

Article

## Radon in Groundwater of the Northeastern Gran Canaria Aquifer

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**Abstract:**  $^{222}\text{Rn}$  has been detected in 28 groundwater samples from the northeast of Gran Canaria (Canary Islands, Spain) utilizing a closed loop system consisting of an AlphaGUARD monitor that measures radon activity concentration in the air by means of an ionization chamber, and an AquaKIT set that transfers dissolved radon in the water samples to the air within the circuit. Radon concentration in the water samples studied varies between 0.3 and 76.9 Bq/L. Spanish radiological protection regulations limit the concentration of  $^{222}\text{Rn}$  for drinking water to 100 Bq/L, therefore the values obtained for all the analyzed samples are below this threshold. The hydrogeological study reveals a significant correspondence between the radon activity concentration and the material characteristics of the aquifer. For a selected group of samples with high radon concentrations, gross alpha activity has been determined to have values higher than the prescriptive screening level (0.1 Bq/L).

**Keywords:** radon activity; gross alpha; groundwater tracer; volcanic terrain; normative

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## 1. Introduction

Radon, a natural byproduct of the radioactive decay of uranium, radium and thorium, is an alpha-emitting noble gas with a half-life of 3.8 days. Radon gas is soluble in water and consequently the gas may be incorporated into groundwater flows. Radon is extracted from the volcanic deposits in which the aquifer resides, its transport taking place basically through the fissure network in the fractured system or from mantle degassing. The quantity of radon dissolved in groundwater depends on different factors such as the characteristics of the aquifer, water-rock interaction, water residence time within aquifer, material content of radium, *etc.* [1–3].

Measurements of radon contents in groundwater have been performed in connection with geological, hydrogeological and hydrological surveys and health hazard studies. On the one hand, the half-life of radon and its solubility have allowed the use of radon gas as a natural groundwater tracer to identify and quantify groundwater discharge to surface waters [4–6] or to attempt to elucidate the type of rocks through which groundwaters flow [5,7]. On the other hand, the presence of high levels of radon in drinking water constitutes a major health hazard [8–10]. The Commission of European Communities (CEC) recommends the monitoring of radon levels in domestic drinking water supplies originating from different types of groundwater sources and wells in different geological areas, in order to determine consumer population exposure. The limit is fixed at below 100 Bq/L [11].

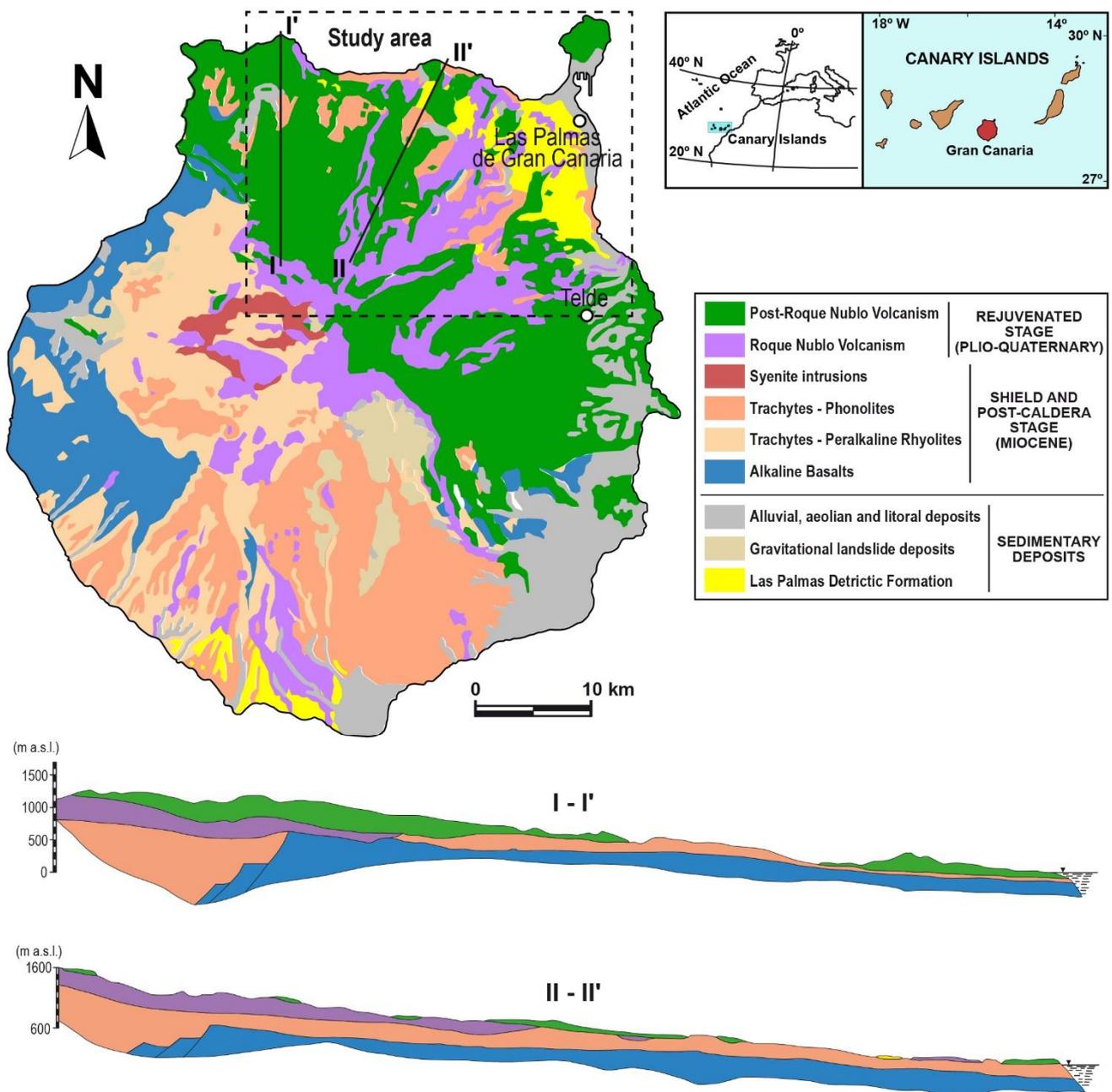
The region studied here is an area of volcanic-origin located in the northeast (NE) of Gran Canaria Island (Canary Islands, Spain). Groundwater in the investigated area plays an important role in guaranteeing water supply for agricultural and domestic purposes, mainly in the higher parts of the island. In coastal areas, desalinated seawater provides for urban needs.

