MEASUREMENTS OF RADIUM LEVELS IN BOTTLED NATURAL SPRING WATER OF MARMARA REGION (TURKEY)

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Received September 14, 2009

Drinking water may contain varying levels of radioactivity. It is therefore important to determine the radium levels in drinking waters for public health and radiation protection. This paper presents results of ^{226}Ra concentrations in bottled spring waters. The concentration of radium has been assessed in the bottled natural spring water samples commercially available in Turkey. Measurements of ^{226}Ra concentrations in bottled natural spring water samples were carried out by AlphaGUARD, measuring the radon in equilibrium with radium. The radium concentration in these samples is found to be between 0.03 and 0.43 Bq·L^{-1}. These values were compared with the recommended safe limits for drinking water.

Key words: Radon, radium, spring water, radioactivity, AlphaGUARD.

1. INTRODUCTION

Water is the most important source for life and makes up 70 - 75% of total body weight. While 70% of the world's surface is covered by water, only 0.3 % of the total water resources on earth is drinkable and suitable for daily use. Human race provides their water from surface water and ground water. Ground water is more radioactive than surface water since it passes through rock and soil formations, dissolves many compounds, minerals and radioactive substances.

The radionuclides in drinking water are members of three natural radioactive series. These are the uranium series, the thorium series and the actinium series. The specific elements of concern are radium, radon and uranium. The nuclides of the uranium series which can be dangerous to health because of their presence in drinking water are ^{226}Ra and ^{222}Rn [1].

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Rom. Journ. Phys., Vol. 57, Nos. 7–8, P. 1204–1210, Bucharest, 2012

Radon is a naturally occurring radioactive element. It is a colourless, odourless and chemically inert gas and has no taste. It can only be measured with special equipment. Radon is also fairly soluble in water and organic solvents. Although reaction with other compounds is comparatively rare, it is not completely inert and forms stable molecules with highly electronegative ions. The radioactive decay of radon produces floating subatomic particles that can damage living cells. When radon is inhaled, 30% of the radon progeny comes in to contact with air passage in the lung and adhere to the surfaces. The ingestion of radon and radium from water can give rise to an additional exposure dose to the stomach and whole body.

In recent years, a great interest arose towards the natural radioactivity in drinking water. Duenas *et al.* [1] analysed concentrations of ^{226}Ra and ^{222}Rn in most of the bottled waters commercially available in Spain and they found that ^{222}Rn values range between 0.22 and 52 Bq·L^{-1}, with a geometric mean of 1.2 Bq·L^{-1}. Xinwei [2] measured radon concentration in drinking water from the municipal water supply system and private wells located in Baoji, China where the mean values of tap water and well water were found to be 12 Bq·L^{-1} with a maximum of 18 Bq·L^{-1} and 41 Bq·L^{-1} with a maximum of 127 Bq·L^{-1}, respectively. Kralik *et al.* [3] analyzed concentration levels of ^{226}Ra, ^{222}Rn and ^{210}Pb in domestic bottled waters commercially available in Austria where the concentrations for ^{222}Rn ranged between 0.12–18 Bq·L^{-1}, the geometric mean being 0.54 Bq·L^{-1} and concentrations up to 0.23 Bq·L^{-1}, with a geometric mean of 0.041 Bq·L^{-1} were found for ^{226}Ra. Chau and Michalec [4] presented activities of ^{238}U, ^{234}U, ^{228}Ra, ^{226}Ra, and ^{224}Ra as well as total α- and β-activities of 23 bottled spring, mineral and therapeutic waters produced and distributed in southern and central Poland where ^{226}Ra and ^{228}Ra concentrations ranged from 1 mBq·L^{-1} to above 500 mBq·L^{-1} while the measured concentrations of uranium isotopes vary from a few tenth mBq·L^{-1} to over dozen mBq·L^{-1}. Damla *et al.* [5] analyzed concentration activity of ^{222}Rn in water taken from eleven different sites in the Eastern Black Sea Region of Turkey and it was found that the activity of ^{222}Rn in tap waters changed from 5.79 Bq·L^{-1} to 18.46 Bq·L^{-1}, with a mean ^{222}Rn activity concentrations of 10.82 Bq·L^{-1}. Singh *et al*. [6] studied the uranium and radon concentrations in drinking water samples collected from different areas of the upper Siwaliks of Kala Amb, Nahan and Morni Hills of Haryana and Himachal Pradesh states, India. The radon concentration in these samples is found to vary from (0.87 ± 0.29) to (32.10±1.79) Bq·L^{-1}. Wallner and Steininger [7] measured the activity concentrations of the Ra isotopes, ^{226}Ra and ^{228}Ra, as well as of ^{222}Rn in Austrian tap waters and concentrations for ^{222}Rn ranged from 1.46 Bq·L^{-1} to 118.7 Bq·L^{-1}. Jia and Torri [8] have evaluated the radiological quality in some samples of drinking water collected in Italy and as far as the measured α or β radionuclides are concerned, the committed effective doses for all the analysed drinking water samples are in the range of 1.80–36.2 mSv·yr^{-1}. Desideri *et al.* [9] measured gross alpha and beta activities and ^{226}Ra, ^{238}U, ^{234}U, and ^{210}Po concentrations in some bottled mineral waters produced in Italy where the results revealed that the concentrations (mBq·L^{-1}) of ^{226}Ra, ^{238}U, ^{234}U and ^{210}Po ranged from <10.00 to 52.50, from <0.17 to 89.00, from <0.17 to 79.00, and from <0.04 to 21.01, respectively. Forte *et al.* [10] had measurements in the range of <0.003-0.186 Bq·kg^{-1}, natural radioactivity content in drinking waters to check the compliance with recent European and Italian rules. Moldovan *et al*. [11] measured ^{226}Ra concentration in 23 types of bottled mineral waters, commercially available in Romania and concentrations for ^{226}Ra ranged from 0.05 Bq·L^{-1} (Limit of Detection, LD) to 443.2 mBq·L^{-1}.

