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

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A B S T R A C T

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; Tollesen 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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boundaries provided by the National Geographic Institute (IGN, 2014). Thus we generated a total number of 5478 cells of NW (433193,87; 4834808,73) SE (692906,43; 3320469,29) PE (98454,23; 2988008,14). These coordinates were converted to longitude-latitude ED50 (decimal degrees) to generate a 10 x 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 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 Acimual Equal Area) suggested by the Joint Research Centre of the European Commission (Dubois et al., 2010; Tollersen 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 x 10 km² grid: Peninsular Spain NW (−1500000; −3010000); SE (−3510000; −13500000) Canary Islands NW (−2700000; −19000000); SE (−21500000; −16500000).

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 x 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. 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 x 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 in coexistence with the ETRS89 in Spain until 2015. The parameters of this system are defined in the ArcGIS.

To decide the number of measurements per cell, it was essential to apply the requirements of European legislation on the radon issue at all administrative levels: national, regional and local.

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, Suarez Mahou et al., 2000; Quindos et al., 2004; Quindos et al., 2008) and lithostatigraphic 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 x 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 x 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 nCy/h [4 µR/h]. 2 additional measurements were added for each location that exceeded 35 nCy/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 nCy/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 lithostatigraphical definition.

<table>
<thead>
<tr>
<th>Lithostratigraphic unit code (IGME)</th>
<th>Lithostratigraphy definition</th>
</tr>
</thead>
<tbody>
<tr>
<td>130</td>
<td>Limestones, calcischists and whiteboards</td>
</tr>
<tr>
<td>131</td>
<td>Mica schists, gneiss, phylites, quartzites and plasters</td>
</tr>
<tr>
<td>132</td>
<td>Mica schists, quartz and gneissies</td>
</tr>
<tr>
<td>79</td>
<td>Slates, sandstone and quartzite. Series of los Cabos</td>
</tr>
<tr>
<td>85</td>
<td>Slates, sandstones and microconglomerates. Slates from Lanza</td>
</tr>
<tr>
<td>86</td>
<td>Quartzites, shales and rocks volcanoclastic and volcanised. Quartzite from Barrios and FM Oville</td>
</tr>
<tr>
<td>91</td>
<td>Slates and sandstones. Huergas slates</td>
</tr>
<tr>
<td>104</td>
<td>Quartzites, slates, sandstones, shales, limestones and dolomites. Paleozoic Iberian Aragon</td>
</tr>
<tr>
<td>117</td>
<td>Quartzites and slates</td>
</tr>
<tr>
<td>369</td>
<td>Shales with interbedded carbonate and gyspsum</td>
</tr>
<tr>
<td>7</td>
<td>Plutonic basic Hercynian rocks (gabbros, dioritas, tonalite, ultramafic rocks)</td>
</tr>
<tr>
<td>2</td>
<td>Acid rocks metamorphic (Otogneas, migmatitas). Gneiss, metarhyolitias (Olo Sapo)</td>
</tr>
<tr>
<td>127</td>
<td>Phyllites, schists, quartzites, limestones, slates and conereal (metamorphic)</td>
</tr>
<tr>
<td>152</td>
<td>Sandstones, sands, sandy limestones, marls, clay and loamy</td>
</tr>
<tr>
<td>153</td>
<td>Sandstones, shales and marls</td>
</tr>
<tr>
<td>173</td>
<td>Limestone reef, rudistas, bioclastic limestones, dolomites and marls</td>
</tr>
<tr>
<td>174</td>
<td>Marls, limestone, clay and dolomites</td>
</tr>
</tbody>
</table>
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 μR/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 × 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

### 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.

<table>
<thead>
<tr>
<th>Region</th>
<th>Lithostratigraphic unit code of interest (IGME)</th>
<th>Measures to take</th>
<th>Criterion</th>
</tr>
</thead>
<tbody>
<tr>
<td>Andalucía</td>
<td>130-131-133</td>
<td>40</td>
<td>In at least 6 different municipalities (covering 3 units) with a minimum of 6 measures per municipality</td>
</tr>
<tr>
<td>Asturias</td>
<td>79-85-86-91</td>
<td>50</td>
<td>In at least 8 different municipalities (covering 4 units) with a minimum of 6 measures per municipality</td>
</tr>
<tr>
<td>Aragón</td>
<td>104-117-369</td>
<td>40</td>
<td>In at least 6 different municipalities (covering 3 units) with a minimum of 6 measures per municipality</td>
</tr>
<tr>
<td>Extremadura</td>
<td>7</td>
<td>30</td>
<td>In at least 4 different municipalities with a minimum of 6 measures per municipality</td>
</tr>
<tr>
<td>Cataluña</td>
<td>2-7-117-127</td>
<td>60</td>
<td>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 per municipality</td>
</tr>
<tr>
<td>País Vasco</td>
<td>152-153-173-174</td>
<td>50</td>
<td>In at least 8 different municipalities (covering 4 units) with a minimum of 6 measures per municipality</td>
</tr>
</tbody>
</table>

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

Calculation of statistical data per cell

Graphical representation

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

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**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$^3$.

