interest were identified and cross checked with the local and regional cartographic databases (IGN, 2014). This increased the number of towns where measurements were to be taken. In order to identify the cells affected by this criterion, the above mentioned towns were superimposed onto the 10 \times 10 km cell base and an additional 270 measurements were done. Figs. 2 and 3 show the implementation of the previous criteria during the two measuring campaigns.

2.3. Radon measurements

The measurements were carried out using CR-39 track-etch detectors over an exposure period of 3–6 months. Having

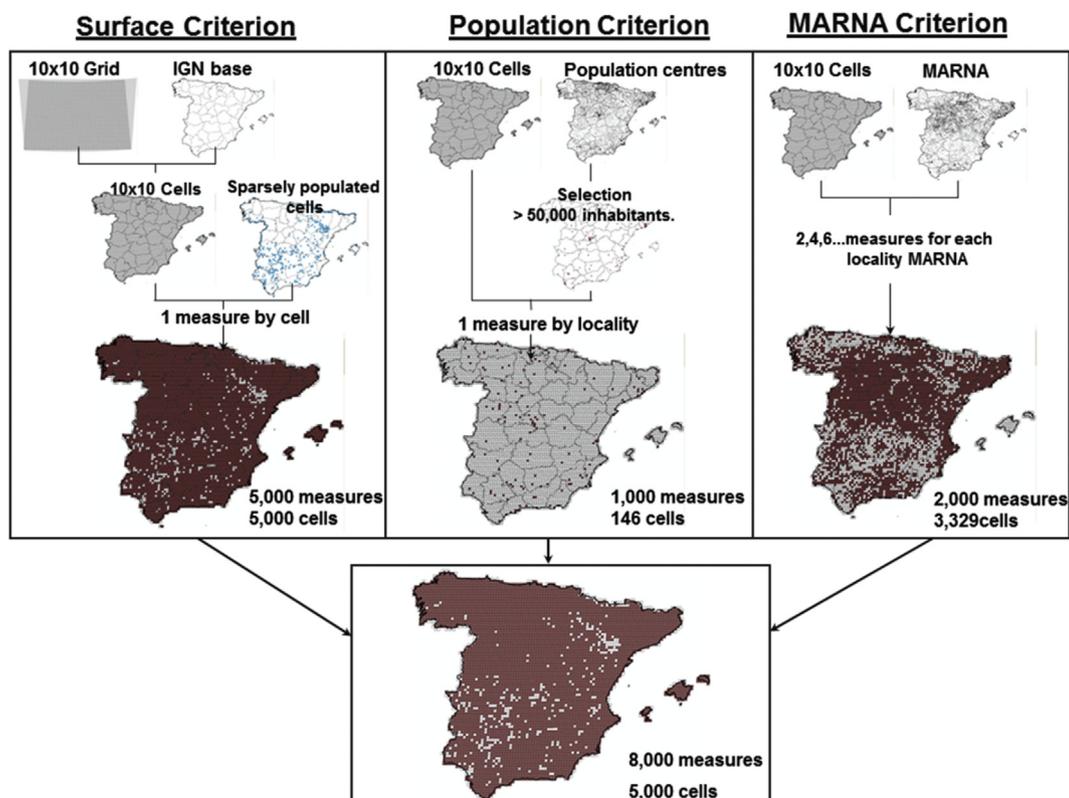


Fig. 2. Schematic view of the criteria used in the first campaign resulting in 8000 measurements.

