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Radon levels in groundwaters and natural radioactivity in soils of the volcanic region of La Garrotxa, Spain

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ABSTRACT

Groundwater radon level and soil radionuclide concentration have been measured in the volcanic region of La Garrotxa (Catalonia, Spain) to further research on the origin and dynamics of high radon levels over volcanic materials found in this region. Water samples from different aquifers have been collected from wells and springs and the water radon levels obtained have been lower than 30 Bq l^{-1}. Soil samples have been collected from different geological formations (volcanic and non-volcanic), being Quaternary sedimentary deposits those that have presented the highest mean values of ^{40}K, ^{226}Ra and ^{232}Th concentrations (448 ± 70 Bq kg^{-1}, 35 ± 5 Bq kg^{-1} and 38 ± 5 Bq kg^{-1}, respectively). Additionally, indoor/outdoor terrestrial radiation absorbed dose rate in air have been measured to better characterize the region from the radiological point of view. Terrestrial radiation absorbed dose rates measurement points have been chosen on the basis of geological and demographical considerations and the results obtained, from 27 to 91 nGy h^{-1}, show a clear relation with geological formation materials. The highest terrestrial gamma absorbed dose rate is observed over Quaternary sedimentary deposits as well. All these results help to better understand previous surveys related with indoor and outdoor radon levels and to reinforce the hypotheses of a radon transport through the fissure network.

1. Introduction

Radon is a naturally occurring radioactive noble gas, the most significant isotope of which is ^{222}Rn with a half life of 3.82 days. This isotope is a member of the uranium-238 decay series and its presence in the environment is associated mainly with the trace amounts of its immediate parent, ^{226}Ra, in rocks and soil. Being radon an inert gas, it can move rather freely through porous media such as soil or fragmented rock, enter into dwellings and accumulate indoors. The inhalation of radon progeny is the largest single source of radiation exposure to population, being the 52% of the total dose due to natural radiation (UNSCEAR, 2000).

Radon is soluble in water and therefore it may be incorporated to groundwater flows. The quantity of radon dissolved depends on different factors as aquifer characteristics, water permanence time in aquifer, material radium content, etc. Water-saturated soil with a porosity of 20% and a radium content of 40 Bq kg^{-1} of dry soil, which is the world-wide average in the earth's crust, causes at equilibrium a radon concentration in groundwater of the order of 50 Bq l^{-1} (UNSCEAR, 2000). The Commission of European Communities (CEC) recommends to measure radon levels of domestic drinking water supplies originating from different types of groundwater sources and wells in different geological areas, to determine the exposure of consumer population. According to this recommendation remedial actions are not necessary when concentrations are below 100 Bq l^{-1} (CEC, 2001). Radon levels of some ground water in Spain have been analysed (Dueñas et al., 1998; Galán López and Martin Sánchez, 2008; Ródenas et al., 2008; González-Díez et al., 2009) obtaining concentrations up to 1868 Bq l^{-1}. The global distribution of values obtained reveals that the level of concentration is closely linked with the specific geological characteristics of the formations where water flows, with high values appearing in areas where there are fractures and the source of the spring is at greater depth.

The gamma emitter members of the three natural decay series, together with primordial gamma emitters, as ^{40}K, account for, on average, 20% of the natural radiation dose. The main source of gamma exposure outdoors is the soil, while indoors building materials are the main contributors. Terrestrial gamma radiation doses received indoors are usually higher than those received outdoors,

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mainly due to the change in the geometry of gamma sources (García-Talavera et al., 2007). Radionuclide distribution in geosphere mainly depends on geology. For instance plutonic rocks, like granite, present higher contents of ^{40}K, ^{238}U and ^{232}Th than other rocks like sedimentary rocks and basalts (Taboada et al., 2006). Radionuclide distribution can also change with time due to some natural and artificial processes that could diminish or concentrate radionuclides in specific places. In some volcanic regions, like Tenerife island at the Canaries archipelago (Fernández de Aldecoa, 2000) and Milos island in the Aegean Sea (Florou and Kritidis, 1991) higher gamma radiation levels and radionuclide content than the world mean values given by UNSCEAR (2000) have been found. In Spain, Suarez et al. (2000) elaborated a nation-wide 1:1.000.000 map of outdoor gamma radiation levels (MARNA), with a higher resolution (up to 1:50.000) in gamma-prone areas. Gamma dose rate levels were obtained from both direct measurements and geological data, with an average value of 76 nGy h^{-1}. Radiation levels were not measured with the same spatial resolution everywhere. Additionally, based on MARNA map, a map of potential indoor radon concentrations was elaborated (Quindós Poncela et al., 2004). This map reflects the trend of indoor radon levels, but no quantitative indoor radon predictions can be calculated from it (Quindós et al., 2008; García-Talavera et al., 2013).

