Determination of ^{226}Ra concentration in bottled mineral water and assessment of effective doses, a survey in Turkey

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ABSTRACT

Background: There is a rapid increase worldwide in the consumption of mineral waters which may contain different level of radioactive elements, especially ^{226}Ra, in addition to varying amounts of beneficial salts. Therefore, a comprehensive study was planned and carried out in order to determine concentration of ^{226}Ra natural radionuclide in bottled mineral waters that commercially available in Marmara Region of Turkey. ***Materials and Methods:*** The method used for ^{226}Ra concentration analysis bases on the measurement of Radon (^{222}Rn) coming from ^{226}Ra dissolved in the water. The measurements were performed using RAD 7, a solid state α detector, with RAD H_2O accessory manufactured by DURRIDGE COMPANY Inc. ***Results:*** The ^{226}Ra concentration in mineral waters was found to vary from <0.074 to 0.625 Bql^{-1} with an average value of 0.267 Bql^{-1}.The committed effective doses due to ingestion of ^{226}Ra from the one year consumption of these waters were estimated to range from 10.8 to 90 μSvy^{-1}, from 9 to 75 μSvy^{-1} and from 3.15 to 26.25 μSvy^{-1}, for infants, children and adults, respectively. ***Conclusion:*** The results obtained in this study indicate that the committed effective doses are below the WHO (World Health Organization) recommended reference level of 100 μSvy^{-1}.

Keywords: *^{226}Ra activity, mineral water, committed effective dose.*

INTRODUCTION

In recent years, consumption of bottled mineral waters has increased in all countries and also in Turkey. As it is known, some kinds of mineral waters contain naturally occurring radionuclides in higher concentration than the usual drinking waters [(1)]. ^{226}Ra which is a radioactive member of ^{238}U series is the most commonly found radioisotopes in groundwater [(2)]. ^{226}Ra is found to be occurring in equilibrium with ^{238}U unless the environmental factors alter this ratio [(3)]. It is an α emitter with a long half-life of 1622 years. It follows the calcium metabolism in human body and deposited in bones. This may lead to enrichment of ^{222}Rn and its daughters, causing potential health implications and high degree of radio toxicity due to long exposure hazards [(4)].In fact people of all ages that consume the water face the risk of some health effects that may result from the significant accumulation of radium in their bones and other vulnerable or radiosensitive soft body tissues [(5)]. The activity concentration of ^{226}Ra in bottled mineral waters can vary considerably since it depends on the origin of the water [(6)]. ^{226}Ra and its daughters in water are responsible for a major fraction of the internal dose received by population from naturally occurring radionuclides [(1)]. Thus, monitoring of ^{226}Ra levels in bottled mineral waters is necessary for radiation protection purpose.

During the last decades, ^{226}Ra has been measured worldwide in the bottled mineral waters [1,2,4,7-28]. In view of these researches, the aim of the study is to determine ^{226}Ra concentration in bottled mineral waters that can be found in the markets in Marmara Region, Turkey. Based on the average concentration values and in the case of a regular consumer, internal doses due to ingestion of radium in mineral water were estimated. In addition to ^{226}Ra, physicochemical parameters of waters such as pH, EC and Eh were measured. Using the measured values of ^{226}Ra, pH, EC, Eh and chemical content of mineral waters presented on label, the impact of these parameters on radium concentration were evaluated.

MATERIALS AND METHODS

Study area

The Marmara Region, with a surface area of 67.000 km^2, is the smallest but most densely populated of the seven geographical regions of Turkey (figure 1). It represents approximately 8.6% of the Turkish national territory and about 30% of its population. Its name derives from the Sea of Marmara [29].

The Marmara Region is surrounded by the Black Sea and Central Anatolia Regions to the east, the Aegean Region to the south and Greece and Bulgaria to the northwest. Edirne, Kirklareli, Tekirdag, Istanbul, Kocaeli, Yalova, Sakarya, Bilecik, Bursa, Balikesir and Canakkale Provinces are located within the borders of the Marmara Regions [30].