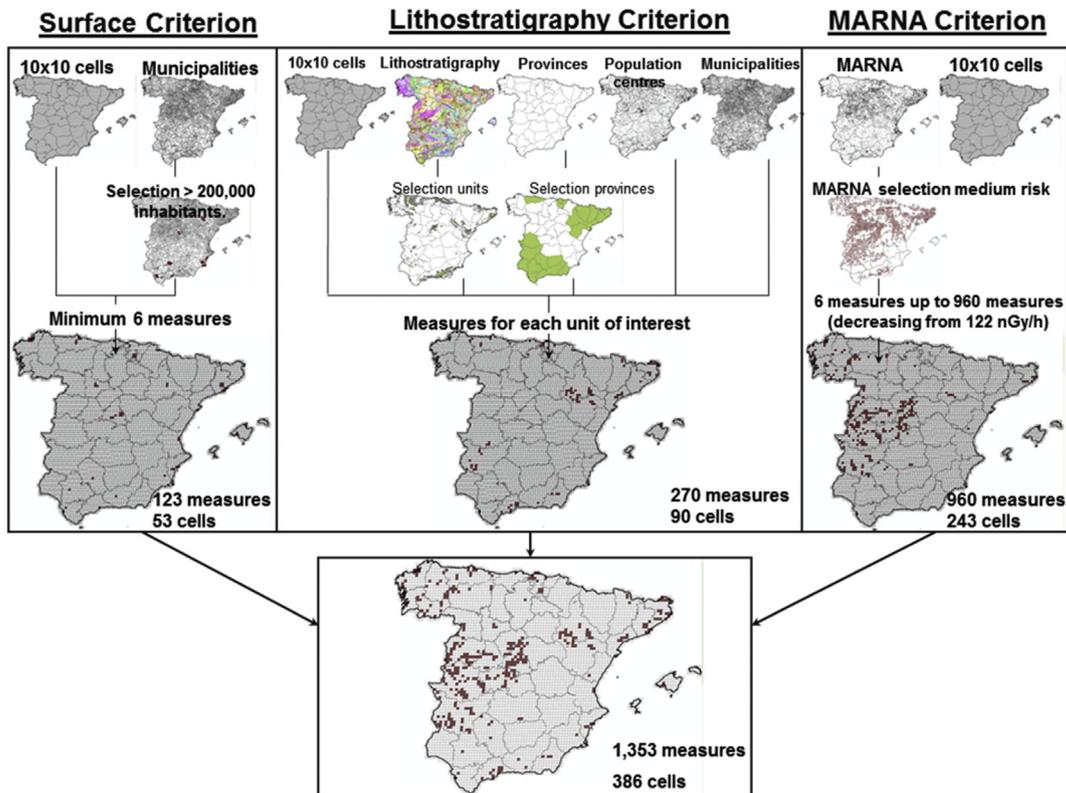


Fig. 3. Extension of measurements to create the Spanish indoor radon map by including additional criteria: surface, lithostratigraphy and MARNA.

