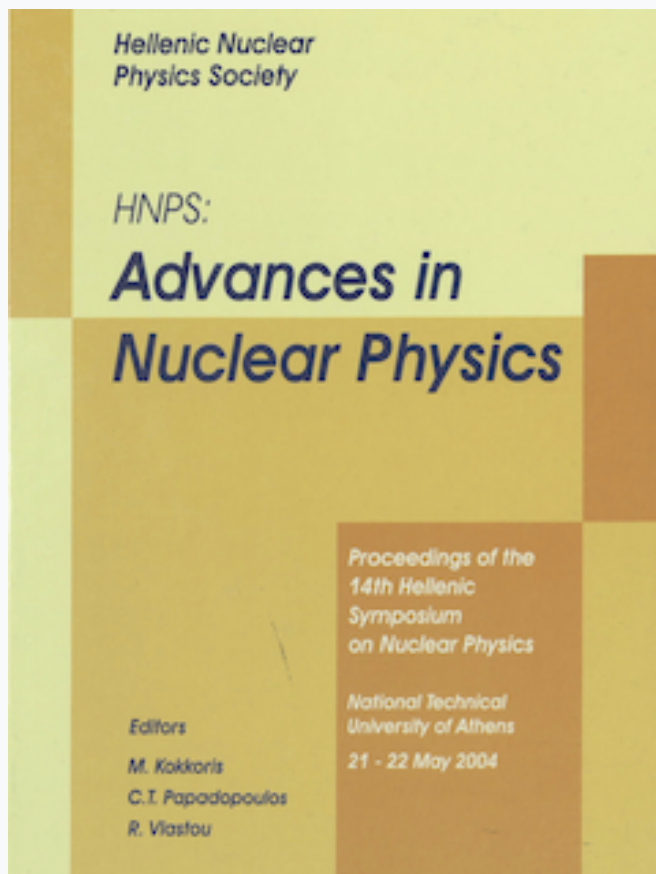


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Radon Measurements in Bottled Waters in Greece

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Abstract

Underground waters pass through various types of rocks, which may contain radium and uranium isotopes in such concentrations and chemical forms that could be diluted in the water body. Thus it is possible to contain significant quantities of these radioisotopes such as radium (^{226}Ra), which decays producing a series of alpha and beta emitters that remain in the water. Among them, radon (^{222}Rn) and isotopes of polonium (^{214}Po and ^{218}Po) contribute to significant doses. In this work, we measured radium/radon concentration levels in bottled waters commercially available in Greece. Samples were concentrated by a factor of 20-100, in order to obtain more accurate results, because in the majority of the samples the concentrations measured initially were very low. To measure the radioactivity concentrations, 10 mL of concentrated sample were added to 10 mL of scintillation cocktail in a 20 mL plastic vial and then were counted with a liquid scintillation analyzer. The results show that radium/radon concentrations of the bottled waters range from 7 - 70 mBq/L.

Key words: radon; radium; water; scintillation counting.

1 Introduction

Natural waters originating from springs or wells may contain natural radionuclides, predominantly of the uranium-radium series. Depending on various factors affecting the solubility of uranium-radium radionuclides in the bedrock, through which the waters pass, such as pH and the chemical form of the elements, as well as on the concentration of the elements in the bedrock, the concentration of these radionuclides may vary significantly. The isotope that contributes significantly to the ingestion dose via consumption of water is the

alpha emitting ^{226}Ra and its daughter radionuclides such as radon (^{222}Rn) and the isotopes of polonium (^{214}Po and ^{218}Po). ^{226}Ra is an alkaline earth metal and follows the metabolic pathways of calcium in the body.

Thus, a significant fraction of the ingested radium is deposited in the bone. ^{222}Rn , may contribute to the ingestion dose at higher concentration levels but such levels are not found in bottled waters due to the short half live of ^{222}Rn (3.8 days).

The consumption of mineral and table bottled water has been increasing in Greece in the past years, following the general trend observed in other European countries. One of the twelve brands of the waters analyzed in this work, claimed that company sales were raised about 1200% in the period 1992-2002. In the present investigation we assess for the first time the concentration levels of ^{226}Ra in bottled waters in Greece.

2 Materials and Methods

The method used to determine the activities of ^{226}Ra in bottled waters is based on the measurement of the concentration of its daughter radionuclide radon (^{222}Rn), which reaches equilibrium with his parent nuclide in about 30 days. Measurements of radon concentration were carried out with the Liquid Scintillation Analyzer Tri-Carb 3170TR/SL (Packard BioScience). This counter is situated in the Archaeometry Center of the University of Ioannina and it is capable to measure alpha and beta emitting radionuclides, using the liquid scintillation technique, with super low-level background.

Each sample was processed as follows: In 1 L of each sample 1 mL of concentrated HNO_3 was added and the solution was concentrated by evaporation in order to enrich the ^{226}Ra in the sample. There were two sets of measurements as shown in Table 1. The ratio of concentration varied from 15 to 98. After this processing, the samples were stored for about one month in order to establish the equilibrium between ^{226}Ra and its daughter ^{222}Rn . Then, 10 mL of each concentrated sample were added to 10 mL of scintillation cocktail Opti-Fluor O (Packard BioScience) in a 20 mL capacity plastic vial. The vials were stirred vigorously for 10 seconds in order radon, which was in equilibrium with radium to be absorbed by the cocktail.