It is Turkey's main industrial region and the most developed area is the Istanbul-Bursa-Izmit triangle. Istanbul, which has been an important trade center since ancient times, is located on the intercontinental transport routes and makes the region superior throughout the country. Processed food items, textiles, ready-to -wear clothing, cement, paper, petrochemical products, durable household items, ships and yachts are among the main industrial goods produced in the region [30].

Measurement Technique

The most frequently produced and consumed total of 15 bottled mineral water samples were collected from the markets located in different part of Marmara Region. In the laboratory, the samples were filled in 250 ml vials using special adaptors. These vials were sealed and stored for 1 month prior to the measurement after radon in the vials was removed. It is well known that after a period of one month, radium is considered in equilibrium with radon [1]. At the end of this period, the growing ^{222}Rn which was equal to

Figure1. Map of the study area.

^{226}Ra activity was measured using RAD 7 (Durridge Co., USA), an electronic radon detector, connected to a RAD H_2O (radon in water) accessory [31, 32]. Briefly, this device offers an accurate measurement, faster reading, it is portable and eliminates the need for noxious chemicals [31]. The schematic diagram of RAD 7 with RAD H_2O accessory is presented in figure 2. In all the used calculation we will refer to radon concentration measurement which is actually the radium activity concentration.

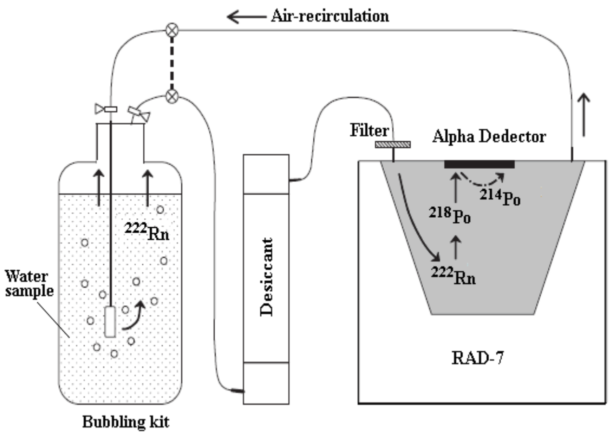

Figure 2. Schematic diagram of RAD H2O assembly.

The RAD 7's internal sample cell is a 0.7 litter hemisphere, coated on the inside with an electrical conductor. A solid-state, Ion-implanted, Planar, Silicon alpha detector is at the centre of the hemisphere. The high voltage power circuit charges the inside conductor to a potential of 2000 to 2500 volts, relative to the detector, creating an electric field throughout the volume of the cell. The electric field propels positively charged particles onto the detector [27]. Inside the RAD7 chamber, there is a solid state alpha detector at the centre of the hemisphere equipped with a nuclear electronic instrument. This solid state alpha detector has semiconductor material that converts alpha radiation directly to electrical pulses [31,33]. One important advantage of solid state devices is ruggedness. Another advantage is the ability to electronically determine the energy of each alpha particle [31]. The RAD7 can compute the energy of each alpha particle which can partially distinguish sample pulse from noise, and makes it possible to recognize to which isotopes (^{218}Po, ^{214}Po, etc.) a radioactive reading belongs [33]. So you can immediately distinguish old radon from new radon, radon from thoron, and signal from noise [31].

For the measurements of water samples, there are two available protocols (Wat-40 and Wat-250) depending on the size of vial (40 or 250 ml) on the RAD 7. It is very important to select the correct protocol before radon measurement, since it controls the pumping and counting cycle, as well as the calculation depending on the size of sample vial that is being used. In the manual Wat-250 protocol is proposed as the basic protocol for low activity measurements while Wat-40 protocol is suggested for high activities [31]. We selected Wat-250 protocol in the present study since the activity concentration of ^{226}Ra is expected to be low.