Identification of the locality and registration of detector

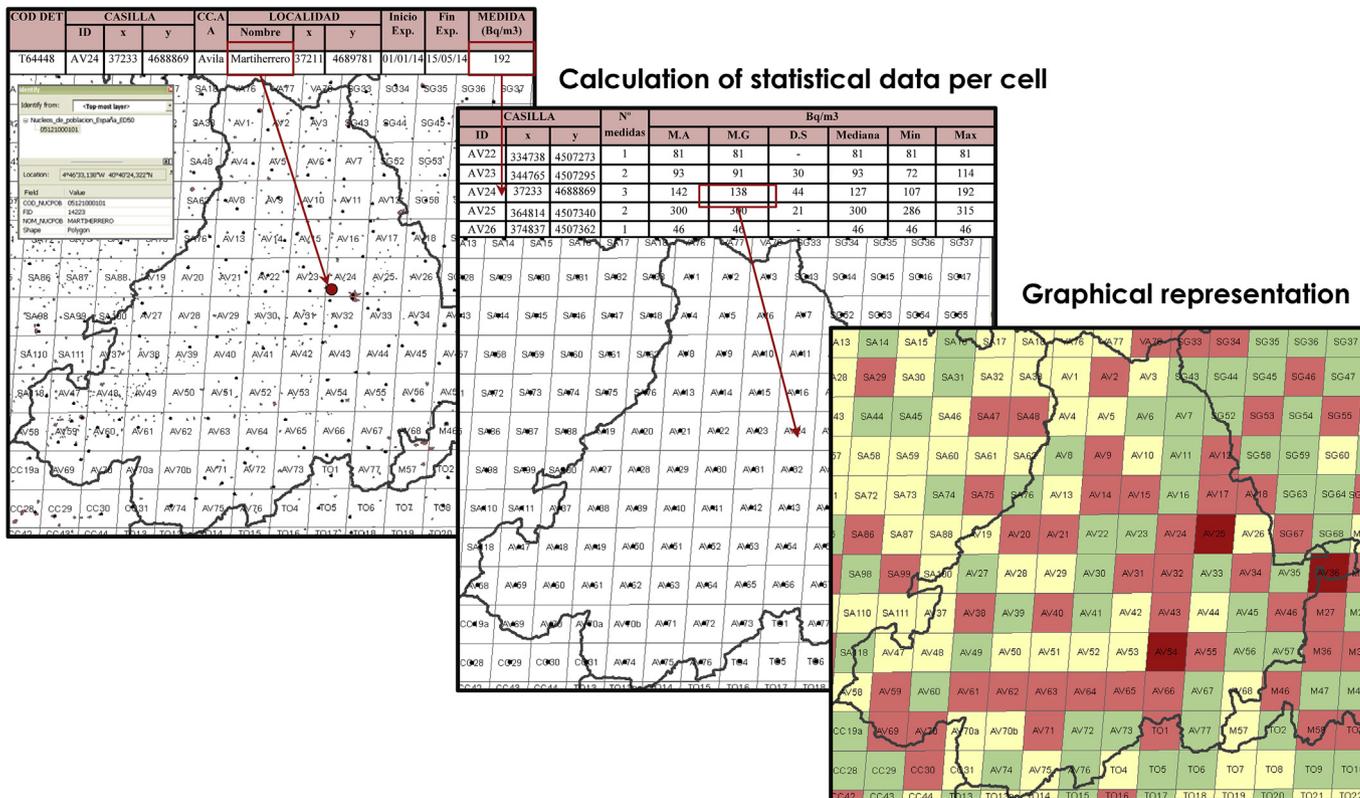


Fig. 4. Process used to incorporate the results of the measurements into the Geographic Information System (GIS).

Distribution of measurements

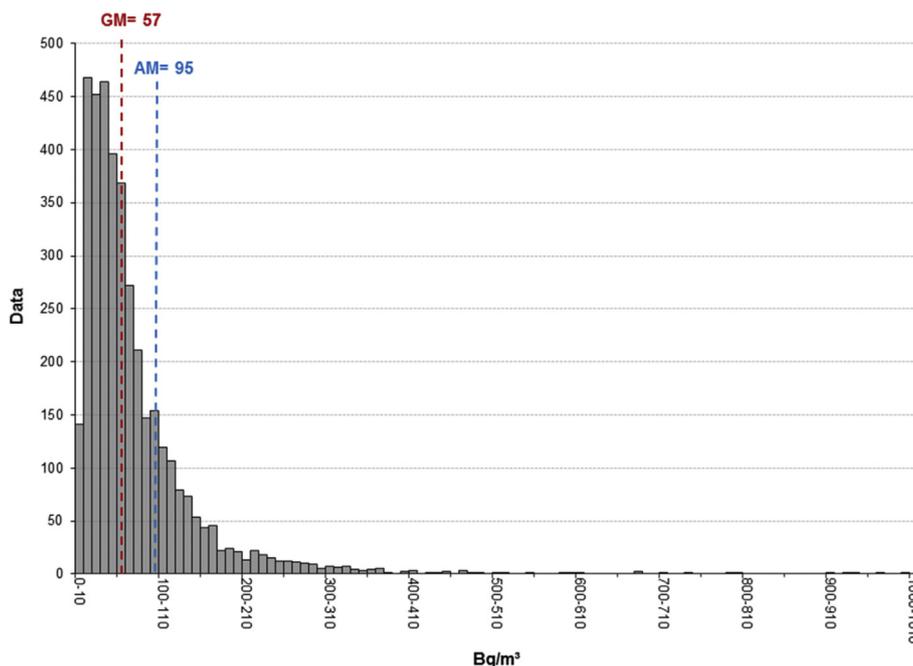


Fig. 5. Log-normal distribution of the current data included in the Spanish indoor radon map. Values of geometric mean (GM) and arithmetic mean (AM) are expressed in Bq/m^3 .

identified the sample cells, houses were selected in different ways. Random locations within the cell were selected and local institutions and government agencies were approached by telephone in most cases.

Each detector was distributed together with installation instructions, a form to be filled up, an information CD, some

explanatory letters and a postage-paid envelope for returns. The form included questions regarding the building's design, building materials and occupants' lifestyle patterns.

The radioactivity laboratory of University of Cantabria (LaRUC) is validated to carry out these types of measurements under the validation scheme designed by PHE (Public Health England, UK)