In this study, we have carried out a survey of ^{226}Ra concentrations in bottled waters that are commercially available. The aim of this study is to draw a general picture of the natural radioactivity of bottled water in Marmara Region of Turkey.

2. STUDIED SPRING WATERS

Twelve natural bottled waters were investigated in the framework of the presented study. The waters were purchased in the period from April to June 2008, from the markets in Bursa, Turkey and most of them are very popular brands in Turkey. All of the studied spring waters originated from Marmara Region which covers an area that has very complex geology and tectonics. Majority of the studied waters originated from Uludag and three of them originated from Hendek which is rich with spring sources, located in Sakarya in the Northwest of Turkey. Water samples' identification number and location of their springs can be found in Fig. 1.

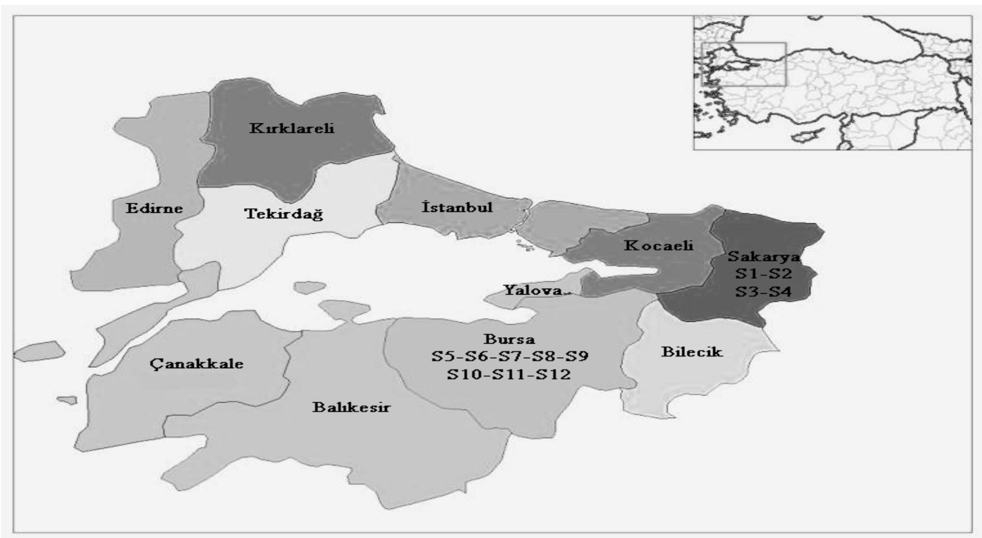

Fig. 1 – Locations of the springs where analysed bottled waters originated from.

All of the examined spring waters contain significant concentrations of magnesium and calcium. Air method enriched with ozone has been used for bottling process by the producers.

Besides different tectonic zones, Uludag is the highest mountain of Western Anatolia, located in Bursa. Bursa is a province full of ground waters, mineral waters and thermal waters since it is located in the juxtaposition of West Anatolian Extensional Zone and the southern branch of North Anatolian Fault Zone (NAFZ). Cold water springs have been discharged from different altitudes from Uludag to Bursa Plain [12]. This region consists of schist, marble, limestone and granite.

3. EXPERIMENTAL TECHNIQUES

An experimental technique described in detail by Kochowska *et al.* (2004) was employed. Radon concentration in water was measured using the professional radon monitor AlphaGUARD PQ 2000PRO (made by GENITRON, Germany). This is an ionisation chamber, designed for measuring radon in air, soil and water. For water measurements the additional equipment AquaKIT was used. Figure 2 shows a schematic view of the radon measurement system in water.

