

^{226}Ra and ^{222}Rn IN NATURAL WATERS IN TWO TYPICAL LOCATIONS IN SPAIN

J. Soto, L. S. Quindós, N. Diaz-Caneja, I. Gutierrez and P. L. Fernández
Departments of Medical and Applied Physics
University of Cantabria, Spain

Abstract — A preliminary study is presented, preliminary to a full-scale national survey on ^{226}Ra and ^{222}Rn levels in surface and underground waters in two regions of Spain of very different geological composition, which were selected as representative of the two main geological types existing in the Iberian peninsula. ^{222}Rn concentrations in the different natural waters collected show values ranging from 0.8 to 75 Bq.l^{-1} in the first area and from 20 to 25,000 Bq.l^{-1} in the second. ^{226}Ra concentrations in domestic water supplies were found to be over the detection limit for almost all the samples collected, with values reaching about 4 Bq.l^{-1} in some cases.

INTRODUCTION

Radium and its daughter products constitute an important part of natural environmental radiation exposure⁽¹⁾. Since ingestion is a major pathway, apart from inhalation of radon, for internal irradiation, the measurement of radioactivity in drinking water from houses or wells is relevant in assessing the contribution of these environmental radiation hazards⁽²⁾. Exposure of the population, therefore, can only be accurately assessed by measuring the actual domestic supplies.

We have carried out a local survey of ^{226}Ra and ^{222}Rn concentrations in natural waters in two regions of Spain of very different geological composition, selected as representative of the two main geological types found in the Iberian peninsula. The first one, Cantabria, is located at the north of the country and sandstone, limestone and loam are the principal components of its soil. Granite and shale are more common in the second area, Arribes del Duero (Salamanca), near Portugal.

This paper reports a preliminary study with a view to developing a full-scale national survey on ^{226}Ra and ^{222}Rn contents in surface and underground waters.

MATERIAL AND METHODS

Water samples were collected in a 2.8 Lucite Marinelli beaker from springs, wells or houses, adding nitric acid to yield a $\text{pH} \approx 1$ acid solution. An appropriate transfer device was used to take the water directly from the source to the container without aeration. Whenever necessary the water was filtered through Whatman 41 filter paper to remove the suspended impurities. Samples were closed and left untouched for three hours afterwards to ensure that ^{222}Rn and its daughters reached secular equilibrium before measurement. The gamma ray spectra were obtained using a $3'' \times 3''$

NaI(Tl) detector coupled to a 1024 multichannel analyser. The detector and the sample to be measured were placed in a shielding chamber with four inch thick lead walls.

The counting time (20,000 s or longer) was selected so that the ^{222}Rn detection limit established at 0.8 Bq.l^{-1} could be reached in experimental conditions. Efficiency of the system was measured using standard solutions of ^{226}Ra from CIEMAT (Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas, Madrid).

^{222}Rn activities of samples were derived from the analysis of the 0.609 MeV and 1.76 MeV lines of ^{214}Bi at the same time taking into account the correction factor for decay between sampling and counting of the sample.

^{226}Ra content in the different samples was determined by means of liquid scintillation according to Pritchard and Gessell⁽³⁾. This method consists of drawing 10 ml from the initial water sample four weeks after the collection time to permit the decay of dissolved ^{222}Rn . This new sample is placed in a glass syringe that can be sealed. At a convenient later time 5 ml of toluene based liquid scintillation fluid is also drawn into the syringe and the two liquids are injected through a rubber septum into an evacuated 20 ml glass scintillation vial. After the three hours required to establish secular equilibrium between radon and radon daughters, the sample is counted in a Beckman LS8000. The detection limit for this technique was evaluated as 20 mBq.l^{-1} also using ^{226}Ra standard solutions.

SAMPLE COLLECTION

Location of the two regions studied is shown in Figure 1. The region of Cantabria has a surface area of 10,000 square kilometres and a population of 500,000. For the Arribes del Duero area, the surface

area is only 2000 square kilometres and the population is not more than 50,000.

²²⁶Ra, ²³²Th and ⁴⁰K content in soils and rocks from both regions were measured by gamma spectrometry in order to try to find a relationship.



Figure 1. Location in Spain of the two regions studied.

Table 1. Average concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K in soil for the regions of Cantabria and Arribes del Duero (Salamanca).