For the measurement of ^{226}Ra via growing ^{222}Rn, the system is purged by using the desiccant tube for 15 minutes in open circuit. In this procedure, the internal pump of RAD 7 begins running and the high voltage circuit turns off in order to clear the sample chamber of radon gas and daughters as quickly as possible. It must be provided clean, desiccated, radon-free air to the inlet in order to push out any radon that was previously sampled. Laboratory air is usually adequate for this purpose [31]. The RAD7 radon detector performs measurements of radon activity in water by measuring its activity in air after the aeration process. Therefore, for measurements of low activity samples, one of the most important questions is the radon concentration in the system air before aeration. It is the so called background. As the number of measurements of the laboratory air was performed, the value of the background reaches hundreds of Bqm^{-3} and the error of its determination with Wat-250 protocol, proposed in the manual as the basic protocol for low activity measurements, also reaches hundreds of Bqm^{-3}. With such a level of background, and its error, it is not possible to perform the measurements of low activity (radium) of water samples [32]. Considering the importance of background in the measurement of radium activity, charcoal adsorption tube was used to reduce background. Charcoal adsorption tube consists of a small column containing 15 grams of activated carbon which can remove up to 98% of the remaining radon from the RAD H_20 system when connected in a closed loop (31). For this purpose, charcoal adsorption tube was added the circuit in a closed loop and the system was purged further. This allowed a decrease of the background level up to tens of Bqm^{-3} with Wat-250 protocol.

After that, as can be seen from figure 2 the inlet of the sampling bottle is connected to the outlet of RAD 7 and the outlet of the sampling bottle passing through a desiccant is connected to the inlet of RAD 7. There is an air filter at the entrance of RAD 7 to keep out particles and attached radon daughters. Wat-250 protocol was started. The internal air pump of RAD 7 runs for five minutes, re-circulating a closed air-loop through the water sample, purging radon from the water into the RAD 7. During the 5 min of aeration, more than 95% of the available radon is removed from the water and the components automatically perform everything required to determine the radon concentration in the water. The system waits a further five minutes for reaching a state of equilibrium. After reaching equilibrium between water, air, and radon progeny attached to the PIPS detector, the system start counting the radon activity concentration of the sample. After five minutes it prints out a short-form report. The same thing happens again five minutes later, and for two more five-minute periods after that. At the end of the run (30 minutes after the start), the RAD 7 prints out a summary, showing the average radon reading from the four cycles counted, a bar chart of the four readings, and a cumulative spectrum. Thus, radon gas is collected through the energy specific windows which eliminate interference and maintain very low backgrounds and later counted for the radon concentration (31). ^{222}Radon activities are then expressed with uncertainty down to under ± 5% (34). All data, except the spectrum, is also stored in memory, and may be printed or downloaded to a PC at any time.

The background of the system was determined from the measurement of a radon free water sample. The distilled water sample stand closed and undisturbed for 4 weeks was used as radon free water sample. The minimum detectible activity (MDA) was found to be 0.074 Bql^{-1}.

RESULTS AND DISCUSSION

The most commonly consumed total of 15 bottled mineral waters was collected from the Marmara Region of Turkey. The results of the measured ^{226}Ra activity concentration and the chemical content of these waters are shown in table 1. The chemical contents were taken from labels on the bottles. The physical properties (pH, Eh and EC) of water samples were measured in laboratory.

As seen from table 1 the radium concentration of bottled mineral waters range from <0.074 to 0.625 Bql^{-1}. These values of ^{226}Ra concentrations are well below the recommended WHO guideline activity concentrations of 1 Bql^{-1} for drinking water (35). The studied bottled mineral waters which arise from different regions of Turkey generally have ^{226}Ra activity concentration less than 0.60 Bql^{-1}, only one sample exceeds this level. The highest values of ^{226}Ra activity of 0.625 Bql^{-1}, 0.589 $Bql^{-1}$and 0.570 Bql^{-1} for S1, S13 and S5, respectively may be related to geological characterisation of the area where water source origins. In general the high ^{226}Ra concentrations relate to sandstone aquifers (28, 36).