The region studied in this work is a volcanic-origin area within La Garrotxa county, located in the North-East of Catalonia, Spain. It covers about 150 km^2 and has Olot as its main city. The main part of La Garrotxa population lives over volcanic materials. In a preliminary indoor radon survey in Olot we obtained higher radon levels in buildings over volcanic materials than on non-volcanic (Baixeras et al., 2005). In a bigger indoor radon survey in Olot and in four villages close to the nearby Amer fault we confirmed these results and observed an influence of the soil type on radon seasonal variations (Moreno et al., 2008). Radon levels obtained in these previous surveys, together with the geological structure of La Garrotxa region and the presence of the nearby Amer fault, which still might be active, suggested the possibility that radon could come from mantle degassing. We considered that this fault could be connected to the fractured system of La Garrotxa and that radon transport could take place basically through the fissure network. Other alternatives were considered as well; radon could come mainly from the soil underneath and/or from deeper substrates with higher radium contents. During last years, in order to continue exploring these hypotheses, we had been studying different radon sources and natural processes. Soil radon levels in the vicinity of the Amer fault were measured, finding very important seasonal variations (Font et al., 2008). We characterized blowholes, rare geologic features in which air is blown through small holes at the soil surface, identifying three different types and finding significant radon level temporal variations correlated with the soil-outdoor temperature differences (Moreno et al., 2009). The next steps have been to analyse groundwater radon levels and soil radium content as radon sources and to better characterize the region from the radiological point of view (Moreno Baltà, 2012). In this paper we present the results of these last steps. The goals have been: i) to measure radon concentration in ground water related with aquifers in contact with volcanic and non-volcanic materials, ii) to determine radionuclide content of soil materials, iii) to measure indoor and outdoor gamma radiation levels over different types of soil and iv) to explore the relation of radon levels and gamma radiation levels with radionuclide content of soil materials.

2. Geological framework

The geological structure of the volcanic region of La Garrotxa consists basically on quaternary volcanic and non-volcanic formations over a tertiary substratum (Fig. 1). This region is located in a framework of a low volcanicity rift model (Martí et al., 2001) affected by a system of longitudinal NW–SE orientated normal faults that developed, since Neogene until today, migrating to southeast direction (Saula et al., 1996). Among these faults, the Amer fault presents evidences of recent activity (Fleta et al., 2001). There are more than 30 volcanoes distributed across the fault network. In general, they are cinder cones surrounded by pyroclastic deposits (Qv). Most of the valleys are covered with lava flows (Qbc) and some of them present interleaved layers of lake deposits (Qlb) consequence of the formation of natural dams by lava flows. The tertiary (Eocene) substratum is composed of sedimentary rocks that can be classified into: (i) polymictic and arkosic sandstones (Eg), (ii) marls and grey lutites with sandstones levels (Em), (iii) polymictic conglomerates and red sandstones with lutites (Ecg) and (iv) alternation of sandstones with marls or lutites (Egm). During Quaternary the fluvial system and gravity joint action also produced deposition of alluvial and colluvial sediments (Qacb) from tertiary substratum (ICC et al., 2007).

According to the aquifer lithology of the region, aquifers can be classified into three main categories: (i) aquifers in alluvial and colluvial quaternary formations (*A*), (ii) aquifers in alluvial and colluvial quaternary formations interleaved with volcanic materials, called alluvial-volcanic aquifers (*AV*), and (iii) aquifers in Eocene formations (*E*). *A* aquifers are formed with materials (sands and gravels) with intergranular medium-low porosity and medium–high permeability. *AV* aquifers, due to the different characteristics of volcanic materials, can present intergranular porosity in pyroclastic materials as well as fissuring porosity in lava flows; therefore permeabilities can present high variability. *E* aquifers, located under the quaternary materials and due to having a more complex geological structure, are the less known and exploited. Some of these formations, constituted by limestone and calcareous sandstone, can present fissuring porosity and kartstification. According to the Parc Natural de la Zona Volcànica de La Garrotxa (PNZVG) water quality control, water fluxes can be classified as local or regional using the nitrate concentration as indicator. Local fluxes are more superficial and collect water from a nearby area, so the water presents higher nitrate concentrations. On the other hand, regional fluxes are interpreted as being deeper, collecting less contaminated water from longer distances before emerging as a spring (Bach, 2005).

3. Radon concentration in groundwaters

3.1. Materials and methods

A total of 53 points has been selected to measure radon concentrations (42 private and public wells and 11 springs). These points correspond to different aquifer systems (Fig. 1). Two samplings have been carried out in 2007. In the first one [May 7 to September 13] a total of 23 wells and 9 springs of alluvial-volcanic aquifers were measured. In the second one [October 22 to November 24] the same wells and springs were measured but two (12 and 25); one because of being dry and the second because became inaccessible. In addition, we expanded the sampling with 21 alluvial aquifer points. Wherever the radon levels obtained in both samplings differed by more than 50% and at least one was higher than the whole survey average, an additional measurement was taken one month later. In springs we took water directly from the source, whereas in wells we had to use different water pumps for sampling from 1 m under water level to avoid superficial and stagnant waters. 250 ml glass bottles were used to avoid radon leakage during transport to our laboratory. Using a flow cell and a