The content of radon in water samples must be determined by reliable methods. Radon is a very mobile gas and it can escape from water with ease during the process of sampling and transportation, hence careful sample preparation is necessary. Several procedures can be found in the literature to perform measurements of radon in groundwater using different techniques such as Lucas-cell, ionization chambers, solid-state detector, or gamma spectrometry [12–16]. The aim of this study was to measure radon concentration in groundwater at most populated zone of Gran Canaria as a starting point for monitoring of the quality of groundwater with respect to radon gas in the whole territory as well as making a first attempt at using this gas as a tracer to establish dynamic knowledge of groundwater flow in Gran Canaria.

## 2. Area of Study

### 2.1. General Description

The Canary Islands, located at the eastern edge of the Central Atlantic Ocean (between 27° N and 30° N and from 19° W to 13° W), close to the north-western continental margin of Africa, comprise seven major volcanic islands (Tenerife, La Palma, La Gomera, El Hierro, Gran Canaria, Lanzarote and Fuerteventura) and six islets (La Graciosa, Alegranza, Montaña Clara, Lobos, Roque del Este and Roque del Oeste). The Canary archipelago extends over approximately 500 km, and the eastern islands are situated about 100 km off the African coast (Figure 1).



**Figure 1.** Location of the study area. Simplified geological map of Gran Canaria (modified from [17]) and geological cross sections of the study area. The sections were made using geological data from wells (modified from [18]).

Gran Canaria Island, located at the center of the Canary archipelago (Figure 1), has a conical morphology and a nearly circular shape with a diameter of 45 km and a maximum elevation of 1950 m above sea level (a.s.l.) at its center. It is shaped by a series of radial gullies that originate in the center of the island and flow into the sea. The population of the island is around 900,000 inhabitants mainly concentrated in the NE area where the most populated cities of the island, Las Palmas de Gran Canaria (400,000 inhabitants) and Telde (100,000 inhabitants), are located.

## 2.2. Geological Setting

The construction of the Canary archipelago is related to the movement of the African plate above an anomalous or pulsating mantle plume [19,20]. The geology of Gran Canaria is conditioned by its origin (Figure 1). The subaerial formation of Gran Canaria occurred in two main phases: (1) a juvenile stage (about 14.5–8.0 Ma), which includes a basaltic shield volcano, a vertical caldera collapse and a post-caldera resurgence characterized by evolved magmas; and (2) a rejuvenated stage (about 5 Ma to present). Both phases are separated by a period of volcanic inactivity that lasted about 3 Ma [20–22].

The geology of the study area includes volcanic and sedimentary materials (Figure 1). On the surface basaltic, basaltic and trachybasaltic lavas of the Roque Nublo and Post-Roque Nublo volcanism, and sediments of Las Palmas Detritic Formation mainly occur. Miocene trachytic and phonolitic lavas crop out, usually in the coastal areas and at the bottom of ravines, where alluvial deposits are also observed [21,23]. The main mineral phases in Basic volcanic rocks consist of diopside, olivine and labradorite, whereas in Salic volcanic rocks andesine, kaersutite, anorthoclase and hauyne are found [23,24]. On the other hand, secondary mineral phases, such as weathering cover or fillings of fractures and vesicles on volcanic rocks, include calcite, dolomite, gypsum and clays (illite, and smectites, usually montmorillonites), and zeolites (analcime, chabazite, phillipsite).

The basic and ultrabasic rocks (peridotites, pyroxenites, gabbros, basalts, *etc.*) do not contain unstable elements in the crystal structure of the mineral and therefore have low radioactivity. The more differentiated rocks (syenites, trachytes, phonolites, carbonatites, *etc.*) contain in its crystal structure some forms of trace elements (La, Ce, Sm, Nd, Sr, Ba, Th, Nb, *etc.*), some of them radioactive, and potassium that is the sources of higher concentrations of radiological activity present in these rocks. In consequence, it is expected that groundwater flowing through acid geological formations will have higher values of  $^{222}\text{Ra}$  concentrations than through basic geological formations.