The comparison of measured ^{226}Ra concentrations with the results of several studies from other countries is presented in table 2.

As can be seen from table 2, the average concentrations of ^{226}Ra in analysed bottled mineral waters are of the same order as those reported for mineral waters from Poland (23), from Yugoslavia (23), from Spain (2) and from Romania (1). The measured activities of ^{226}Ra in the present study are considerably lower than those reported from Portugal (9), Hungary (7,8,11) and Spain (2). However, the levels of ^{226}Ra in the investigated water samples are slightly higher

Table 1. ^{226}Ra concentrations and the chemical contents of mineral waters.

Sample ID	pH	EC (mS)	Eh (mV)	K^+ (mgl^{-1})	Na^+ (mgl^{-1})	CA^{2+} (mgl^{-1})	Mg^{2+} (mgl^{-1})	Cl^- (mgl^{-1})	Total (mgl^{-1})	^{226}Ra (Bql^{-1})
S1	6.34	2.28	+ 64.9	13.4	845	52.4	17.2	141.5	3567.5	0.625±0.08
S2	5.84	0.89	+ 94	8.2	40.04	289	36.2	40.7	1853	0.418±0.08
S3	6.04	1.10	+ 81.5	53.6	425	326	111.4	166	2724	0.112±0.01
S4	5.68	1.19	+ 101.9	10.7	72.59	116.8	102.2	32.4	1417.4	<0.074
S5	5.73	1.52	+ 99.4	28.9	189.8	223.6	81.4	95.1	2705	0.570±0.08
S6	5.36	0.83	+ 120.5	13.4	356.76	35.67	10.6	189.3	1707.2	0.210±0.07
S7	6.32	2.07	+ 65.7	30.7	434.2	146.8	66.1	62	2737.6	<0.074
S8	5.71	1.45	+ 99.3	17	38.4	234	55	10.2	1264.9	0.285±0.05
S9	5.71	0.63	+ 99.4	n.a.	11.8	155	6.8	25	679.7	0.209±0.06
S10	5.40	1.25	+118.2	2	34	179	52	55	948	<0.074
S11	6.19	1.70	+ 73.1	1.6	107.7	163	91.5	26.4	1715	0.075±0.01
S12	5.85	1.78	+ 91.8	63	265.2	235.5	108.4	25.8	2767.5	0.401±0.04
S13	6.03	1.99	+ 82.2	8	752.3	202.9	238.5	74.9	3600.4	0.589±0.08
S14	6.11	2.69	+ 78.9	39.6	1014	28.5	27.3	257.9	3522.4	0.301±0.03
S15	5.91	1.82	+ 89.1	5.4	22.2	393.2	28.8	5.1	3920.3	0.210±0.04

n.a.: non-available

Table 2. The comparison of ^{226}Ra activities for bottled waters with the results of other studies.

^{226}Rn Activity (Bql^{-1})	Study	Location
0.002-0.6	Duenas *et al.* (2)	Spain
0.003-2.185	Bettencourt *et al.* (9)	Portugal
0.007-0.614	Kobal *et al.* (23)	Yugoslavia
0.012-0.042	Amrani (12)	Algeria
<0.002-0.23	Kralik *et al.* (25)	Austria
<0.01-0.053	Desideri *et al.* (13)	Italy
0.06-0.443	Moldovan *et al.* (1)	Romania
0.013-2.89	Baradacs *et al.* (7)	Hungary
0.004-0.211	Wallner and Jabbar (26)	Austria
0.043-2.94	Kovacs *et al.* (8)	Hungary
ND-1.86	Sanchez *et al.* (10)	Spain
0.106-3.169	Somlai *et al.* (11)	Hungary
0.008-0.015	Fatima *et al.* (20)	Pakistan
<0.4-1.77	Fredj *et al.* (19)	Tunisia
0.0008-0.525	Chau *et al.* (17)	Poland
<0.074-0.625	***This Study***	***Turkey***

than those reported from Algeria (12), Austria (26), Tunisia (19), Pakistan (20) and Italy (13).