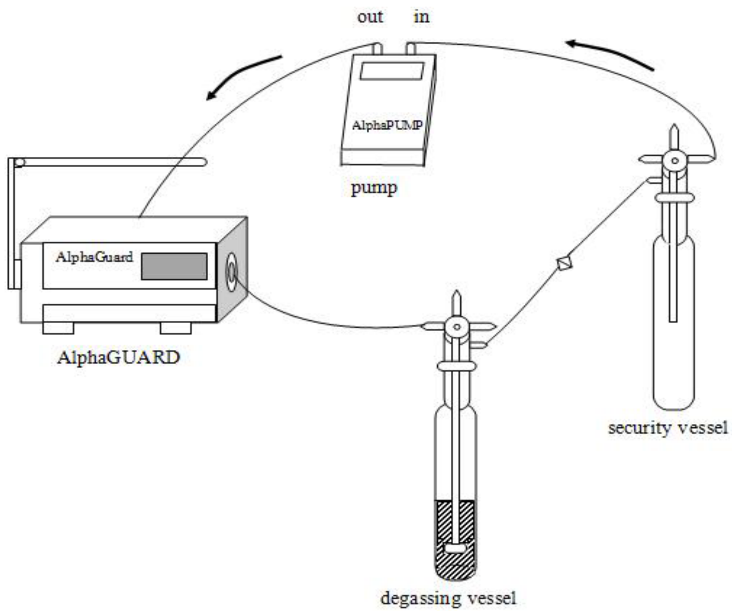

Fig. 2 – Schematic view of the experimental set-up.

In a close gas cycle, radon was expelled from water samples (placed in degassing vessel) using a pump. The security vessel was connected with the degassing vessel. All drops would deposit in it if they had got into the gas cycle during the degassing process. This way the stress of the water vapour was minimized for the radon monitor. The background of empty set-up was measured for a few minutes before every water-sample measurement. After that the water was injected into the degassing vessel, and the AlphaGUARD and AlphaPUMP were switched on. After 10 min the pump was switched off and the AlphaGUARD remained switched on for another 20 min, so the radon measurement was continued. This cycle was repeated three times in order to obtain a better precision. The AlphaGUARD monitor worked in a ‘flow’ mode and radon concentration was recorded every minute. The flow rate of the pump was 0.3 L/min. Radon concentration in the system was determined with the AlphaGUARD, whose ionisation chamber was also a part of gas cycle.

The determination of radon concentration in water samples was based on radon concentration indicated by the AlphaGUARD. This value was not the radon concentration in the water sample yet because radon driven out had been diluted in the air within the measurement set-up and a small part of the radon remained diluted in the watery phase. The following equation served to determine radon concentration in the measured water samples [13].

$$C_{water} = \frac{C_{air}\left(\frac{V_{system} - V_{sample}}{V_{sample}} + k\right) - C_0}{1000}$$

where

C_{water} - radon concentration in water sample ($Bq \cdot L^{-1}$)

C_{air} - radon concentration in the set-up after expelling radon from water ($Bq \cdot m^{-3}$);

C_0 - background ($Bq \cdot m^{-3}$);

V_{system} - interior volume of the measurement set-up (mL);

V_{sample} - volume of the water sample (mL); and

k - radon distribution coefficient.

Because our samples are “old” the initial radon is died and in these circumstances we measured the radon generated from radium during storage in the shops. After 23 days, 98,5% from radium-radon equilibrium is attained [11].

4. RESULTS AND DISCUSSION

Commercially available bottled spring waters were analyzed. The radium concentration in these samples was determined by using AlphaGUARD PQ 2000PRO radon gas analyser and the results are tabulated in Table 1.

Table 1

Results of radium measurements in bottled waters in Marmara Region of Turkey

Sample code	Origin of the water	^{226}Ra concentration ± standard error [Bq·L^{-1}]
S1	Hendek	0.03 ± 0.01
S2	Hendek	0.09 ± 0.02
S3	Hendek	0.07 ± 0.02
S4	Hendek	0.43 ± 0.09
S5	Uludag	0.23 ± 0.06
S6	Uludag	0.20 ± 0.03
S7	Uludag	0.03 ± 0.01
S8	Uludag	0.08 ± 0.02
S9	Uludag	0.10 ± 0.03
S10	Uludag	0.15 ± 0.03
S11	Uludag	0.11 ± 0.03
S12	Uludag	0.16 ± 0.03

The radium concentration in these samples varies from 0.03 to 0.43 Bq·L^{-1}. All of the radon concentration results are below 0.5 Bq·L^{-1} in our study.

5. CONCLUSIONS

Radium measurements in some bottled spring waters originating from Marmara Region were performed and the concentrations for ^{226}Ra ranged from 0.03–0.43 Bq·L^{-1}, the arithmetic mean being 0.14 Bq·L^{-1}. The recorded radium concentrations in all the water samples lie below the internationally recommended safe limit of 0.555 Bq·L^{-1} [14,15] and hence safe for drinking purposes.

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