## 2.3. Hydrogeological and Hydrogeochemical Characteristics

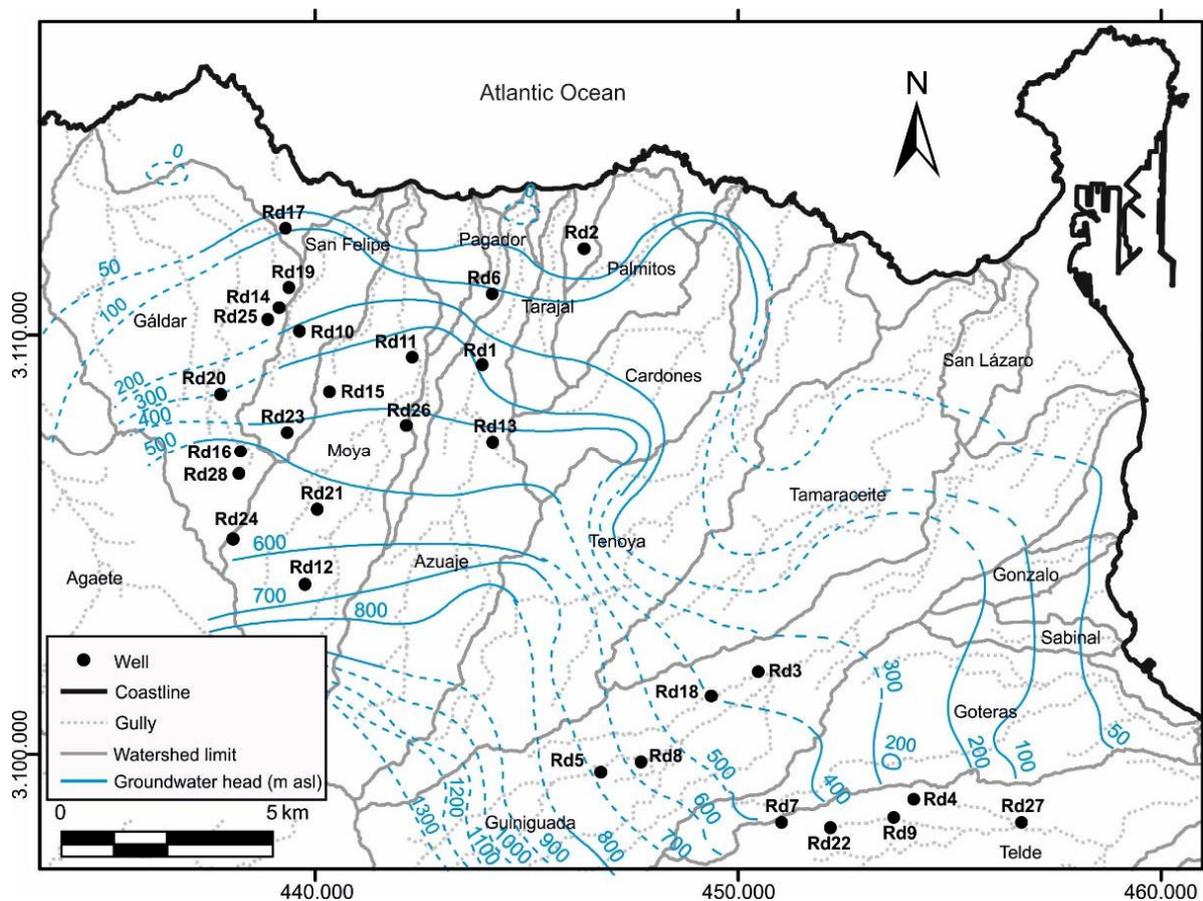
The island aquifer is conceptualized as a single, stratified and heterogeneous water-body, with groundwater flows from the recharge area (at the summits in the central part of the island) towards the coast. There are hydraulic connections between waters occurring in different rocks, which actually form one common hydrogeological system in the island [25,26].

In this framework, groundwater in the study area is part of the northern insular aquifer and flows from the south to the north. In the study area, groundwater is extracted predominantly from the Roque Nublo group rocks and underlying Miocene trachytic and phonolitic lavas. Groundwater exceptionally flows towards the deep gullies (“*barrancos*”) and generally discharges to the coast. Recharge is mainly a result of rainfall recharge, the study area constitutes the main recharge area of the island, and also from irrigation return flows. Agriculture, which is mainly practiced in the coastal areas, is supplied from both surface and groundwater resources, and locally causes important irrigation return flows [26,27], which promote increases in soil and groundwater salinity. Discharge occurs towards the sea and through withdrawals from wells and galleries. Groundwater is mainly of Mg-Ca-HCO<sub>3</sub> and Mg-Ca-Cl type. At the coast, groundwater can be Na-SO<sub>4</sub> due to irrigation return flows [25].

## 2.4. Area of Study

The study area, about 323 km<sup>2</sup>, is bordered by Atlantic Ocean to the north and watersheds to the east and west. The overall average annual rainfall is 375 mm; in the highlands the average rainfall reaches 820 mm/year, while the coastal areas remain dry with an average rainfall of 115 mm/year. Average annual temperature varies from 12 °C in the highlands to 22 °C at the coast, with an average temperature of 18 °C.

A total of 28 groundwater points were selected in the northeast of the island of Gran Canaria corresponding to 26 large-diameter wells, one borehole and one spring. In Figure 2 the selected points are shown.



**Figure 2.** Spatial distribution of the groundwater sampling points and groundwater head contours 2008–2009, modified from [28,29]. The names of the gullies are indicated. Coordinates in UTM WGS84 28N.

