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RADON MEASUREMENTS IN GROUNDWATER MINES IN LA PALMA AND EL HIERRO, CANARY ISLANDS (SPAIN)

In the Canary Islands, groundwater is the main source of drinking water. Groundwater mines have been the system used by the engineers of the archipelago to collect water from the ground. The Canary Islands are volcanic with soils characterized by being rich in uranium, the disintegration of which gives rise to radon gas. In this study, radon gas levels in the mines on two islands of the archipelago have been measured to study exposure to this gas in the galleries. Results show values much higher than the European regulatory limit concentrations.

Keywords: radon gas, radiation, groundwater, mines, Canary Islands

1. Introduction

Radon is a radioactive element that has different forms in nature (radioactive isotopes) and one of them, the most stable and abundant, is Rn-222, hereinafter referred to as radon. The chain of disintegration of the atomic elements involved in the generation of radon (Rn-222) begins with uranium (U-238), passing through thorium, radium and finally radon [1]. Since radon gas is heavier than air, under certain conditions, it tends to accumulate in underground structures

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(tunnels, basements, mines etc.) and can reach levels that are harmful to health. Medical and environmental studies indicate that radon-associated radiation is the second cause of lung cancer after tobacco and the first for non-smokers [2].

The emission of radiation by atomic elements is associated with radioactive isotopes. The structure of atoms can be simplified into a nucleus formed by protons and neutrons, with a cloud of electrons surrounding it. An atomic element has a fixed number of protons in the nucleus, but the number of neutrons can vary, giving rise to different isotopes of that element [3]. The element maintains the same number of protons permanently, but isotope may vary their number of neutrons over time, becoming unstable components; these are known as radioactive isotopes or radionuclides. Thus, radioactivity is a property by which radioactive isotopes disintegrate into new atoms and emit either particle radiation (alpha, beta, neutrons, protons) or gamma radiation. All this radiation is known as ionising radiation, since it interacts with the surrounding matter and can dissociate it to form ions.

In the Canary Islands, most of the hydrological resources come from groundwater, with 70% of water being obtained from this source [4]. Mines are drilled into the natural terrain, mainly by private initiative, seeking to find the aquifer and extract water resources for the population. These underground works are covered by volcanic soils and with high levels of humidity, so high concentrations of radon gas can be expected in them. Water also represents a potential source of radon, since once radon dissolves in it, it can be transported to the area where the water is extracted and consumed [5]. The areas that present the greatest danger from waterborne radon are, as might be expected, those where the rocks or soil through which the water flows have high levels of uranium and other radon parents.

The European Union, through the Council Directive [6] establishes basic safety standards for protection against the dangers arising from exposure to ionising radiation. The Directive specifies that when radon enters from the ground into closed work areas, it should be considered an existing exposure situation, since the presence of radon is largely independent of human activities carried out within the workplace. Such exposures may be significant in certain areas or in specific types of workplaces to be determined by the Member States and, if the national reference level is exceeded, appropriate measures to reduce radon and exposure should be taken. If the levels continue to exceed the national reference level, such human activities within the workplace should not be considered practical. However, Member States should ensure that such workplaces are notified and that, where worker exposure may exceed a corresponding time-integrated radon exposure value, the workplaces are managed as a planned exposure situation, dose limits are applied, and operational protection requirements are determined. Member States will take radon measurements to identify the types of workplaces and buildings with public access, e.g. schools, underground workplaces or those in certain areas, where measurements are required on the basis of a risk assessment, taking into account, for example, the hours of occupation. Regarding the limits of radon exposure, it is not advisable that the value of 300 Bq/m^3 be exceeded, according to European Directive 2013/59 EURATOM.

2. Materials and methods

This study was undertaken on the islands of La Palma and El Hierro, in the Canary Islands, Spain, which are the youngest islands in the archipelago. On both islands most lavas are basaltic, and none are prior to the Quaternary. However, the last aerial eruptions of the archipelago took place in 1949 and 1971 on the island of La Palma, and they occurred in the North-South Dorsal formed by the Recent Basaltic Cycle, which comprises several historical eruptions [7].