The radium concentration of investigated mineral waters varies over a wide range. One reason for this can be the geological differences of the aquifers and also the depth of the well (17). Another reason can be the different chemical characteristic of water (24). In order to evaluate this, the relation between the chemical characteristics and the measured ^{226}Ra activities of water were investigated (figure 3).

It was reported by many researcher that the activity concentrations of mineral waters can be reach higher values with increasing mineralization (26, 10). This behaviour was observed clearly in the investigated water samples (the correlation coefficient is 0.36). We found that the measured ^{226}Ra concentrations in the investigated water samples increases with the total mineralization (figure 3).

Several studies have revealed a negative correlation between pH and elevated ^{226}Ra concentration. On the other hand, there are some studies exhibits the positive correlations between pH and ^{226}Ra activity (37). In the present study it was observed that the concentrations of ^{226}Ra increasing with pH. Although there is a positive correlation between ^{226}Ra concentrations and pH, the link between them is generally poor (the correlation coefficient is 0.40). Several studies have revealed a positive correlation between ^{226}Ra and chloride (Cl) content (38-40) as well as sodium (Na) (39). Although a positive correlation is observed between chloride content and ^{226}Ra activity of water samples, correlation coefficient is very weak (r=0.14). However, there is a relatively strong correlation (r=0.50) between Sodium (Na) content and ^{226}Ra activities of analysed waters.

A reverse correlation was registered between ^{226}Ra activity and redox potential (E_h) (r=-0.40). It is known that low redox potential conditions decrease the adsorption of radium onto aquifer surfaces by reducing the stability field of sulphate minerals [41]. As a result, the solubility of ^{226}Ra in water increases with reducing redox potential.

Also, there appears to be a positive correlation between the EC and ^{226}Ra concentrations in mineral waters (R=0.45). The elevated ^{226}Ra concentrations with an increase of the electrical conductivity has been reported for bottled drinking waters [6,22].

Dose estimations

The effective dose to an individual from intake of ^{226}Ra radionuclide via ingestion of bottled mineral water is calculated using the relation [42]:

$$D_{ing.} = C_{Ra} \times I_a \times D_f$$

where C_{Ra} is the concentration of ^{226}Ra (Bql^{-1}), I_a (l.y^{-1}) is yearly intake of drinking water and D_f is dose conversion factor. For the age dependent dose calculations the dose conversion factor (D_f) used in the calculations are given in table 3.