## 3. Materials and Methods

### 3.1. Sample Characteristics and Sampling Procedure

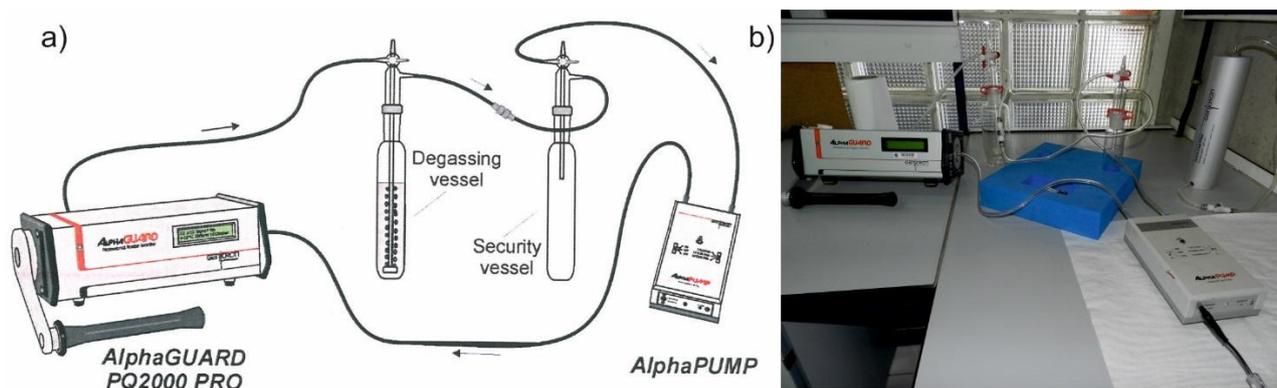
Twenty-eight groundwater samples were taken during the field campaign carried out in the summer of 2014. Sampled groundwaters exhibit temperatures of up to 25 °C and/or high concentrations of dissolved volcanic gases. The depth of the wells varied from 125 to 600 m, passing through several geological structures.

The samples were taken by pumping using submersible pumps, and so the results obtained can be influenced by the large purging volumes and pumping rates that vary from one sampling point to another. The samples were contained in dark glass bottles of 1 L capacity; the procedure to collect the samples was designed to reduce radon losses due to bubbling. At each location, the bottles were filled to the edge with the sampled water and then immediately closed to avoid loss of radon by degassing during transport to the laboratory. Radon levels were determined within 3–6 h after sample collection in order to minimize the influence of radioactive decay.

### 3.2. Equipment Setup

The concentrations of radon in the groundwater samples were measured using the continuous active radon monitor ALPHAGUARD (Model PQ2000PRO) of Genitron-Saphymo, Germany [30]. This detector was selected due to its proven calibration stability and fast response to concentration gradients, which have been confirmed in numerous studies [31]. It utilizes a pulse-counting ionization chamber of 0.65 liter active volume to detect radon, and it is suitable for long-term monitoring of radon gas concentrations from 2 to  $2 \times 10^6$  Bq/m<sup>3</sup>. The measurement procedure is based on the alpha spectrometry of radon and its progeny in the air that enters into the detection volume of the ionization chamber. In addition, the ALPHAGUARD registers the values of the main environmental parameters (temperature, humidity and atmospheric pressure) during the time of measurement.

For the measurement of radon concentration in water, an AquaKIT system [32] was used. This system consists of a 500 mL container with a degassing device that works by passing bubbles through the sample, a gas pump (AlphaPUMP), and a safety glass bottle which is connected to the detector within a closed loop, as shown in Figure 3. An active carbon filter is also used to reduce background radon concentrations in the system before every measurement.



**Figure 3.** (a) Schematic view of the experimental set-up [30]; and (b) practical setup in the laboratory.

Following the prescription of the supplier [33], before every water sample measurement, the system is first purged to reduce radon activity concentrations in the detector to background values for 10 min. After that, the water is introduced into the degassing vessel, and the AlphaGUARD and AlphaPUMP are turned on. After 10 min, the pump is switched off and the AlphaGUARD still measures the radon activity concentration for another 20 min. The AlphaGUARD monitor works in a “flow” mode and the radon concentration is recorded every minute. The flow rate of the pump is 0.5 L/min.

The AlphaGUARD monitor indirectly measures the activity of radon in water, since the radon that is expelled is diluted in air within the measurement setup, and a small part determined by the partition coefficient of the radon remains diluted in the aqueous phase. The concentration in water,  $C_{\text{water}}$ , in Bq/L is obtained by using:

$$C_{\text{water}} = \frac{C_{\text{air}} \times \left( \frac{V_{\text{system}} - V_{\text{sample}}}{V_{\text{sample}}} + k(T) \right) - C_0}{1000} \quad (1)$$

where  $C_{\text{water}}$  is the radon concentration in the water sample (Bq/L),  $C_{\text{air}}$  is the value of radon concentration in the measurement system provided by AlphaGUARD (Bq/m<sup>3</sup>),  $C_0$  is the system background radon concentration,  $V_{\text{system}}$  is the inner volume of the measurement system (mL), and  $V_{\text{sample}}$  is the volume of the water sample (mL).  $k(T)$  is an empirical diffusion coefficient which depends on the temperature of the water sample through the following expression [32]

$$k(T) = 0.105 + 0.405 \times e^{0.502 \times T(^{\circ}\text{C})} \quad (2)$$

To determine if part of the radon in the water sample is generated from <sup>226</sup>Ra, which could be dissolved in the own sample [13], a part of each water sample has been stored in their glass containers in order to re-measure the radon content after the time required for <sup>226</sup>Ra to reach secular equilibrium with its progeny.

## 4. Results and Discussion

### 4.1. Radon as a Natural Radioactive Tracer to Study Aquifer Systems

The results of groundwater radon concentrations for the 28 samples measured are shown in Table 1 and the spatial distribution of the results is shown in Figure 4. The average concentrations range from 0.3 to 76.9 Bq/L; the arithmetic mean is 12.8 Bq/L; the median of the distribution is 5.3 Bq/L; and the standard deviation is 17.7 Bq/L.