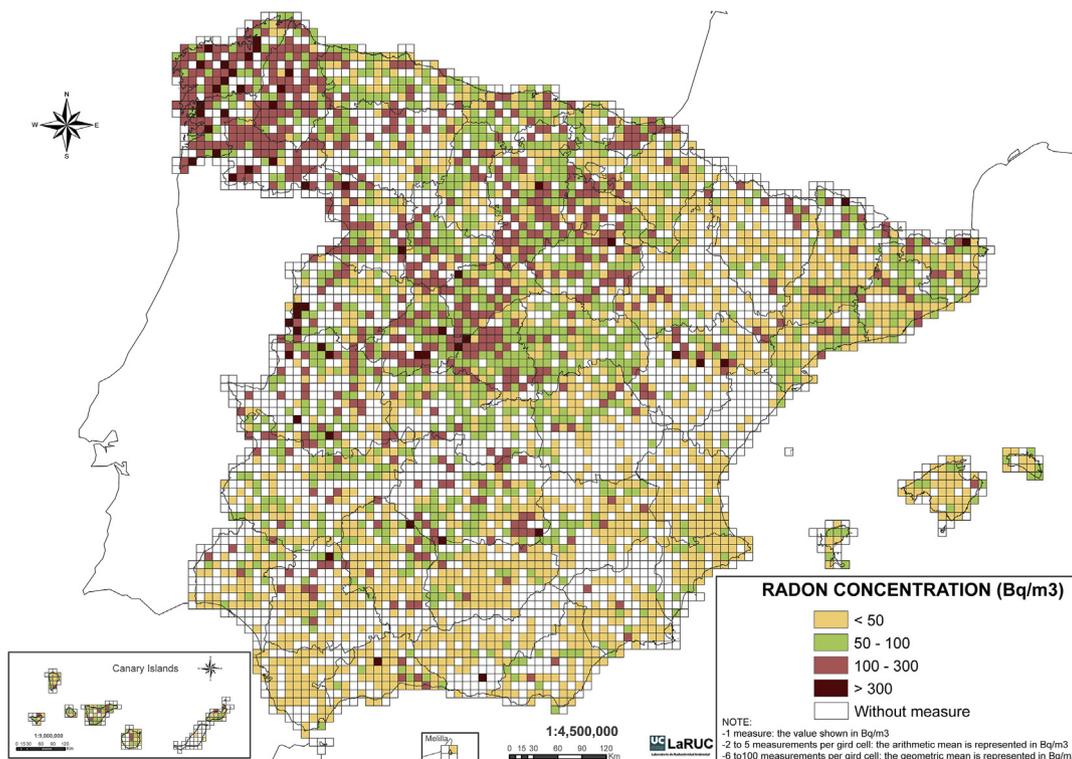


Fig. 6. The Spanish indoor radon map up-to-date. The cells include a total number of 9211 radon determinations.

Table 3
Concentration of radon: Number of cells and data.

Range (Bq/m ³)	Number of cells	Number of data
<50	1606 (29%)	4294 (46%)
50–100	967 (18%)	2922 (32%)
100–300	602 (11%)	1902 (21%)
>300	42 (1%)	93 (1%)

Table 4
Number of measures per cell.

Number of measures (Per cell)	Number of cells (%)
No data	41
1	29
2	12
3	6
4	3
5	2
6 to 100	7

every year. Additionally, the laboratory evaluates the quality of the detector calibration by means of inter-comparison exercises at national and international levels on a regular basis (Gutierrez-Villanueva et al., 2011, 2015).

The measurements took place in ground-level buildings following the protocol of placing the detector in the main room at a height between 1 and 2 m on a shelf or wardrobe separate from the wall, and always away from air currents and heat sources. Once the measurement had been taken, a report was sent to the collaborator informing of the radon level in the buildings and including recommendations in order to reduce the concentration if it were deemed high.

2.4. Data representation

The measurement data were stored using the Geographic Information System's database including: identifying code of each detector; the result of the measurement (in Bq/m³); location where the measurement was made; "x" and "y" coordinates and the cell of the location; and detector exposure time. Thence, the following data were obtained for each cell: number of measurements; geometric mean; arithmetic mean; median; minimum and maximum values and standard deviations. Their graphic representation was achieved by assigning these data to the "x" and "y" coordinates of the centroids of each cell. By doing so we ensured the anonymity of the project's collaborators, as the original data regarding the exact locations of the buildings were not shown on the map (Fig. 4).

3. Results

The Spanish indoor radon map now includes a total number of 9211 measurements distributed across the country. The data conform to a log-normal distribution (Fig. 5), as is commonly found when dealing with radon. The geometric mean and geometric standard deviation are the parameters often used to characterize this type of distribution.

The final result is the map shown in Fig. 6. The cells with 1 measure were represented by the value of radon concentration

obtained. The cells with a number of measurements between 2 and 5 were represented by using the arithmetic mean, and the cells with more than 6 radon measurements used the geometric mean.

