

^{238}U , ^{234}U and ^{226}Ra concentrations in mineral waters and their contribution to the annual committed effective dose in Turkey

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Abstract Activity concentrations of ^{234}U , ^{238}U and ^{226}Ra in mineral waters were determined on the basis of nine water bottling facilities using alpha particle spectrometry. The mineral water samples were collected from three geographic regions of Turkey. The radiochemical separation used in the uranium analysis is based on the isolation of uranium radioisotopes from other radionuclides such as Th, Am, Pu and Np using UTEVA resin. Alpha sources were prepared using electrodeposition method. The activity concentration of ^{226}Ra was determined after deposition on a membrane using BaSO_4 co-precipitation method. The activity concentrations (mBq L^{-1}) of ^{226}Ra , ^{238}U and ^{234}U ranged from <0.56 to 165, from <0.42 to 439 and from <0.42 to 464, respectively. The measured activity concentrations were used for the calculation of the average total annual effective ingestion doses for children and adults. The committed effective doses were calculated for three different scenarios according to mineral water consumption rate. In the most extreme scenario (for age group 12–17), all water samples except MW1 and MW2 cause annual committed effective doses below the reference level ($0.1 \text{ mSv year}^{-1}$) recommended by World Health Organization (WHO).

Keywords Mineral water · Uranium · Radium · Alpha spectrometry · Dose assessment

Introduction

Humans are exposed to radiation from natural or man-made sources through inhalation, ingestion by food and water consumption. According to The United Nations Scientific Committee on the Effects of Atomic Radiation, exposure to natural radiation sources contributes more than 98 % of the radiation dose to the population and the contribution from nuclear power production and nuclear weapon testing is very small. The global average human exposure from natural sources is $2.4 \text{ mSv year}^{-1}$ [1].

The quality of water needs to be strictly and regularly controlled due to its importance in human life. Assessment of radiological content is a part of this control to ensure lower levels of health risks posed by radioactivity in drinking waters (natural or mineral). The reference level of the effective dose due to consumption of drinking water is $0.1 \text{ mSv year}^{-1}$ in the European Drinking Water Directive 98/83/EC [2].

This value is within the recommendations given in the second edition of WHO guidelines [3]. The effective dose value excludes the dose received from ^3H , ^{40}K , ^{222}Rn and radon decay products. In the second edition of WHO guidelines, the recommended screening levels for drinking water are 0.1 Bq L^{-1} for gross alpha activity and 1 Bq L^{-1} for gross beta activity. The total alpha radioactivity value was raised to 0.5 Bq L^{-1} in the third edition of WHO guidelines [4], while the recommended screening level for gross beta activity remained the same as in the previous edition. Below these recommended screening levels, drinking water is acceptable for human consumption and no further action is required. Otherwise, investigation of the concentrations of individual radionuclides and assessment of the dose exposure is required. In any case, it is necessary to investigate the activity concentrations of

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individual radionuclides for accurate dose evaluation since the dose coefficients are related to specific radionuclides.

The current directives and recommendations do not cover mineral waters. However, there is an increasing tendency to replace tap water for human consumption with mineral water. Mineral water can be defined as water containing various minerals which are natural compounds formed through geological processes or other dissolved substances that alter its taste or give it therapeutic properties [5]. Mineral waters stemming from very deep aquifers usually show higher natural radionuclide content compared to surface water and water obtained from shallower wells [6]. Thus, higher levels of mineral water consumption may lead to increased doses from natural radiation sources.

Alpha emitters, particularly uranium and radium compounds are very toxic for human health [7]. Uranium exposure causes both radiological and chemical toxicity in the body. The primary chemical effect of uranium in the body is its accumulation in the kidneys causing kidney failure [8]. For this reason, uranium concentration in drinking water is controlled by several countries even when the total annual committed effective dose from alpha emitting radionuclides is less than 0.1 mSv a^{-1} . It is also covered by the WHO Guidelines, although its contribution to the internal dose is minor due to its low dose conversion factor. WHO set the most stringent limitation with $2 \mu\text{g L}^{-1}$ in its 1998 report [3], but later changed this limit to $15 \mu\text{g L}^{-1}$ [4]; and finally to $30 \mu\text{g L}^{-1}$ [9]. USA [10] set the limit to $30 \mu\text{g L}^{-1}$ and Germany set a maximum level of $2 \mu\text{g L}^{-1}$ for mineral water considered suitable for infants [11].

^{226}Ra is another frequently monitored alpha emitting naturally occurring radionuclide in drinking waters due to its long half-life (about 1,600 years) and radiological effects on human health [12]. ^{226}Ra is mainly accumulated in the bones when ingested, thus increasing the internal radiation dose of individuals [13]. Therefore, the activity concentration of ^{226}Ra in drinking waters has to be determined in order to evaluate its potential contribution to the internal dose as well.

Turkey lies on the “Alpine–Himalayan orogenic belt” and is a region of great tectonic complexity. Continuing tectonic activities have resulted in the formation of various geological structure and numerous cold and hot water springs [14]. This leads to a profusion of natural mineral waters all throughout Turkey. Western Anatolia is especially significant in terms of mineral waters since most of the bottling facilities are located in this region [15]. Those mineral waters are consumed by local people and transferred to the other parts of country. Consumption of mineral waters in Turkey is lower than in other European countries [16]. The average mineral water consumed per

person in Turkey is 7.6 L per year where this value is 150 L in European countries [17, 18]. However the consumption rate has increased 8.2 % annually since 1997 and expected to increase in the future [15].