**Table 1.** Radon activity concentrations in the measured samples.

| Samples | Elevation (m above Sea Level) | Depth (m asl) | Depth Elevation (m asl) | Radon Concentration (Bq/L) |
|---------|-------------------------------|---------------|-------------------------|----------------------------|
| Rd1     | 235                           | 0             | 235                     | 76.9 ± 8.3                 |
| Rd2     | 108                           | 150           | −42                     | 51.9 ± 8.9                 |
| Rd3     | 430                           | 176           | 254                     | 0.3 ± 0.3                  |
| Rd4     | 354                           | 192           | 162                     | 5.7 ± 1.9                  |
| Rd5     | 725                           | 151           | 574                     | 7.4 ± 3.7                  |
| Rd6     | 235                           | 123           | 112                     | 1.3 ± 1.0                  |
| Rd7     | 679                           | 290           | 389                     | 12.5 ± 3.1                 |
| Rd8     | 603                           | 340           | 263                     | 30.8 ± 6.4                 |
| Rd9     | 345                           | 150           | 195                     | 19.2 ± 4.6                 |
| Rd10    | 515                           | 360           | 155                     | 3.8 ± 2.4                  |
| Rd11    | 377                           | 125           | 252                     | 28.2 ± 6.1                 |
| Rd12    | 996                           | 450           | 546                     | 1.8 ± 1.0                  |
| Rd13    | 499                           | 200           | 299                     | 14.8 ± 4.1                 |

Table 1. Cont.

| Samples | Elevation (m above Sea Level) | Depth (m asl) | Depth Elevation (m asl) | Radon Concentration (Bq/L) |
|---------|-------------------------------|---------------|-------------------------|----------------------------|
| Rd14    | 452                           | 400           | 52                      | 0.9 ± 0.8                  |
| Rd15    | 620                           | 315           | 305                     | 9.7 ± 2.6                  |
| Rd16    | 740                           | 280           | 460                     | 2.8 ± 1.2                  |
| Rd17    | 260                           | 170           | 90                      | 0.8 ± 0.8                  |
| Rd18    | 523                           | 230           | 293                     | 37.3 ± 8.1                 |
| Rd19    | 420                           | 372           | 48                      | 4.2 ± 1.9                  |
| Rd20    | 625                           | 400           | 225                     | 4.9 ± 1.7                  |
| Rd21    | 810                           | 302           | 508                     | 2.6 ± 1.4                  |
| Rd22    | 481                           | 143           | 338                     | 12.4 ± 3.3                 |
| Rd23    | 750                           | 608           | 142                     | 1.8 ± 0.9                  |
| Rd24    | 1050                          | 540           | 510                     | 3.1 ± 1.4                  |
| Rd25    | 445                           | 340           | 105                     | 3.4 ± 1.5                  |
| Rd26    | 598                           | 394           | 204                     | 10.9 ± 2.8                 |
| Rd27    | 182                           | 150           | 32                      | 6.6 ± 2.5                  |
| Rd28    | 827                           | 355           | 472                     | 3.7 ± 1.5                  |