The classification was carried out into four categories: <50, 50–100, 100–300 and > 300 Bq/m³, in compliance with recommendations of the World Health Organization (WHO, 2009), the International Commission on Radiological Protection (ICRP, 2009, 2014) and the Nuclear Safety Council (CSN, 2012a) which establish the reference levels in the range 100 and 300 Bq/m³. Among the Spanish data, 12% of the cells were in this range and 1% had geometric means higher than 300 Bq/m³. This means that a large proportion of Spain's surface surpasses the limits recommended by international institutions and the national authority (Table 3).

The majority of the Spanish territory is now covered by at least one measurement (Fig. 8 and Table 4), but there are areas with few measurements (between 1 and 5 measures, 52% of the surface). Incomplete cells (41%) are the result of not being able to carry out a complete analysis in successive sampling campaigns (inside this category 6% of the cells is included without population). Whereas 7% of the surface has had greater than 6 measurements. This density of measures by cell is explained in the own criteria of sampling of the project.

Given that a part of the national territory is covered by only one measure, singular points with high concentration have been found in non prone areas. The significance of the values associated to these cells will increase with a higher number of measurements in future campaigns.

Table 5 shows a summary of the overall statistics of the data. As we can see, in comparison with previous measurement campaigns (CSN, 1998; Martín Matarranz, 2004; García Talavera et al., 2013a; Sainz Fernández et al., 2014), both the national geometric and the arithmetic mean have increased. This is due to the fact that in this second phase of the project the focus has been upon areas expected to have high radon exposure.

An interesting aspect is that areas with a high radon exposure were selected in accordance with lithostratigraphic criterion instead of lithologic or geologic criteria. The first criterion takes into account the soil permeability. Lithological maps show the most representative lithologic associations and geological maps show chronolithostratigraphic units (IGME, 2014b). The use of the lithostratigraphic map is justified by the fact that it incorporates the lithologic permeability factor, which is of great interest when analysing the behaviour of radon. Additionally, it represents units in a smaller scale and, consequently, in greater detail than the geologic or lithologic maps used up until now.

Previous studies have used lithostratigraphy to identify high radon areas in Spain (García Talavera et al., 2013b). To test whether this criterion is valid or not, we investigated if it is possible to identify cells with radon concentrations higher than 300 Bq/m³ and in the range 100–300 Bq/m³.

In order to verify the validity of lithostratigraphic criterion, 40 units identified as radon prone areas were selected and compared with available measurements in those areas. By overlapping the 42 cells over 300 Bq/m³ with the above mentioned units, it was found that 88% of these cells were included within these areas. In the same way, the comparison of the 602 cells with a range of between 100 and 300 Bq/m³, led to a 70% of agreement (see Fig. 7 and Table 6).

Table 5
Descriptive statistics of data used to produce the Spanish indoor radon map up-to-date (Bq/m³).

Number of measurements	Arithmetic mean	Arithmetic standard deviation	Geometric mean	Geometric standard deviation	Median	Range
9211	95.0	270	56.6	2.6	54	10–15,400