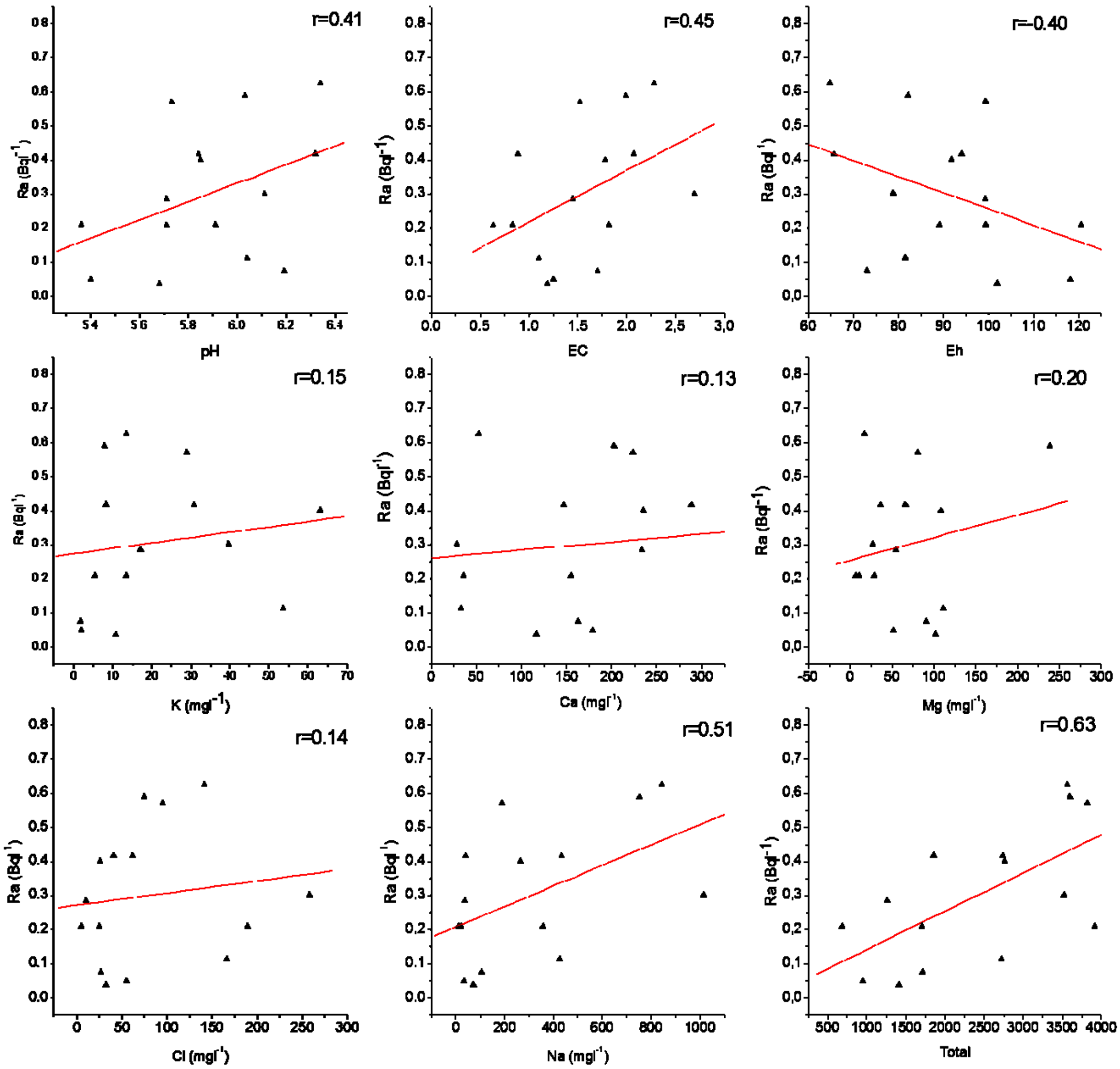

Figure 3. The correlation between ^{226}Ra activity and the chemical content of mineral waters.

Table 3. Dose conversion factors for ingestion of ^{226}Ra.

Age Group	D_f (in µSvBq^{-1}) for Ingestion of ^{226}Ra [42]
Babies (1 year)	0.96
Children (13-17 year)	0.80
Adults (>17 year)	0.28

The daily intake values of bottled mineral waters were estimated considering the dietary habits of the Turkish population and statistical data on mineral water consumption. Although the consumption of bottled mineral water has been increasing every year in Turkey, the annual consumption is estimated at 6-7 litres per person which is well below the other European countries averages of 150ly^{-1} per person [43]. However, in calculations consumption rate of mineral water was taken as 150ly^{-1} per person living in Turkey. Figure 4 shows the results of dose estimations for ^{226}Ra due to ingestion of bottled mineral waters.

