



Radon in spring waters in the south of Catalonia



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ABSTRACT

Spring waters in the south of Catalonia were analysed to determine the ^{222}Rn activity in order to be able to establish a correlation between the obtained values with the geology of the area of origin of these samples, and also estimate the potential health risks associated with ^{222}Rn . Most of the analysed samples (90%) show ^{222}Rn activities lower than 100 Bq/L (exposure limit in water recommended by the World Health Organisation and EU directive 2013/51/EURATOM). However, in some cases, the activity values found for this isotope exceeded those levels and this can be attributed to the geology of the area where the spring waters are located, which is predominantly of granitic characteristics. To verify the origin of the radon present in the analysed samples, the obtained activity values were compared with the activities of its parents (^{226}Ra , ^{238}U and ^{234}U). Finally, we have calculated the annual effective dose from all the radionuclides measured in spring water samples. The results showed that the higher contribution due to spring water ingestion come from ^{222}Rn and ^{226}Ra . The resulting contribution to the annual effective dose due to radon ingestion varies between 10.2 and 765.8 $\mu\text{Sv/y}$, and the total annual effective dose due to his parents, ^{226}Ra , ^{234}U and ^{238}U varies between 0.8 and 21.2 $\mu\text{Sv/y}$ so the consumption of these waters does not involve any risks to population due to its natural radioactivity content.

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1. Introduction

Drinking water is usually provided by groundwater or surface water sources. Different substances present in the water source, such as radioactive elements, can dissolve in water and contaminate drinking water resources. The content of these natural radionuclides in groundwater can vary in several orders of magnitude and are influenced by physical (temperature), chemical (pH) and geological properties of the aquifer (Duggal et al., 2013; Isam Salih et al., 2002; Jobbágy et al., 2010; Przylibski and Gorecka, 2014). For instance, felsic rocks, such as granite, present high contents of incompatible elements such as ^4K , ^{238}U and ^{232}Th (Moreno et al., 2014). Radium can occur in rocks, soils and sediments, so it can be found in virtually every aquifer matrix (Sahin et al., 2013; Todorovic et al., 2012).

Radon (^{222}Rn), a decay product of ^{226}Ra , is a naturally occurring inert radioactive gas with a half-life of 3.82 days. When ^{226}Ra decays, ^{222}Rn atoms might be ejected from the soil grain by α -recoil and transferred to groundwater or void air and finally escape

through the atmosphere (Abdallah et al., 2007; Somlai et al., 2007; Tabar and Yakut, 2014).

The primary health effect of ^{222}Rn is lung cancer, resulting from inhalation in indoor air. There is also evidence from epidemiology that ingestion of ^{222}Rn causes stomach cancer (Ravikumar and Somashekar, 2014; Todorovic et al., 2012).

In order to protect the population from radiation exposure from ^{222}Rn it is necessary to analyse the presence of this radionuclide on the water sources, particularly underground sources. In this respect, an European Directive has been published recently laying down the requirements for the protection of the health of the general public with regard to radioactive substances in water intended for human consumption (EURATOM, 2013). The recommended reference level of effective dose received from drinking water consumption is 100 $\mu\text{Sv/y}$. This value excludes the dose received from ^3H , ^{40}K , ^{222}Rn and its decay products but a specific reference value of 100 Bq/L for ^{222}Rn has been also established. However, it should be pointed out that these recommendations do not apply to natural mineral waters and waters that are classified for medical benefits.

This study focuses on the determination of ^{222}Rn at different points in northeastern Spain, in the region of Catalonia, where numerous lithologies outcrop. The presence of numerous natural

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springs points to the possibility of elevated radioactivity and ^{222}Rn activity depending on the mineral composition of the rocks. In this respect, one important objective is to correlate the possible origin of this isotope through the analysis of its parents (^{226}Ra , ^{234}U and ^{238}U). Another aim is to determine the health hazards from the consumption of this water through the evaluation of the annual effective radiation dose.

2. Materials and methods

2.1. Materials and reagents

All chemical reagents used in this study were of analytical grade. Hydrochloric acid (37%), nitric acid (65%), hydrogen peroxide (30%), ammonia solution (25%), iron chloride (III), citric acid anhydrous and sulphuric acid were supplied by J.T. Baker (Deventer, Holland). Barium chloride dehydrate, lead nitrate, calcium nitrate tetrahydrate and sodium sulphate were supplied by Sigma–Aldrich. UTEVA columns were supplied by Eichrom (Bruz, France). A mineral oil scintillator (Ultima Gold F) and Optiphase Hisafe™ 3 were provided by Perkin Elmer™ (Turku, Finland).

A solution of ^{241}Am with a nominal activity concentration of 4.68 ± 0.41 KBq/g was provided by Amersham International, PLC (Buckinghamshire, England). Standards of ^{232}U and ^{226}Ra certified solutions with a well-known activity concentration value of 15.01 ± 0.15 Bq/g and 175.9 ± 0.7 Bq/g were provided by CIEMAT (Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas - Madrid, Spain).

Uranium was electroplated onto stainless steel planchets (diameter 25.15 mm and thickness 0.6 mm) supplied by Tecnasa (Madrid, Spain). Samples were filtered with a $0.45 \mu\text{m}$ filter supplied by Whatman (Maidstone, United Kingdom).