According to the Security Guide 11.4 [8] developed by the Spanish Nuclear Security Council (CSN), studies on the radiological risks linked to radon must estimate the annual exposure for workers and, if applicable, for the public as well. Measurements should be made at locations that are representative of the actual or reasonably foreseeable occupation of workers (Fig. 1). For this reason, measurements need not be taken in areas such as corridors, toilets, etc., that are unoccupied or infrequently occupied. An under-occupied area can be considered as one where a person is unlikely to spend more than 100 hours per year of his or her working time (approximately two hours per week).

Fig. 1. Passive detector placed inside the gallery, where the workers spend their working hours

Thus, the results must be based on measurements with passive detectors which are exposed for a minimum of three months. Passive detectors are those recommended by the World Health Organization [2] and ISO 11665-4 [9] because long-term radon measurements are preferred for assessing the annual average radon concentration within a house or a workplace. High temporal variation of indoor radon makes short-term measurements unreliable for most applications. For this reason, nuclear traces passive devices were chosen, specifically Radtrak[2], alpha-track detectors (ATD) from Radonova, in order to meet all the CSN requirements. These detectors consist of a radiation-sensitive film located within a capsule made of a special antistatic plastic, which enables radon gas to enter by diffusion. The specifications of the detectors used are as follows: measuring range (from 15 Bq/m^3 up to 25000 Bq/m^3) and measuring period (from 2 to 6 months).

In each of the studied areas (one area was in the middle of the gallery and the other one at the end of it) it was placed a passive detector, which covered a period of three months each one. That is, in each gallery there were placed two passive detectors per period, in each one of the mentioned zones. Therefore, the length at which the passive detectors were placed at the front of the gallery corresponds to the total number of meters excavated. Thus, the length in the middle of the gallery, corresponds to half of the meters excavated. The measurement periods on the island of El Hierro cover August 2018 to February 2019. On the island of La Palma it was measured from February to September 2019.

Once the detectors were removed after the measurement periods, they were sent to an accredited laboratory in Sweden for analysis. In order to ensure the readings taken by the passive detectors were within the correct range, we took extra measurements in some of the mines in La Palma with active detectors (Radon Scout from SARAD Company) (Fig. 2). The active equipment can be operated simply and offers protection against unauthorised manipulation, and the readings may be transferred to a computer using a USB connection. In addition, the active device is so sensitive that it can accurately detect variations in the activity of concentrations, even though when radon levels are low. Therefore, we went to the groundwater mines and placed the active detectors at points around the mid- length of the galleries, leaving them for one hour.

Fig. 2. Active detector taking measurements in a gallery in La Palma

3. Results and discussion

Table 1 reports radon concentrations values measured during 2018-2019 with passive detectors, in water galleries in La Palma and El Hierro.

Table 2 collates all the measurements with the active detector, in the period of one hour on the island of La Palma, in order to compare them with the passive detector's results. The sensitivity of the active detector is 3/7 calculations/minute: 200 Bq/m^3 with 10% statistical error (1σ) in 1h sample interval; 10 Bq/m^3 with 25% statistical error (1σ) in 4h sample interval. In most cases,

TABLE 1

Results of radon gas measurement in groundwater mines in La Palma and El Hierro.
Location: deepest area and mid-length

Radon gas concentrations at the front of the gallery							Radon gas concentrations in the middle of the gallery line						
Island	Municipality	Location: Length (m)	Minimum Bq/m^3	Average Bq/m^3	Maximum Bq/m^3	Uncertainties Bq/m^3	Island	Municipality	Location: Length (m)	Minimum Bq/m^3	Average Bq/m^3	Maximum Bq/m^3	Uncertainties Bq/m^3
La Palma 1	Garafía	3800,0	1570,0	1920,0	2270,0	350,0	La Palma 1	Garafía	1900,0	2110,0	2580,0	3050,0	470,0
La Palma 2	Puntallana	2000,0	5810,0	7090,0	8370,0	1280,0	La Palma 2	Puntallana	1000,0	3040,0	3710,0	4380,0	670,0
La Palma 3	Los Llanos	3278,0	3660,0	4470,0	5280,0	810,0	La Palma 3	Los Llanos	2700,0	3960,0	4840,0	5720,0	880,0
La Palma 4	Breña Alta	2185,0	5010,0	6120,0	7230,0	1110,0	La Palma 4	Breña Alta	1000,0	4280,0	5230,0	6180,0	950,0
La Palma 5	Fuencaliente	170	890	1.050	1210	160	La Palma 5	Fuencaliente	145	430	500	570	70
Island	Municipality	Location: Length (m)	Minimum Bq/m^3	Average Bq/m^3	Maximum Bq/m^3	Uncertainties Bq/m^3							
El Hierro 6	Valverde	300	41	71	101	30							
El Hierro 7	Valverde	80	460	530	600	70							
El Hierro 8	La Frontera	Pumping Area	2440,0	2980,0	3520,0	540,0							
El Hierro 9	Valverde	20	410	480	550	70							