The results presented in figure 4 shows that the committed effective doses due to ingestion of ^{226}Ra in bottled mineral waters varied from 10.8 to 90 µSvy^{-1}, 9 to 75 µSvy^{-1} and 3.15 to 26.25 µSvy^{-1}, for infants, children and adults, respectively. As can be seen from figure 4, the committed effective doses from all the bottled waters are well below the WHO's recommended value of 100µSvy^{-1} for infants, children and adults [35].

The results obtained in this study were compared with the data from the studies carried out in Turkey and other nations (table 4). The comparison of our results with similar works shows that the observed effective dose of ^{226}Ra (3.15-90 µSvy^{-1}) was lower than those reported from Tunisia [19], from Spain [2], from Portugal [9], from Romania [1], from Hungary [11] and from Austria [25]. Only in two studies (from Pakistan [20] and from Algeria [12]) reported doses are lower than our results.

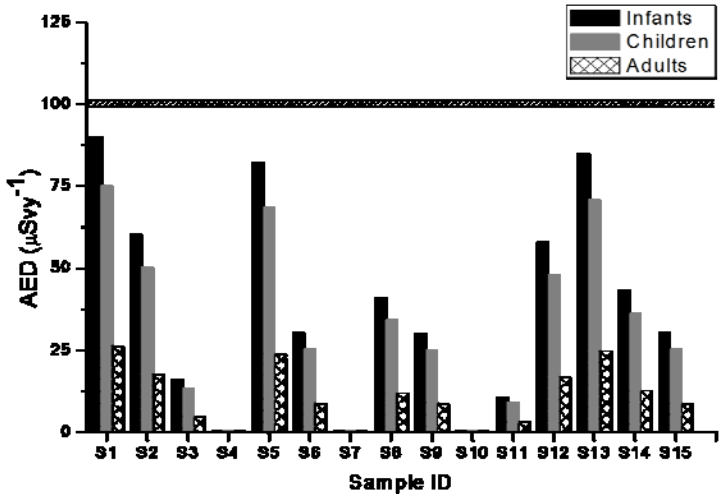

Figure 4. The committed effective doses due to ingestion of ^{226}Ra. The dashed line of 100µSvy^{-1} represents WHO reference value for drinking water.

Table 4. The comparison of annual effective doses with the results of other studies.

^{226}Ra (µSvy^{-1})	Study	Country
1.5-2.9	Fatima et al. [20]	Pakistan
180-2080	Fredj et al. [19]	Tunisia
1-220	Kralik et al. [25]	Austria
1.84-6.44	Amrani [12]	Algeria
2-102	Duenas et al. [2]	Spain
9-1600	Somlai et al. [11]	Hungary
5-200	Bettencourt et al. [9]	Portugal
9-242	Moldovan et al. [1]	Romania
3.15-90	***This Study***	***Turkey***

CONCLUSION

The present study has been the first one in which bottled mineral waters consumed in Marmara Region of Turkey were analysed for ^{226}Ra activity concentrations. The results of the study showed that concentrations of ^{226}Ra in bottled mineral water vary from <0.074 to 0.625 Bql^{-1} with an average value of 0.267 Bql^{-1}. The annual effective doses for infants, children and adults were calculated from the measured activities of ^{226}Ra, considered the UNSCEAR recommended age-dependent dose coefficients and assumed 150 l for yearly consumption. The annual doses for all age groups are found to be significantly below the 100 μSvy^{-1} reference dose level.

In addition to ^{226}Ra other physicochemical parameters of water like pH, EC and Eh were also measured in order to evaluate the impact of these parameters on radium concentration. On the basis of the measured data for ^{226}Ra, pH, EC and Eh and the ion content presented on label of bottles, the correlations between the ^{226}Ra activities and chemical contents in the analysed waters, as well as their mineralization were investigated bottled waters.

In conclusion, the study shows that bottled mineral waters consumed in Marmara Region have good radiological quality according to WHO guidelines.

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