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Radon-Specific Activity in Drinking Water and Radiological Health Risk Assessment: A Case Study

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Abstract: In this paper, the evaluation of the Rn-222 radioactivity content in drinking water samples from the Calabria region, southern Italy, is reported as a case study. The Rn-222-specific activity in the analyzed groundwater samples for human use was evaluated by using the PerkinElmer Tricarb 4910 TR setup and compared with the parameter value (100 Bq L^{-1}) reported in the reference Italian legislation, i.e., D.Lgs. 28/2016, derived from the European Directive 2013/51/Euratom. The radiological health risk for the population of the investigated area, due to the ingestion and inhalation of Rn-222 dissolved in water, was then evaluated by calculating the total annual effective dose, only in those cases where the parameter value was exceeded. The obtained results represent a main reference for the investigated area and are useful for determining any possible radiological health risk for human beings related to the ingestion of the investigated radionuclide. Moreover, they can also be used as a baseline for future investigations regarding background radioactivity levels.

Keywords: radon; drinking water; liquid scintillation counting; radiological risk; effective dose

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1. Introduction

The natural radioactivity in the environment, due to the presence of cosmogenic and primordial radioisotopes in the Earth's crust, contributes the highest percentage to the dose received by the population [1–3]. Although environmental aspects of natural radioactivity have been widely discussed in the literature [4–6], the presence of natural radioisotopes in drinking water has not been sufficiently addressed up to now, despite the fact that water represents a critical component of the surrounding environment. In fact, it is worthy of note that the health protection of the population cannot be separated from ensuring the quality of water for human consumption, as drinking water should not pose a health risk over a lifetime [7–9]. In particular, as water is a critical component of the environment, its quality can be harmed by increased pollution, human activity and a high concentration of naturally occurring radioactive elements [10,11].

As widely reported in the literature, drinking water has a natural radioactivity content that is strictly dependent on its origin [12]. In particular, among the naturally occurring radioisotopes, radon is one of the most remarkable, since, as is well known, its ionizing radiation provides the major contribution of internal human exposure compared with other natural sources [13,14]. Radon exhales from rocks and easily migrates and enters fracturing groundwater bodies [15]. Each rock type has a characteristic radon content, with the highest values for granitic and metamorphic rocks, phosphate rocks, black shales and some carbonate rocks [16]. Although the solubility of radon in water is relatively low [17], its specific activity in this environmental matrix may instead be several orders of magnitude higher than that of other natural radionuclides [18]. This could also lead to an increase in indoor airborne radon activity concentration, since it was estimated that the transfer coefficient of radon from water to indoor air is 10^{-4} [19]. Therefore, a very high concentration of radon in drinking water poses a serious public health hazard because it can lead, in the long term, to the development of diseases of internal organs, i.e., stomach cancer from ingestion and lung cancer from inhalation [19,20].

In light of this, in order to protect the public from possible health consequences, it is necessary to investigate radon levels in drinking water [21,22]. This is usually achieved by using reliable measuring devices with reasonably low detection limits, affordable price and simple operation, now available in many laboratories [2,23].

The results of a campaign to measure the specific activity of radon in groundwater samples for human use coming from civic springs of the Reggio Calabria district, an aquifer system located in the Calabria region, southern Italy, are reported in this article in order to (i) increase the available experimental data with respect to the presence of Rn-222 in groundwater samples for human use from the area under investigation and (ii) to ascertain the relative health risk to members of the population. The overall approach consisted of an initial screening to assess if activity concentration values (in Bq L^{-1}) were below the parameter value (100 Bq L^{-1}) reported in the Italian D.Lgs. 28/2016 [24], and a second, deeper investigation, based on the assessment of radiological risk to public health by computing the total annual effective dose associated with ingestion and inhalation of the investigated radionuclide, only in those cases where the parameter value was exceeded [25].

2. Materials and Methods

2.1. Sample Collection

Four samples of groundwater for human use (one for each season of the year 2022) were collected for each of the nine selected Calabrian locations (ID#, # = 1, . . ., 9), southern Italy, as detailed in Table 1 and indicated in Figure 1.

Table 1. The IDs and GPS details of the sampling locations.

