Analele Universităţii de Vest din Timişoara
Vol. XLVII, 2005
Seria Fizică

RADIUM CONCENTRATION IN SOME BOTTLED MINERAL WATERS FROM ROMANIA

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Abstract
The most important radionuclids from water and gas radioactivity point of view are: the uranium, the radium and the radon with it's short life descendents. This paperwork wants to determinate the radium concentration from 20 type of bottled mineral water commercially available in Romania. We used Luk-3A-a Czech device. Using this device we can determine the radium concentration directly, after we measure the radon resulted from balanced radium-226; the direct forerunner of radon is secular balanced with the radon-222. It has been determined in many cases—in different countries—that the ^{226}Ra concentration in mineral water is higher than that measured in tap water (Fernández, Cerretero, Liger, Canete, & Duenos, 1999; Marovic, Sencar, Franic, & Lokobauer,1996; Szerbin, Guczi, Stúr, Sztanyik, & Ugron, 1997). Enrichment of radium in drinking water and food enhances the ingestion dose due to its long physical and biological half-life (*T*: 1622 years and *T*b: 45 years). The elevated ingestion of ^{226}Ra might provide an annual internal dose near to the 0.1 mSv year^{-}1 reference level recommended by the World Health Organization (WHO) (*Guidelines for Drinking- Water Quality*, 1993). Therefore determination of the concentration of ^{226}Ra in mineral water has become important.
The results of the studies about radium in mineral water show that the radium concentration values measured are comparable with values measured and given in literature and they are under the maximum accepted values. According to the PHS estimations, the maximum level of contamination for radium (combined ^{226}Ra and ^{228}Ra) from the public water is 5 pCi/l.

Keywords: radium, radon, mineral water

1. Introduction

The risc of radium exposure and the radiation effects on living tissue have large perspective affinity in physics, medicine and geology.[1] The involvement of radium and especially of radon in population irradiation (60%) have lead to the conclusion that this element represents the second majour risc factor, after smoking, in pulmonary cancer. The first phase in preventing the exposure risk at radium and radon is the identification of the sources and then finding the best way to rectify the situation. Radium is a radioactive element, of silver white colour and it can't be found in natural form. It is formed by uranium and thorium desintegration in the environment. Radium only desintegrates by radioactive emision.

Due to the fact that uranium can be found throughout the terrestrial crust, radium's presence can be considered everywhere, it can be found in soil, water, rocks, plants, construction materials and in lower concentrations in food. Radium exists in many forms of different isotopes. There are four isotopes of radium that are naturally obtained and more isotopes produced by man through desintegation. The soil and rocks content in ^{226}Ra can be determined by γ spectrometry or by quantitative analysis of ^{222}Rn which is in secular equilibrium with ^{226}Ra in solid or liquid samples.

^{224}Ra can be determined in good precision by considering the secular equilibrium with its life long parent ^{232}Th by α spectrometry. This last method can also be used in ^{223}Ra determination. ^{226}Ra discovered by Marie Curie was used for medical purposes and it is up-to-dateness again as the parent of radon, the last one becoming a real threat in houses and work places when its concentration is too high. Nowadays the consumption of bottled mineral waters has become very popular. The average consumption of these is 0.36 l/day per person in Europe. A considerable segment of the population drinks almost only mineral water as drinking water, which is about 1l/day. As is known, some kind of mineral waters contain naturally occurring radionuclides in higher concentration than the usual drinking (tap) water. The WHO (1993) legislation concerning the drinking waters does not include the mineral waters.

In our work, the concentrations of ^{226}Ra, were determined in mineral waters available in Romania.

Radium in water

The concentration of ^{226}Ra and ^{228}Ra in the drinking water are usually low, but there are areas where the high concentration of Radium is due to the geological sources. The radioactivity levels in water are usually very low and they are measured in pCi/l. The surface water usually has a low Radium concentration, but the underground waters may contain higher concentrations due to the local geology. The drinking water from wells may contain ^{226}Ra and ^{228}Ra at higher levels than the standard ones. These high levels of Radium can be due to the rocks or sand in wells.