For the measurement of ^{222}Rn , Teflon-coated polyethylene vials with a volume of 20 mL were used, equipped with urea screw caps and aluminium foil liners. They were supplied by Perkin Elmer™.

2.2. Detectors

^{226}Ra was measured using a zinc sulphide (ZnS [Ag]) scintillator counter (photo multiplier tube and base preamplifier) model 2000 Canberra (Meriden, Connecticut) with a voltage range of 0.76–0.90 KeV. A planchet containing ^{241}Am with an activity concentration of 100 Bq was used for the monthly calibration. The background of each detector was determined by counting an empty planchet for 5000 min.

An alpha spectrometer was used to determine the individual isotopes of uranium (^{238}U , ^{235}U and ^{234}U). The alpha spectrometer (EG&G ORTEC, Model 676A, Oak Ridge, Tennessee) includes an ion-implanted silicon detector (ORTEC, size: 450 mm^2 ; alpha resolution: 25 keV FWHM at 5.48 MeV of ^{241}Am) in a vacuum chamber (Edwards Model E2M8, Sanborn, New York), a detector bias supplier, a preamplifier, a linear amplifier, and a multichannel pulse height analyser. Throughout the measurement, the pressure of the chamber was maintained at 1.06 kPa (10^{-2} Torr).

^{222}Rn was determined by ultra-low level liquid scintillation counting (LSC) in a Quantulus 1220™ from Perkin–Elmer (Turku, Finland) with WinQ and Easy View software.

2.3. Methods

The method used to determine the uranium isotopes (^{234}U , ^{235}U and ^{238}U) was described in a previous study by Mola et al. (Mola et al., 2013). In brief, this method consists of the precipitation of the actinides and separation of the uranium using an UTEVA resin (Eichrom, Bruz, France). The uranium solution was then transferred

to an electrodeposition cell and the uranium isotopes were electrodeposited for 2 h, under a current density of 1.5 A/cm^2 . One minute before the end of the electrolysis, 1 mL of concentrated ammonium hydroxide was added to ensure uranium deposit attachment on the stainless steel disk.

To determine ^{226}Ra , a method described by Palomo et al. (Palomo et al., 2007) was followed. This method is based on coprecipitation using barium chloride and sulphuric acid. Different clean-up steps were then applied to adjust the solution to different pH levels. Finally, the barium sulphate/radium sulphate precipitate was formed and isolated. This precipitate was deposited on a planchet for measurement in the ZnS (Ag) counter.

In the case of ^{222}Rn , the determination method consisted of taking an aliquot of 10 mL of sample (sampling details explained in Section 2.4) which was placed in a Teflon-coated polyethylene vial. This was then mixed with the scintillation cocktail Ultima Gold F until there was no empty space in the vial (about 12 mL). The vial was stirred vigorously and left in the instrument for 4 h prior to the measurement to attain equilibrium between ^{222}Rn and its progeny. The window for ^{222}Rn was set between 650 and 910 channels to be able to determine ^{222}Rn and its daughters ^{214}Po and ^{218}Po (Aleissa et al., 2013; Jowzaee, 2013; Salonen, 2010). Counting was performed for two cycles of 500 min. Pulse shape analysis (PSA) measurement mode was used for ^{222}Rn and in order to separate alpha and beta pulses into different spectra electronically.

2.4. Water samples

Sample locations of the analysed natural spring waters (15 samples) were considered on the basis of the different lithologies encountered (Table 1). Natural spring water samples were collected directly in 2L nalgene plastic containers for the determination of different radioactive isotopes. In addition, for the determination of ^{222}Rn , 100 mL of each sample were also collected in a glass bottle to avoid gas bubble formation. The samples were then capped immediately to prevent ^{222}Rn to degas into the atmosphere.

2.5. Ingestion doses

The annual effective radiation dose (E_D) for an individual consumer due to the water intake is calculated using the equation:

$$E_D = A_c \cdot I_a \cdot C_f$$

where A_c is the activity concentration from each radionuclide of ingested water in terms of Bq/L, I_a describes the annual intake of drinking water (the WHO and EPA estimate the average consumption of water in 2 L of water per day, 730 L/y) and C_f is the coefficient dose for adults, with a value of $6.5 \cdot 10^{-8}$, $2.8 \cdot 10^{-7}$, $4.9 \cdot 10^{-8}$, $4.5 \cdot 10^{-8}$ and $1 \cdot 10^{-8}$ Sv/Bq, for ^{224}Ra , ^{226}Ra , ^{234}U , ^{238}U and ^{222}Rn , respectively (UNSCEAR, 2008).

3. Results and discussion

3.1. Optimization of LSC method for ^{222}Rn determination

Firstly, we evaluated the effect of several parameters on the counting efficiency by LSC. In particular, we focused on the selection of the liquid scintillation cocktail and the sample volume needed to achieve the highest efficiency possible. For the optimization, a ^{226}Ra standard was used and, prior to the measurement, Teflon-coated vials was sealed to avoid ^{222}Rn degassing. An equilibration time of 21 days was needed to ensure decay equilibrium between ^{226}Ra and its progeny (^{222}Rn). For background measurement, an additional vial was prepared containing a mixture of

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