active measurements resulted in lower values than those obtained with passive measurements. This may be because the passive measurements collect periods of three months, where seasonal variations are suffered that may be the cause of the high results obtained.

TABLE 2

Results of radon gas measurement with an active detector in groundwater mines in La Palma

Code	Island	Municipality	Length (m)	Average Bq/m^3
1	La Palma 1	Garafía	1.900	1.356
2	La Palma 2	Puntallana	1.000	2.171
4	La Palma 4	Breña Alta	1.000	1.425
5	La Palma 5	Fuencaliente	25	2.292

In almost all the galleries studied, the radon limit levels set by the Directive Euratom were exceeded by a wide margin. These radon levels are most probably due to the igneous materials present in the aquifer [10]. Several studies of radon in groundwater show that high radon activity occurs in uranium-bearing granite aquifers, in magmatic and metamorphic bedrock aquifers [11]. Natural radioactivity is partly due to primordial radionuclides contained within the Earth's crust, such as ^{40}K, ^{238}U, ^{232}Th (potassium, uranium and thorium) and the products of their decay series. For instance, a study carried out in the state of Pernambuco in Brazil [12], which analysed the spatial distributions of natural radionuclides, found that the radionuclide concentrations decreased in the following order: igneous > metamorphic > sedimentary, an occurrence that has been verified as well in Italy [13] and Greece [14]. The Canary Islands offer a potential wide variety of naturally occurring radioactive volcanic rocks. Volcanic felsic rocks contain more radionuclides than volcanic mafic rocks. The presence of felsic rocks dominates in La Palma and El Hierro and radon concentrations in groundwater mines are significant from the point of view of worker's health.

The amount of radon present in a mine depends mainly on the concentration of uranium in the rocks that make up the site [15,16], atmospheric conditions and the existence of ventilation. In fact, the only groundwater mine (in El Hierro) whose radon concentration is lower than 100 Bq/m^3, has with two entrances that favour the mine ventilation. Besides, the whole mine length is covered by gunite concrete, which helps to isolate it from the ground. Therefore, in closed spaces without ventilation, high concentrations of radon can accumulate in the trapped air [17]. In active facilities, a forced ventilation system can be installed to help keep radon concentrations below the limits specified by current regulations. However, in disused or abandoned galleries this is option in most cases is not feasible.

Predictably, radon moves more easily through permeable soils, such as gravel and coarse sand, than through clay soils, which are highly porous but not very permeable. In turn, radon dissolved in water moves much more slowly than radon in air [18]. The distance that radon can move before it disintegrates in water-saturated rocks and soils is only a few centimetres, while in dry soils it can be up to several metres. For this reason, in areas with relatively normal radon values (between 1000 and 50,000 Bq/m^3) but whose subsoil is very permeable and dry, high surface radon levels can be detected.

4. Conclusions

Although the current legislation establishes a radon concentration limit of 300 Bq/m^3 for workplaces, the average values obtained in groundwater mines of La Palma range between 500 and 5810 Bq/m^3, and in El Hierro between 71 and 2980 Bq/m^3. Therefore, workers' exposures to radon gas should be controlled at all times and minimised as far as possible. To this end, workers must carry an individual dosimeter inside the gallery, and, in addition, constructive and/or administrative solutions must be taken whose cost must be met by the organization [8]. Construction measures to be taken include extraction and/or forced ventilation systems and administrative measures include control of the time employees spend in areas with high radon concentrations.

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