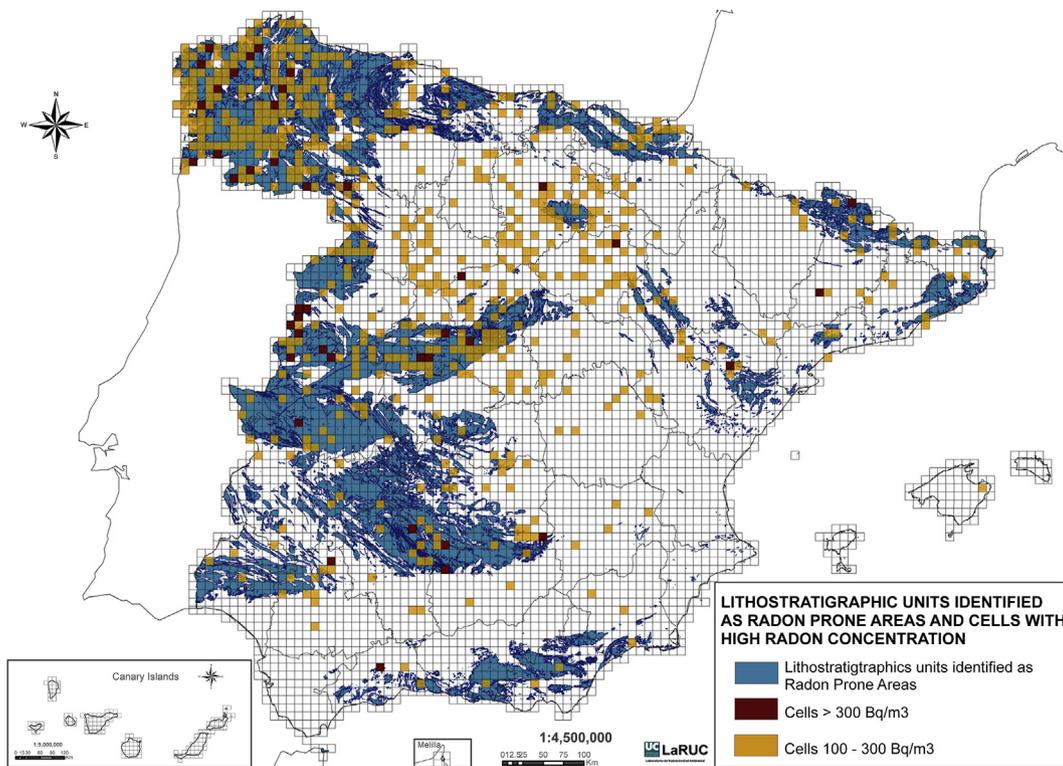


Fig. 7. Lithostratigraphic units identified as radon prone areas and cells with high radon concentration.

The reason to find cells out of the mentioned comparison might be due to the different factors involved with high radon concentrations indoors (building materials, geological faults, ventilation rates, etc.). So it is necessary to perform extra

measurements in those cells located in radon prone areas to validate the preliminary results. This would confirm the importance of lithostratigraphy to identify areas with high radon levels.

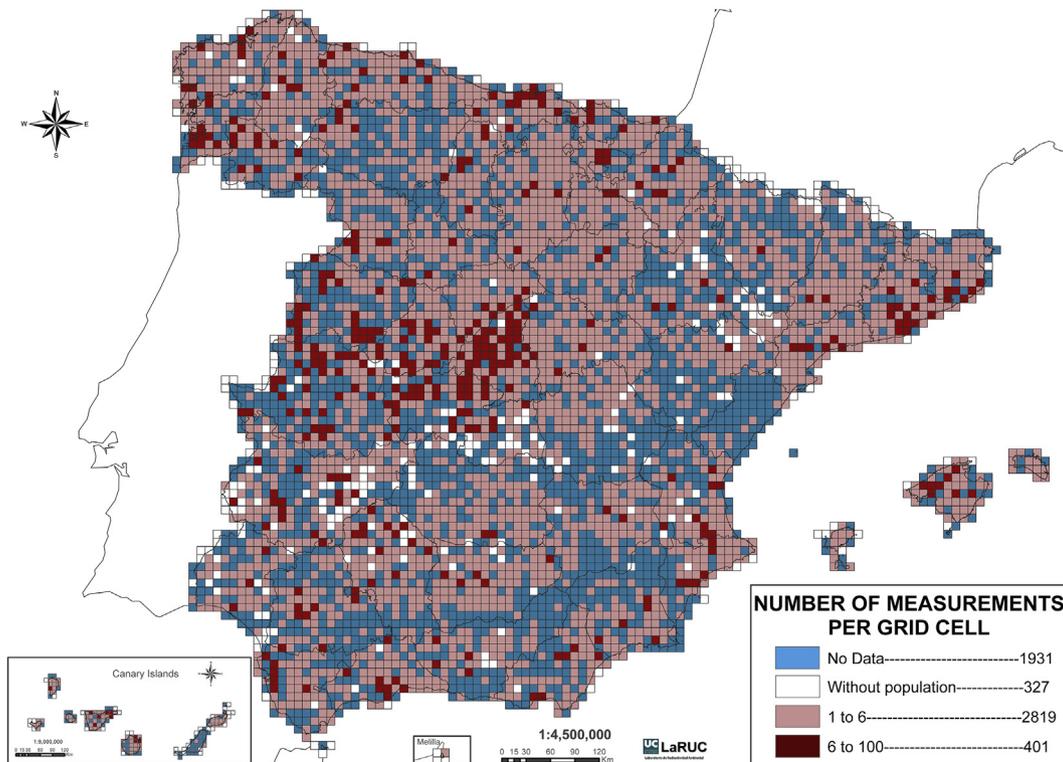


Fig. 8. Graphical view of the situation of the cells in terms of the number of data included in each cell.