Site ID	GPS Coordinates	
	Latitude	Longitude
1	38°21′03.0″ N	16°10′35.0″ E
2	38°23′39.1″ N	16°11′17.3″ E
3	38°25′30.8″ N	16°09′41.5″ E
4	38°16′42.4″ N	16°09′46.7″ E
5	38°18′17.4″ N	16°06′40.2″ E
6	38°19′57.4″ N	16°09′37.1″ E
7	38°16′24.7″ N	16°04′47.3″ E
8	38°20′00.5″ N	16°09′20.7″ E
9	38°13′24.4″ N	15°59′43.8″ E

The sample collection was carried out according to the local weather conditions, which severely limit the access to the monitoring stations at times [25]. Moreover, the sampling, packaging and preservation of the samples were carried out according to [26].

2.2. Liquid Scintillation Counting (LSC) Measurements

The activity concentration of *Rn*-222 in the investigated drinking water samples was obtained through liquid scintillation counting (LSC) measurements, according to [27]. Specifically, 10 mL of each sample was inserted with a gas-tight syringe into the bottom of a 25 mL plastic vial previously filled with 10 mL of Perkin Elmer Opti-Fluor O scintillating cocktail immiscible in water, stored and, after a rest time of 5 h, counted for 60 min together with a background [28]. The scintillator was a Perkin-Elmer Tricarb 4910 TR, with an energy range of 0–2 MeV (β particles) and 0–10 MeV (α particles). Its minimum acceptable efficiency is 60% for H-3 (0–18.6 keV) and 95% for C-14 (0–156 keV). Its average background is 17 CPM for H-3 and 26 CPM for C-14. It operates in normal/low-activity–high-sensitivity mode, with the external Ba-133 standard to account for chemical and optical quenches and to assess the counting efficiency by using the tSIE/AEC [29].

Figure 1. The map of the investigated area.

A picture of the detector is presented in Figure 2.

During the radon-in-water analysis or if a water sample is taken and analyzed sometime later (rather than immediately), the sample's radon concentration will diminish, mainly due to radioactive decay and, partly, to the degassing phenomenon [30]. Then, it is essential to correct the resulting activity concentrations in order to take into account the decay from the sampling time to the analysis time. The decay correction is described by a simple exponential function with a time constant of 132.4 h, coming from the exponential law for radioactive decays:

$$C_{Rn-222} = C_{0,Rn-222}e^{-\lambda t}, \quad (1)$$

where $C_{Rn\text{-}222}$ is the measured *Rn*-222 concentration, $C_{0,Rn\text{-}222}$ is the initial concentration at the sampling time and t is the time elapsed since collection (hours).

Figure 2. A picture of the Perkin-Elmer Tricarb 4910 TR detector.

The time elapsed from sampling to measurement is less than 48 h in all cases, in order to minimize the *Rn*-222 content due to the decay of Ra-226 in the investigated samples.

Moreover, the uncertainty associated with the measurement of the *Rn*-222 concentration for each of the four investigated samples (per single sampling location) is given by [27]

$$U(C_{Rn-222}) = C_{Rn-222}\sqrt{\left(\frac{U_{CN}}{C_N}\right)^2 + \left(\frac{U_\varepsilon}{\varepsilon}\right)^2 + \left(\frac{U_V}{V}\right)^2}, \quad (2)$$

where U_{CN} is the uncertainty associated with the net counts C_N of the detector calibration source, U_ε is the uncertainty associated with the detection efficiency ε and U_V is the uncertainty associated with the volume V of the analyzed sample [27].

The quality of the LSC experimental results was certified by the Italian Accreditation Body (ACCREDIA). This implies the continued verification (with annual periodicity) of the maintenance of the LSC method's performance characteristics [31].

2.3. Evaluation of the Radiological Health Risk

The radiation dose from radon gas in drinking water is ingested and inhaled. Thus, with the aim of monitoring the radiation exposure of the population, the assessment of the annual effective dose due to the ingestion of *Rn*-222 in drinking waters was carried out [32]:

$$H_{ing}\left(Svy^{-1}\right) = DCF_{ing} \times C_{Rn-222} \times I_w \times 365, \quad (3)$$

where DCF_{ing} (Sv Bq^{-1}) is the dose conversion factor for ingestion of *Rn*-222 in water samples (23, 5.9 and 3.5 nSv Bq^{-1} for infants, children and adults, respectively) and I_w (L day^{-1}) is the average daily water consumption rate [20]. In detail, a per capita consumption of 150, 350 and 730 L per year for infants, children and adults, respectively, was defined [33].

Moreover, the contribution to the total effective dose due to inhalation of *Rn*-222 present in the investigated samples is given by [34]

$$H_{\boldsymbol{inh}}\left(Svy^{-1}\right) = C_{Rn-222} \times R \times F \times O \times DCF_{inh}, \quad (4)$$

where R is the transfer coefficient of radon from water to indoor air, equal to 10^{-4}; F is the equilibrium factor between radon gas and its progeny, equal to 0.4; O is the average annual number of hours spent indoors by a single individual, equal to 7000, and DCF_{inh} is the inhalation dose conversion factor of *Rn*-222, equal to 9 nSv Bq^{-1} h^{-1} m^3 [35].