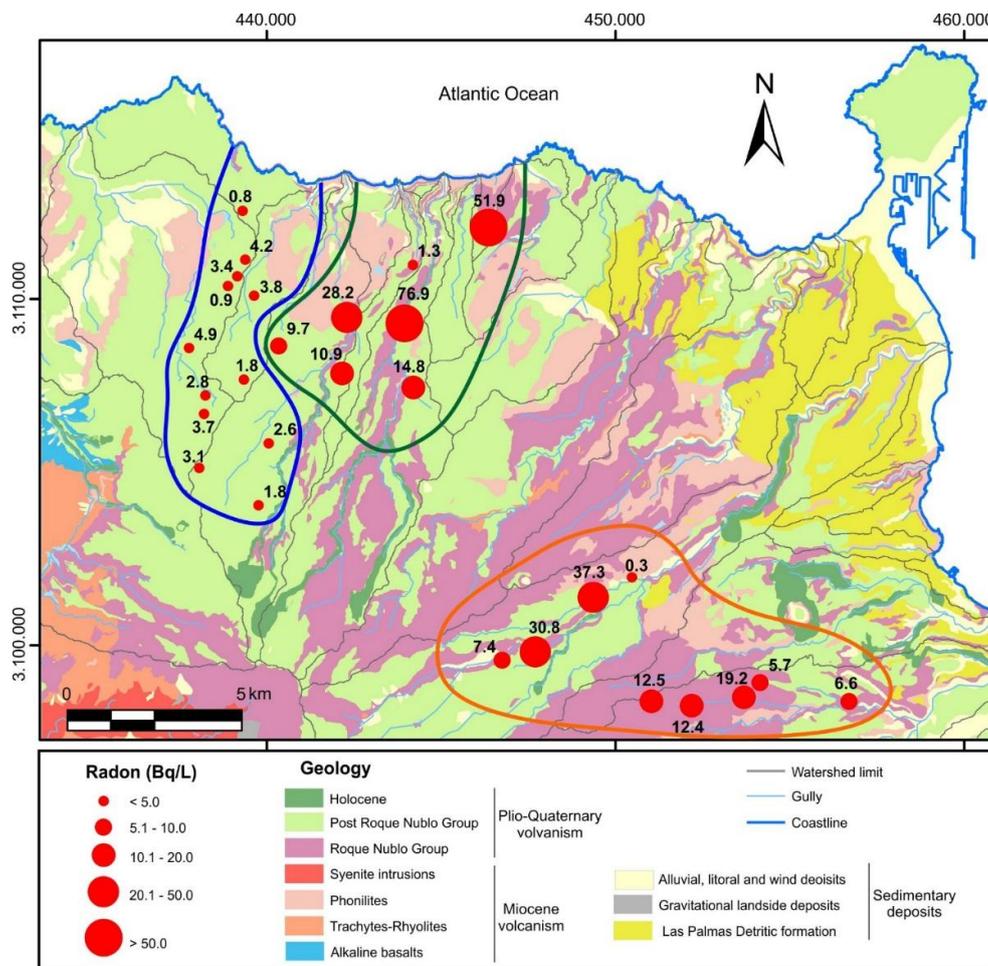


Figure 4. Spatial distribution of radon activity concentration. Samples are grouped depending on concentration of radon and geographic distribution. The blue group is located in the Guia-Moya basins, the green group in the Moya-Azuaje basins, and the orange group in the Guiniguada-Telde basins.

Several studies related to the use of radon as a hydrogeochemical tracer can be found in the literature, [7,34,35]. The main factors that influence the presence of  $^{222}\text{Rn}$  in groundwater are the content of  $^{226}\text{Ra}$  in the reservoir rock and its emanation coefficient, as well as the feasibility of mixing of various groundwater components [36,37].  $^{226}\text{Ra}$  is present in all rocks and soils in variable amounts and, as mentioned above, it is more abundant in volcanic acid rocks such as phonolites than in basic ones such as basalts. Therefore, higher concentrations of  $^{222}\text{Rn}$  would be characteristic of groundwaters flowing through volcanic acid rocks. The behavior of dissolved  $^{222}\text{Rn}$  in groundwater is strongly influenced by the properties of the mother rock, in particular by the distribution of  $^{226}\text{Ra}$  (precursor of  $^{222}\text{Rn}$ ), in relation to surface of pores and fissures in the rock where the interchange with groundwater occurs [38]. The half-life of radon is 3.82 days and it disappears in approximately 38 days by radioactive decay. Taking into account the representative velocity of groundwater, a radon particle cannot move more than a few dozen meters away from where it was incorporated [36]. This distance could vary depending on the aquifer transmissivity and  $^{222}\text{Rn}$  will reach greater distances (hundreds of meters) when groundwater circulates through fissures or fractures. In consequence, for a porous medium, the radon concentration in a sample is representative of the location where the sample was taken. Nevertheless there are many factors that can affect this ideal behavior, such as the recharging or mixing processes with water with low radon concentrations, or the temperature of the groundwater, since the solubility of radon gas increases as temperature rises.

The spatial distribution of radon allows the establishment of three classes or groups (Figure 4) that could explain the similar behavior of groundwater in wells that are near each other, but located in different catchments.

- (a) Northwestern group (Guía-Moya). This group (blue line in Figure 4) has lower radon concentration values (lower than 10 Bq/L) and it is mainly located in the Guía basin but includes some points associated with the Moya basin. The samples included in this group are Rd10, Rd12, Rd14, Rd16, Rd17, Rd19, Rd20, Rd21, Rd23, Rd24, Rd25, and Rd28. Some statistical parameters are: arithmetic mean, 2.82 Bq/L; geometric mean, 2.47 Bq/L; median, 2.95 Bq/L; and standard deviation, 1.29 Bq/L. According to the deep geology of the area (cross section I-I' in Figure 1), groundwater in these areas flow through basic rocks with little content in radon precursors.
- (b) Northwestern group (Guía-Moya). This group (blue line in Figure 4) has lower radon concentration values (lower than 10 Bq/L) and it is mainly located in the Guía basin but includes some points associated with the Moya basin. The samples included in this group are Rd10, Rd12, Rd14, Rd16, Rd17, Rd19, Rd20, Rd21, Rd23, Rd24, Rd25, and Rd28. Some statistical parameters are: arithmetic mean, 2.82 Bq/L; geometric mean, 2.47 Bq/L; median, 2.95 Bq/L; and standard deviation, 1.29 Bq/L. According to the deep geology of the area (cross section I-I' in Figure 1), groundwater in these areas flow through basic rocks with little content in radon precursors.
- (c) Northern group (Moya-Azuaje). This group (green line in Figure 4) has the highest radon concentration values (reaching 76.9 Bq/L) and it is mainly located in the Azuaje basin but includes some points from the Moya basin. The samples included in this group are Rd1, Rd2, Rd6, Rd11, Rd13, Rd15 and Rd26. These data have an arithmetic mean of 27.67 Bq/L; geometric mean of 15.64 Bq/L; median of 14.8 Bq/L; and a standard deviation of 27.31 Bq/L.

In this case the high values of radon could suggest that the underlying territory, through which the groundwater flows, has a great proportion of acidic rock with higher contents of radon precursors as shown by the deep geology of the area (cross section II-II' in Figure 1).

- (d) Northeastern group (Guiniguada-Telde). This group (orange line in Figure 4) has intermediate radon concentration values and combines points belonging to both the Guiniguada and Telde basins. The samples included in this group are Rd3, Rd4, Rd5, Rd7, Rd8, Rd9, Rd18, Rd22, and Rd27. These data have an arithmetic mean of 14.69 Bq/L; geometric mean of 8.7 Bq/L; median of 12.4 Bq/L; and a standard deviation of 12.28 Bq/L. The characteristics of this group are similar to the Moya-Azuaje group. Although there is no geological cross section for this area, and many of the wells are located at the Roque Nublo group, phonolite outcrops are observed, so that a greater proportion of basic rock in the subsurface is plausible.