Table 1.

Concentration factors of water samples.

NM = not measured

Water brand	Concentration ratio	
	1st measurement	2nd measurement
Vikos	34	65
Aura	32	89
Vittel	15	80
Zagori	41	61
Ioli	21	NM
Velouchi	23	NM
Ivi Loutraki	22	98
Samarina	48	81
Contrex	30	32
Joumerka	36	55
Dirfis	19	NM
Korpi	NM	29

The time of separation of radon from the sample was recorded for later use. The cocktail used does not mix with water and the vials were left aside in the dark for at least three hours in order to let the two phases to be well separated and also all the radon to be absorbed by the cocktail. Then the vials were counted using the alpha-beta discrimination technique.

All samples were measured for 200 min and the background was found to be 7.7 ± 0.1 cpm for the sum of alpha and beta counts. The total efficiency of the measured radium concentration using the above technique was calculated $0.27 \text{ Bq}\cdot\text{sec}\cdot\text{counts}^{-1}$. The minimum detectable activity for concentration ratio about 100 was calculated 4 mBq/L , for the above-mentioned values of background and efficiency.

3 Results and Discussion

The results of radium measurements are presented in Table 2. The average value of all measurements was $0.032 \pm 0.021 \text{ Bq/L } ^{226}\text{Ra}$ and the values varied from 0.006 to $0.071 \text{ Bq/L } ^{226}\text{Ra}$. The concentration values are in the same range with measurements performed by other authors elsewhere.

Table 2.

Measured radium concentrations of bottled water samples. The concentration is presented in Bq/L and error is 1σ .

Water brand	^{226}Ra concentration			
	1st measurement		2nd measurement	
	Bq/L	1σ	Bq/L	1σ
Vikos	0.019	0.004	0.018	0.001
Aura	0.024	0.004	0.017	0.002
Vittel	0.060	0.009	0.035	0.002
Zagori	0.028	0.003	0.006	0.003
Ioli	0.071	0.007	NM	
Velouchi	0.036	0.006	NM	
Ivi Loutraki	0.046	0.006	0.011	0.002
Samarina	0.019	0.003	0.010	0.001
Contrex	0.045	0.005	0.066	0.001
Joumerka	0.024	0.004	0.007	0.003
Dirfis	0.048	0.007	NM	
Korpi	NM		0.042	0.005

NM = not measured

Kralik et al [1], measured natural radioactivity in bottled water in Austria and found concentrations that varied from below the minimum detectable limit of their method (20 mBq/L), to a maximum concentration of 225 mBq/L in mineral waters. Bomben and Palacios [2], measured drinking water samples from municipal taps and private wells for natural radionuclides in Argentina, assessed that ^{226}Ra concentrations did not exceed 22 mBq/L. Measuring gross alpha and beta radioactivity in bottled waters sold in Mexico, Davila Rangel et al [3], found total alpha radioactivity that did not exceed 100 mBq/L in most of the water samples, except in six brands where their concentration in total alpha radioactivity was found up to 600 mBq/L. In a survey carried out in Taiwan, Yen-Chuan Kuo et al [4], assessed the activity concentrations and calculated the corresponding population dose from ^{226}Ra in food and drinking water. They measured the concentration of ^{226}Ra in mineral water samples and in drinking and underground waters and found levels that did not exceed 30 mBq/L. Martin Sanchez et al [5] and Manjon et al [6] have measured the concentration of ^{226}Ra in bottled mineral and drinking waters in Spain. The values varied from 60 mBq/L to 1860 mBq/L in the work of Martin Sanchez, and from 3 mBq/L to 267 mBq/L in the survey of Manjon.

The ^{226}Ra concentration levels assessed in the present work do not exceed the value of 100 mBq/L, which is the maximum acceptable concentration for ^{226}Ra in drinking water based on the U.S. Drinking Water Regulation [7]. The National Institute of Health in Italy [8], calculated the maximum acceptable activity of all radionuclides in the water so the annual committed dose would not exceed the total indicative dose, 0.1 mSv/y for the general public set in the European Council Directive 98/83 EC on the quality of water intended for human consumption [9]. The derived activity value for ^{226}Ra in the drinking water and for the highest risk age group, (less than one year old), was 0.085 Bq/L. It should be noted that the assessed values in the present work do not exceed this indicative maximum concentration.

4 Conclusions

Twenty bottled water samples from twelve domestic and foreign water brands were analyzed and the ^{226}Ra concentration was calculated. The concentrations did not exceed the maximum concentration 0.085 Bq/L for ^{226}Ra in drinking water. This value was calculated by the National Institute of Health in Italy for the highest risk age group, ages less than one year old, based on the total indicative dose 0.1 mSv/y as this was set in the Council Directive 98/83 EC on the quality of water intended for human consumption. The results show that some of the samples tend to reach this upper limit and need monitoring for a long period.

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