3. Results and Discussion

In Table 2, the annual mean value (±standard deviation) of the *Rn*-222-specific activity in the investigated drinking water samples is reported for each collection site.

Table 2. The annual mean value (±standard deviation) of the *Rn*-222-specific activity in the investigated drinking water samples, for each collection site.

Site ID	$C_{Rn\text{-}222}$ (Bq L^{-1})
1	164 ± 46
2	7.4 ± 1.4
3	13.1 ± 2.4
4	9.1 ± 1.6
5	76 ± 18
6	38.9 ± 8.3
7	15.5 ± 2.8
8	64 ± 15
9	32.4 ± 6.6

It can be noticed that that the radon activity concentration varies from a minimum of (7.4 ± 1.4) Bq L^{-1} for the site ID2 to a maximum of (164 ± 46) Bq L^{-1} for the site ID1, thus suggesting that the provenance of these drinking water samples is different and that they originate from different depths and pass through distinct geological strata. This uneven distribution of the specific activity may be dependent on the amount of *Rn*-222 in the aquifer rocks of different locations and on the residence time of water/rocks-soils in contact [36]. Specifically, the highest value was recorded for a site situated within the geological context of the "Calabrian-Peloritan arc" [37]. This particular geological setting is known for its abundance of uranium-rich rocks, resulting in elevated levels of radon gas.

In addition, the relative uncertainty for the site ID1 is higher than in other cases; this is due, according to Equation (2), to the higher *Rn*-222 activity concentration measured in the four investigated samples picked up from this sampling location.

Moreover, in all cases, with the only exception of site ID1, the activity concentration values were found to be always lower than 100 Bq L^{-1}, i.e., the parameter value according to the Italian legislation [24]. Then, in this case, the assessment of the total annual effective dose due to the ingestion and inhalation of *Rn*-222 by infants, children and adults was carried out by using Equations (3) and (4), respectively. Specifically, the annual effective dose due to ingestion of *Rn*-222 was 0.56 mSv y^{-1}, 0. 4 mSv y^{-1} and 0.42 mSv y^{-1} for infants, children and adults, respectively. These values fall within the acceptable range of 0.2–1.8 mSv y^{-1} reported in the literature [38]. Furthermore, the annual effective dose due to inhalation of *Rn*-222 in the analyzed water sample was 0.41 mSv y^{-1}. Therefore, the total annual effective dose was found to be 0.97 mSv y^{-1}, 0.75 mSv y^{-1} and 0.83 mSv y^{-1} for infants, children and adults, respectively. All these results, obtained in a fully precautionary scenario, are below the 1 mSv y^{-1} limit value recommended by the World Health Organization (WHO), thus allowing us to reasonably exclude any possible radiological health risk related to the radon exposure for the population living in the investigated area [39].

4. Conclusions

In this paper, the evaluation of the radon content in groundwater samples for human use coming from nine selected locations of Calabria, southern Italy, representative of the investigated area, is reported as a case study.

The radon-specific activity was measured by means of a liquid scintillation counting setup, and the experimental results provide evidence that the activity concentration of radon is below the parameter value indicated by the current Italian legislation (100 Bq L^{-1}), except for the site ID1. In this latter case, the corresponding annual effective doses for infants, children and adults due to the ingestion of *Rn*-222 were shown to fall within the acceptable range of 0.2 mSv y^{-1}–1.8 mSv y^{-1} reported in Recommendation 2001/928/Euratom. Moreover, the total (ingested and inhaled) annual effective doses for infants, children and adults were found to be below the 1 mSv y^{-1} limit value recommended by the World Health Organization (WHO), thus reasonably ensuring the safety of the analyzed samples for drinking purposes, and no remedial actions are demanded.

Although these results display low levels of radon-specific activities and doses, in complete accordance with official institutional guidance, it should be highlighted that regular monitoring is necessary to ensure the safety of drinking water. In light of this, the data reported in this paper will be supplemented in the near future by increasing the sampling points and the number of analyzed drinking water samples. Moreover, it should be remarked that the approach reported in this article might be applied, in principle, for the assessment of any potential radiological hazard for human beings due to the presence of radioactive elements in drinking water, by constituting a guideline for investigations focused on the monitoring of the radiological quality of these samples.

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