A box-and-whisker plot is shown in Figure 5. In this graphic, the values of radon activity concentration have been compiled for the three groups (Guia-Moya basins, Moya-Azuaje basins, and Guiniguada-Telde basins). The results show significant statistical differences between the groups depending on the location of the samples. Based on the knowledge of the deep geology of the area, we can infer that measurement of radon concentration can provide reliable indications of the type of rock in the vicinity of the sampling point, which does not necessarily coincide with surface geology.

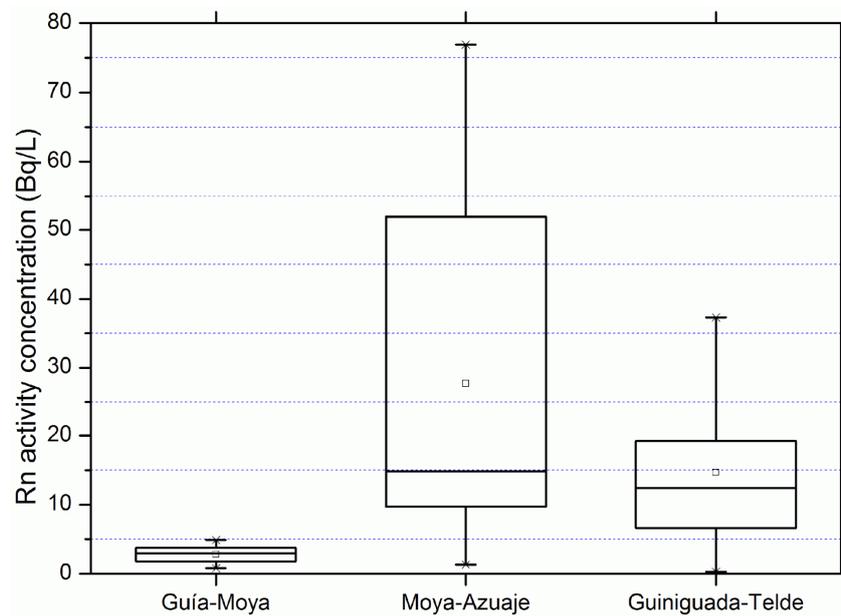
Since well depth is known, the presence of layers of different subsurface materials can be estimated. Figure 6 shows a Box-and-whisker plot for concentration of radon grouped by well depth elevation (m a.s.l.). Wells with a depth elevation, between 0 and 200 m a.s.l., have a lower concentration of radon; this may be due to an environment formed by basic rocks. Wells with a depth elevation, between 200 and 400 m a.s.l., have a higher concentration of radon, which could be due to the presence of phonolites (intermediate volcanic rocks) around groundwater at these depths. Finally, the Rd5, Rd12, Rd16, Rd21, Rd24 and Rd28 wells that exploit groundwater below 400 m have a lower concentration of radon. This could suggest that the surrounding area around these wells is dominated by basaltic basic rocks, but these low concentrations could also be attributed to the double pumping system of these wells, so loss of radon gas during the extraction process is possible, although the possible infiltration of surface water must be also considered because these wells are located at higher levels (Figure 2) where most of the recharge occurs in the area [39].

To determine the origin of the radon dissolved in the groundwater, after two months the radon activity concentration of each sample was measured again. This allows us to test if the samples contained a dissolved radon precursor ( $^{226}\text{Ra}$ ). The values of radon activity were under 1 Bq/L for all the samples. This allows us to assume that the radon is sourced directly from the surrounding bedrock through which it flows.

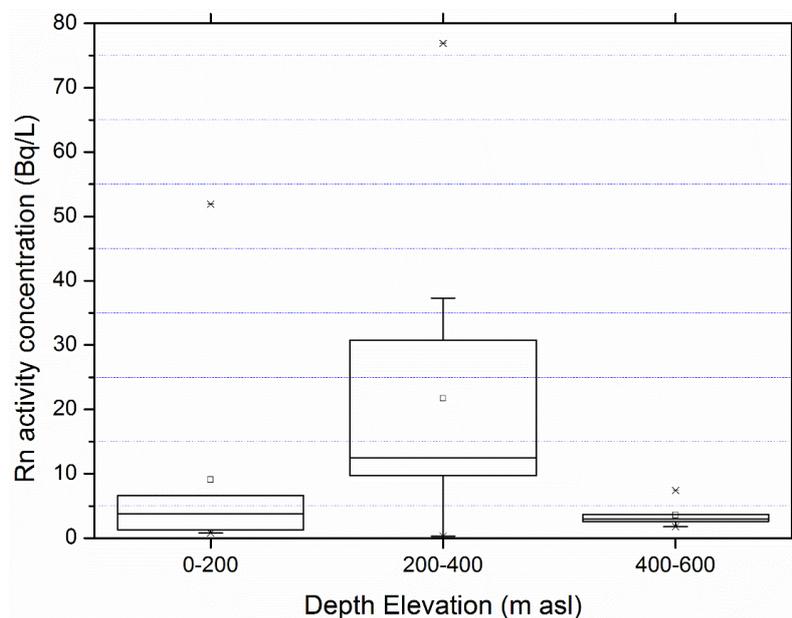
#### 4.2. Radiological Quality of Ground Waters: Radon and Gross Alpha Activity

The International Commission on Radiological Protection (ICRP) suggests that radionuclides in water are absorbed more easily than radionuclides in food [40]. As mentioned above, in groundwater  $^{222}\text{Rn}$  occurs in dissolved form and radon activity concentrations range from a few to thousands of Bq/L, in contrast to the surface water where  $^{222}\text{Rn}$  generally has very low concentrations [41,42]. The Council of the European Union states in its directive 2013/51/EURATOM of 22 October 2013 that the

“requirements for the protection of the health of the public with regard to radioactive substances in water intended for human consumption has established a limit of 100 Bq/L of radon activity”. As shown in Table 1, all measured values are below this limit.