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

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every year. Additionally, the laboratory evaluates the quality of
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 measure-
ment 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 In-
formation 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. Hence, the following
data were obtained for each cell: number of measurements; geo-
metric 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
measurement were represented by the value of radon concentra-
tion 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 recom-
endations of the World Health Organization (WHO, 2009), the
International Commission on Radiological Protection (ICRP, 2009,
2014) and the Nuclear Safety Council (CSN, 2012a) which estab-
lish 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 den-
sity 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 criteria 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 3
Concentration of radon: Number of cells and data.

<table>
<thead>
<tr>
<th>Range (Bq/m³)</th>
<th>Number of cells</th>
<th>Number of data</th>
</tr>
</thead>
<tbody>
<tr>
<td>&lt;50</td>
<td>1606 (29%)</td>
<td>4294 (46%)</td>
</tr>
<tr>
<td>50–100</td>
<td>967 (18%)</td>
<td>2922 (32%)</td>
</tr>
<tr>
<td>100–300</td>
<td>602 (11%)</td>
<td>1902 (21%)</td>
</tr>
<tr>
<td>&gt;300</td>
<td>42 (1%)</td>
<td>93 (1%)</td>
</tr>
</tbody>
</table>

Table 4
Number of measures per cell.

<table>
<thead>
<tr>
<th>Number of measures (Per cell)</th>
<th>Number of cells (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>No data</td>
<td>41</td>
</tr>
<tr>
<td>1</td>
<td>29</td>
</tr>
<tr>
<td>2</td>
<td>12</td>
</tr>
<tr>
<td>3</td>
<td>6</td>
</tr>
<tr>
<td>4</td>
<td>3</td>
</tr>
<tr>
<td>5</td>
<td>2</td>
</tr>
<tr>
<td>6 to 100</td>
<td>7</td>
</tr>
</tbody>
</table>

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

<table>
<thead>
<tr>
<th>Number of measurements</th>
<th>Arithmetic mean</th>
<th>Arithmetic standard deviation</th>
<th>Geometric mean</th>
<th>Geometric standard deviation</th>
<th>Median</th>
<th>Range</th>
</tr>
</thead>
<tbody>
<tr>
<td>9211</td>
<td>95.0</td>
<td>270</td>
<td>56.6</td>
<td>2.6</td>
<td>54</td>
<td>10–15,400</td>
</tr>
</tbody>
</table>
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. 7. Lithostratigraphic units identified as radon prone areas and cells with high radon concentration.

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

We have used different approaches to find areas with high radon concentrations: lithostratigraphy, lithology and geology. We conclude that the lithostratigraphy is most useful since it includes the soil permeability as a parameter, which is one of the keys to explain the radon transport in the soil. However, our study revealed that some of the high radon zones cannot be related to lithostratigraphical units. Therefore we need to increase the effort on gathering more radon data in order to generate a map that shows homogeneous geological units that may be identified as potential radon risk areas. The lithostratigraphical classification used in this paper at 1:200,000 is sufficient to a national scale but questionable when we want to go into more local scales.

Despite the fact that the number of measurements in Spain has increased with the latest sampling campaigns, we have realised that there are areas where the number of measurements remains low. Future campaigns should be focus on those areas with not enough radon measurements, as well as in those cells where high radon values have been found without correlation with lithostratigraphical classification.

We are going to develop probabilistic models of indoor radon presence to identify high radon risk areas on the basis of geologically similar units where the amount of measurements is lower. Also, in the future this model will enable us to verify the correlation between the different geographic variables in order to identify those which bring about the presence of higher concentrations to a greater extent.

Acknowledgements

Authors express their gratitude to the Spanish Nuclear Safety Council that made possible the national measurement campaigns through different agreements. The authors would also like to thank the staff at the LaRUC laboratory for their invaluable help and cooperation during this study.

Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.jenvrad.2016.07.007.

Table 6: Comparison on the effectiveness of different approaches to identify high radon risk areas.

<table>
<thead>
<tr>
<th>Lithostratigraphy scale 1:200,000</th>
<th>Lithology scale 1:1,000,000</th>
<th>Geology scale 1:1,000,000</th>
</tr>
</thead>
<tbody>
<tr>
<td>All units</td>
<td>329</td>
<td>26</td>
</tr>
<tr>
<td>Units within radon prone areas</td>
<td>40</td>
<td>20</td>
</tr>
<tr>
<td>Number of cell inside radon prone areas</td>
<td>&gt;300 Bq/m³ 37 (88%) and 100–300 Bq/m³ 421 (70%)</td>
<td></td>
</tr>
</tbody>
</table>

References


Web references


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