Area	²²⁶ Ra (Bq.kg ⁻¹)	²³² Th (Bq.kg ⁻¹)	⁴⁰ K (Bq.kg ⁻¹)	Number of samples
Cantabria	25 ± 4	26 ± 4	440 ± 50	30
Arribes del Duero	70 ± 9	35 ± 5	800 ± 80	20

Table 1 gives the geometric mean concentrations of these three radionuclides found in the two locations. Results, which are higher for the second region, are closely related to geological composition: limestone, sandstone and loam for Cantabria, slate and granite for Arribes del Duero. In this latter zone we can also find several old uranium mines, one of them still working.

The water samples in the two regions were collected during the winter of 1986 and spring of 1987. A total of 30 surface water samples from the eight main rivers and 40 from the domestic supplies from houses and wells in different towns around the region were analysed for Cantabria. For the Arribes del Duero area 23 samples from the two principal rivers and tributaries and 32 from houses and wells were measured.

EXPERIMENTAL RESULTS AND DISCUSSION

The ²²⁶Ra and ²²²Rn geometric mean concentrations for each type of water and location are given in Table 2. Also shown in brackets are the percentage of samples collected which correspond to each one of the different average concentrations obtained and the highest value found in each location.

A first analysis of these results shows that the concentrations of ²²⁶Ra and ²²²Rn in surface waters are similar in the two regions: almost all measurements are below detection limits. Only a few of the samples (5%) collected near the uranium mine mentioned show a higher value directly related to the mineral washing activity of the mine.

For domestic water supplies, ²²⁶Ra average concentrations obtained are about 20 mBq.l⁻¹ and

Table 2. ²²⁶Ra and ²²²Rn concentrations for the regions of Cantabria and Arribes del Duero (Salamanca).

		Cantabria		Arribes del Duero	
Surface water	²²⁶ Ra (Bq.l ⁻¹)	<0.02	(100%)	<0.02	(95%)
	²²² Rn (Bq.l ⁻¹)	<0.8	(100%)	<0.8	(100%)
Domestic supplies: houses and wells	²²⁶ Ra (Bq.l ⁻¹)	<0.02 0.02 – 0.1 >0.1	(80%) (10%) (10%)	<0.5 0.5 – 3.0 >3.0	(55%) (30%) (15%)
		Highest value: 0.4		Highest value: 4.0	
	²²² Rn (Bq.l ⁻¹)	<1.0 1.0 – 4.0 >4.0	(30%) (30%) (40%)	<100 100 – 1,000 >1,000	(30%) (60%) (10%)
		Highest value: 75		Highest value: 25,000	

1000 mBq.l^{-1} for Cantabria and Arribes del Duero, respectively, with over 15% of samples from Arribes del Duero showing values higher than 3000 mBq.l^{-1} .

For ^{222}Rn the average concentrations are now about 3 Bq.l^{-1} and 600 Bq.l^{-1} for Cantabria and Arribes del Duero, respectively. The origin of this difference must be sought in the water source, either rivers or wells, and the route from these points to the final consumer. In the water of Cantabria, the processing consisted of an aeration stage with subsequent slow filtration through a bed containing oxidising substances to precipitate metal contamination. In these conditions, ^{222}Rn is lost dramatically by aeration, giving low concentrations for the samples measured. The contrary occurs for the water in Arribes del Duero, where it goes directly from wells to the houses. The ^{222}Rn concentrations then rise to rather high levels, showing the influence of the processing method on the final concentration measured in drinking water⁽⁴⁾.

From the data given, the ^{226}Ra content classifies

the Arribes del Duero area as one of enhanced exposure for people living there, although their dose equivalent rate to bone surface from ^{226}Ra and its retained daughters would never be more than 0.5 mSv.y^{-1} compared to 0.1 mSv.y^{-1} estimated for average background areas such as the region of Cantabria and others⁽⁵⁾.

If, from the ^{222}Rn concentrations found in Arribes del Duero, we assume an intake of 0.5 l of unboiled water a day, with an average concentration of 600 Bq.l^{-1} , according to NCRP 77⁽⁵⁾ a stomach dose of approximately 16 mSv.y^{-1} and a whole-body dose equivalent of one hundred times less will be reached.

Although the contribution of drinking water to the total intake is small for average background conditions⁽⁶⁾, the high values measured in the Arribes del Duero area justify the interest in carrying out a national survey in Spain, if we bear in mind that a third of the country has a similar geological composition and that the population affected in this area is more than ten million.

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