**Figure 5.** Box-and-whisker plot for the radon activity concentration values, grouped depending on concentration of radon and geographic distribution.



**Figure 6.** Box-and-whisker plot for concentration of radon (Bq/L) grouped by well depth elevation (m a.s.l.).

According to the Przylibski and Gorecka classification [37], three samples with a radon concentration between 0.1 and 1.0 Bq/L belong to Radon Free Water, 15 in a range of 1.0–10.0 Bq/L are Radon Poor Water and 10 between 10.0 and 100.0 Bq/L are part of Low Radon Water. Despite that

the radon concentrations of this last group (Low Radon Water)) are lower than the threshold value of 100 Bq/L, a new radiological analysis to determine the gross alpha activity was done.

Gross alpha activity is a frequently used parameter for monitoring the radiological quality of water intended for human consumption and involves the activity due to all alpha emitter radionuclides in the water. The aforementioned European directive also recommends a screening level for gross alpha activity of 0.1 Bq/L, above which analyses for specific radionuclides should be required.

To determine the gross alpha activity of the well water samples, a radiochemical procedure, the coprecipitation method [43,44], was followed to prepare samples for counting in a ZnS(Ag) scintillation detector.

The gross alpha activity ( $A$ ) and its uncertainty ( $u(A)$ ) in Bq/L were calculated using Equations (3) and (4), respectively [44]:

$$A = \frac{cpm_S - cpm_B}{60 \times E \times F_a \times V} \quad (3)$$

$$u(A) = \frac{2}{60 \times E \times F_a \times V} \sqrt{\frac{cpm_S}{T_S} - \frac{cpm_B}{T_B}} \quad (4)$$

where  $cpm_S$  is the count-rate of the sample;  $cpm_B$  the count-rate of the blank;  $E$  is the efficiency determined from the certified standard of  $^{241}\text{Am}$  P2177/LMRI/RN/1068 provided by CIEMAT (Centro de Investigaciones Medioambientales y Tecnológicas, Madrid, Spain);  $F_a$  is the self-absorption factor, which is calculated as a function as the weight of the final precipitate in milligrams [43];  $V$  is the volume in liters;  $T_S$  is the measuring time of the sample in minutes; and  $T_B$  is the measuring time of the blank in minutes.

Some gross alpha activity values exceed the screening level for human consumption water (Table 2), so detailed analysis should be conducted in order to determine their radioisotopic composition.

**Table 2.** Gross alpha activity values for water with radon activity concentrations higher than 10 Bq/L.

| Samples | Group     | A (Bq/L)      |
|---------|-----------|---------------|
| Rd18    | Northeast | 0.044 ± 0.004 |
| Rd8     | Northeast | 0.172 ± 0.007 |
| Rd9     | Northeast | 1.08 ± 0.02   |
| Rd7     | Northeast | 0.046 ± 0.004 |
| Rd22    | Northeast | 0.034 ± 0.004 |
| Rd1     | North     | 0.306 ± 0.004 |
| Rd2     | North     | 0.94 ± 0.02   |
| Rd13    | North     | 0.004 ± 0.001 |
| Rd11    | North     | 0.344 ± 0.009 |
| Rd26    | North     | 0.087 ± 0.006 |

## 5. Conclusions

Water samples were collected from 28 groundwater locations and analyzed for their radon content. The results obtained for the radon activity concentration present minimum, maximum, arithmetic mean and standard deviation of 0.3 Bq/L, 76.9 Bq/L, 12.85 Bq/L and 17.72 Bq/L, respectively.

The hydrogeological study provides some evidence for a relationship between radon activity concentration and the aquifer material characteristics, suggesting the feasibility of using radon groundwater measurements as tracers for the geological composition of subsurface material flows. In order to obtain more conclusive results, a larger number of samples, and other analysis techniques, such as determination of the concentration of radioisotopes in the groundwater and their geological environment would be appropriate.

From the point of view of radiological protection, the radon levels measured in groundwater in the present work are lower than the minimum level established by the European Commission (100 Bq/L) for taking remedial action in order to comply with requirements for the protection of human health. Nevertheless, in this study, gross alpha activity values higher than the prescriptive screening level (0.1 Bq/L) for water fit for human consumption have been found for some samples showing radon activity concentrations between 10 and 100 Bq/L (Low Radon Water). Therefore, even for water samples with low level of radon activity concentration, other radiological analyses, such as gross alpha activity determination, are recommended to ensure water quality that is adequate for human consumption.

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### Author Contributions

Authors collaborated together for the completion of this work and in the discussion about the role of the radon as hydrogeological tracer. The radon measurement was done by Héctor Alonso, Jonay González-Guerra and Miguel A. Arnedo. The gross alpha determination and the radiological protection analysis were performed by Alicia Tejera, Jesús G. Rubiano and Pablo Martel. The manuscript was mainly written by Héctor Alonso, Tatiana Cruz-Fuentes and Jesús G. Rubiano and reviewed by all authors. Tatiana Cruz-Fuentes, Alejandro Rodriguez-Gonzalez, Francisco J. Pérez-Torrado and María del Carmen Cabrera performed the fieldwork, sample and data collections and geological discussion.

### Conflicts of Interest

The authors declare no conflict of interest.

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