A research project lead by the USA Public Health Service (PHS) made a retrospective study of the homes of 111 communities from Illinois and Iowa, which were given water containing over 3pCi/l of Radium. A total of almost 908.000 inhaitants was the exposed population. Another study made on the public water from these communities showed that the averege level of Radium in water was 4,7 pCi/l.[2]

According to the PHS estimations, the maximum level of contamination for Radium (combined ^{226}Ra and ^{228}Ra) from the public water is 5 pCi/l.[3]

The measuring method of the Radium in water

Generally, ^{226}Ra, the direct forerunner of radon, is in secular equillibrium with the last one. We note with λ_{Ra} and λ_{Rn} the desintegration constants of radium, respectively radon, and with N_{Ra} and N_{Rn} the number of atoms of radium, respectively radon. In the case of secular equilibrium, for a time $t<<T_{1/2}(Ra)$, where $T_{1/2}(Ra)=1620$ years, the parent radium desintegration speed is actually constant, therefore we can approximate $e^{-\lambda_{Ra}t} \approx 1$, meaning $N_{Ra}=N_{Ra}(0)$ and the number of radon atoms is given by:

$$N_{Rn} \approx N_{Ra} \frac{\lambda_{Ra}}{\lambda_{Rn}} \left(1 - e^{-\lambda_{Rn}t}\right)$$

More, if $t \geq T_{1/2}(Rn)$, whre $T_{1/2}(Rn)=3,82$ days, then $e^{-\lambda_{Rn}t} \approx 0$, which leads to

$$N_{Rn} = N_{Ra} \frac{\lambda_{Ra}}{\lambda_{Rn}}$$

or $\lambda_{Rn} \cdot N_{Rn} = \lambda_{Ra} \cdot N_{Ra}$, which means that the activities of the parent (^{226}Ra) and the daughter (^{222}Rn) become equal.

The procedure used for radium measurement in water

The radon concentration is equal to the radium concentration, this happens after a a period of 28 days when radium can be considered in secular equilibrium with radon. In all the used calculations we will refer to radon concentration measurement which is actually the activity concentration of radium.

The procedure can be resumed to the following steps:

1. the water sample processing:
   - the water samples will be taken in 0,5 l vessels, the vessels will be fully filled and perfectly closed
2. the vessel will be brought to room temperature and the water temperature will be read using a thermometer

3. the water sample from the bottle, after reaching equilibrium is poured in the LUK-VR (scrubler) radon extraction device from water. After one minute of stirring, the scrubler is connected to the Lucas cell from the inside of the Luk-3A device.
4. The number of impulses given by the sample is recorded on a pre-established time interval. This number of impulses is chosen so that the static errors are around ±5%, which means that the total number of measured impulses is around 400. As an example we give the following values: for a nominal volume of 300 ml of water, evacuation volume of 510 ml and a temperature of 24^{o}C, results that α = 7,9 n/s. Thus at 20 Bq-l we get 2,5 impulses per second and the measuring period is 160 s.

2. Experimental method

For Radium determinations, we actually measure Radon using a LUK-3A Czech device. The special device for Radon extraction from water that accompanies the LUK 3A device is called LUK VR. The LUK VR device is a device built for the measurement of Radon (^{222}Rn) concentration in water.

The device components

The system is delivered as a complete unit with the following components:

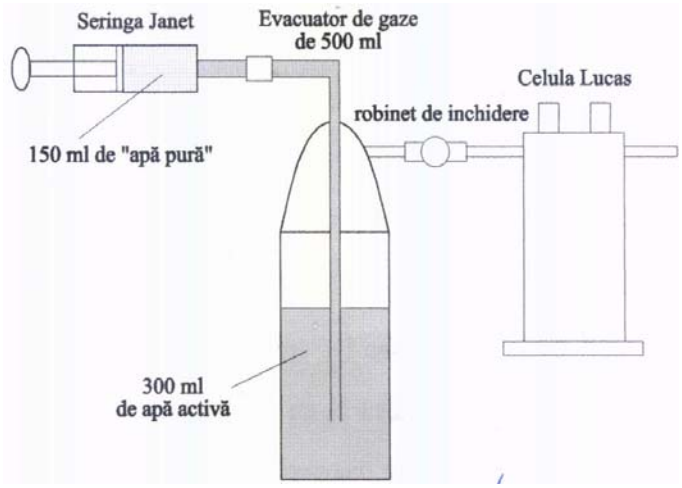
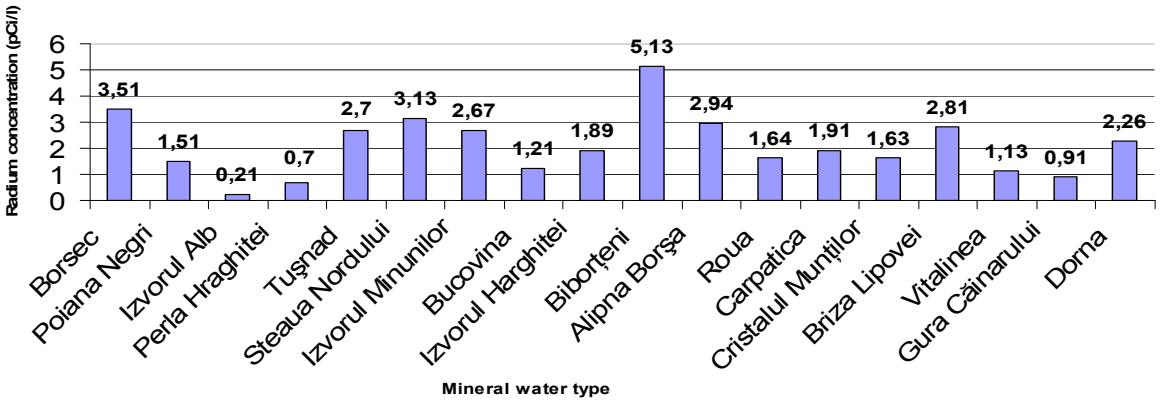


Figure 1. The principle scheme of the LUK-VR device:
- 500 ml gase evacuation
- tap with robber connectors
- termometer

3. Results:

The radium concentration was determined for 18 kinds of bottled mineral waters available in Romania:



The value of radium concentration obtained is between 0.21 pCi/l and 5.13pCi/l whith the maximum value is in Biborţeni mineral water and the minimum value in Izvorul Alb mineral water. In most cases the ^{226}Ra concentration is less than 5.pCi/l

The next table contains the types of mineral water whith the radium concentration. In most cases the ^{226}Ra concentration is less than 5.pCi/l.

Mineral water type	Nr. of counter /1000sec	Radium concentration (Bq/l)	Radium concentration (pCi/l)
Borsec	22,6	0,13	3,51
Poiana Negri	11,7	0,056	1,51
Izvorul Alb	11	0,008	0,21
Perla Hraghitei	13,3	0,026	0,70
Tuşnad	26,1	0,10	2,7
Steaua Nordului	36,42	0,116	3,13
Izvorul Minunilor	29,4	0,047	2,67
Bucovina	19,57	0,045	1,21
Izvorul Harghitei	23	0,07	1,89
Biborţeni	32,14	0,19	5,13
Alipna Borşa	25,14	0,109	2,94
Roua	16,85	0,061	1,64
Carpatica	21,14	0,071	1,91
Cristalul Munţilor	27,85	0,060	1,63
Briza Lipovei	20,15	0,1043	2,81
Vitalinea	17,13	0,042	1,13
Gura Căinarului	19,17	0,034	0,91
Dorna	21,36	0,0842	2,26

4. Conclusions

The results of the study on radium in mineral water show that the radium concentration values measured are comparable with values measured and given in literature and they are under the maximum accepted values. According to the PHS estimations, the maximum level of contamination for radium (combined ^{226}Ra and ^{228}Ra) from the public

water is 5 pCi/l. In recent years the consumption of mineral water in Romania has increased to a great extent. We determinated the ^{226}Ra concentrations of different types of mineral water. The value of radium concentration obtained is between 0.21 pCi/l and 5.13pCi/l whith the maximum value in Biborțeni mineral water and the minim value in Izvorul Alb mineral water. Due to the relatively long time in which Radium gets in equilibrium with Radon, the presented method requires a period of time of 28 days between the collecting point and the measurement point. [4]

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