Determination of naturally occurring radionuclides such as radium and uranium in mineral waters and assessment of effective annual doses from mineral water consumption is useful as a direct input to public health studies. Several studies were performed previously on radioactivity in various water samples (tap water, spring water, ground water, surface water, geothermal water) collected from different regions of Turkey [19–24]. However the available literature indicates that there are only a few studies on the radioactivity in Turkish mineral waters [5, 25]. Taşkin et al. reported the radiological assessment of bottled mineral waters collected from various natural sources in Turkey [25]. This study only refers to gross alpha/beta activities. Kobya et al. reported the chemical composition and activity concentrations of ^{226}Ra , ^{232}Th , ^{137}Cs , ^{40}K , gross alpha and gross beta of mineral water samples collected from Eastern Black Sea Region of Turkey [5]. To the best of our knowledge, the detailed information on the activity concentration of naturally occurring alpha emitter nuclides (^{226}Ra , ^{234}U , ^{238}U) of mineral waters in Turkey is not available.

In this study we investigated the activity concentrations of ^{234}U , ^{238}U and ^{226}Ra in mineral water samples selected from three different regions of Turkey. The choices of these three regions were based on considerations regarding elevated levels of gross alpha activities [26] and presence of the majority of the bottling facilities in these regions. The activity concentrations of these radionuclides were determined by alpha particle spectrometry. The potential committed effective doses for three different age groups were calculated for three different mineral water consumption rates using the measured activity concentrations of investigated radionuclides and comparing these values to the maximum total indicative dose of $0.1 \text{ mSv year}^{-1}$ given in the EC Drinking Water Directive 98/83/EC [2].

Materials and methods

Sampling

Nine mineral water samples taken from three different regions of Turkey were investigated in this study. Mineral water samples (MW) were collected from different bottling facilities; MW1–5 from Marmara region, MW6, MW7 and MW9 from Aegean region and MW8 from Black Sea Region. Samples were collected in 5 L polyethylene containers between the filtering and bottling steps in the facilities. Containers were washed with HNO_3 and rinsed



Fig. 1 Locations of the mineral water sampling sites in this work (modified from d-maps.com [28])

with the water sample before sampling. Samples were acidified to pH 2 by adding HNO_3 to avoid precipitation and adsorption on the walls of the sample bottle [10, 27]. The locations of the water sources are shown in the map of Turkey in Fig. 1.

Reagents and tracers

All reagents used in this study were of analytical grade and used without further purification. HCl , $\text{Pb}(\text{NO}_3)_2$, $\text{BaCl}_2 \cdot 2\text{H}_2\text{O}$, Na_2SO_4 , EDTA, NaOH , CH_3COOH , $(\text{NH}_4)_2\text{HPO}_4$, $\text{Ca}(\text{NO}_3)_2$, $\text{Al}(\text{NO}_3)_3$, KOH , NaHSO_4 , phenolphthalein, ethanol, acetone and pH indicator solution (pH 0–5) were supplied from Merck. HNO_3 , H_2SO_4 , NH_4OH , ascorbic acid were supplied from Sigma-Aldrich and $(\text{NH}_4)_2\text{C}_2\text{O}_4 \cdot \text{H}_2\text{O}$ was from Fluka. 0.45 micron polypropylene filter was purchased from PANI. Prepacked columns of UTEVA resin (100–150 μm particle size) were supplied from Eichrom Technologies, Inc.

^{133}Ba tracer solution with an activity concentration of $3.715 \text{ kBq g}^{-1} \pm 1.0 \%$ ($k = 1$) was purchased from Isotope Products Lab. SRM 1443-59-1. The tracer solution was diluted with 0.1 M HCl for reducing the activity concentration to approximately 30 Bq mL^{-1} . ^{133}Ba standard disc used for determining chemical recovery of radium analysis was prepared by using drop deposition method on a polypropylene filter. The ^{232}U tracer solution with an activity concentration of $38.22 \text{ kBq g}^{-1} \pm 0.8 \%$

($k = 1$) was obtained from NIST SRM 4324B. The tracer solution was diluted with 0.1 M HCl for reducing the activity concentration approximately to 0.1 Bq mL^{-1} .

Radiochemical procedure

The water samples collected before the bottling step in the facilities have not been filtered prior to radiochemical separation since no suspended substances were observed in the samples.

Determination of ^{238}U and ^{234}U

A calcium phosphate precipitation technique was used to concentrate and remove actinides from water samples [29]. The sample container was vigorously shaken prior to sub-sampling. 0.1 Bq of ^{232}U tracer was added to 1 L of water sample at the beginning of the sample preparation stage. 0.5 mL of 1.25 M $\text{Ca}(\text{NO}_3)_2$ was added to the sample and the solution was heated until boiling. 2–3 drops of phenolphthalein indicator and $200 \mu\text{L}$ of 3.2 M $(\text{NH}_4)_2\text{HPO}_4$ solution were added to the sample. The uranium was co-precipitated with $\text{Ca}_3(\text{PO}_4)_2$ by addition of NH_4OH . The precipitate was allowed to settle overnight. The supernatant was discarded. The precipitate was centrifuged 5 min at 3,000 rpm, washed with distilled water and centrifuged until no ammonia odour persisted. The precipitate was dissolved in 5 mL HNO_3 and evaporated to dryness. The

residue was dissolved with 10 mL of 3 M HNO_3 –1 M $\text{Al}(\text{NO}_3)_3$. The uranium in this solution was purified and isolated radiochemically using chromatographic resin. The radiochemical separation is based on the isolation of uranium radioisotopes from other radionuclides such as Th, Am, Pu and Np in the sample using Eichrom resins prior to measurement by alpha particle spectrometry [30]. UTEVA resin was preconditioned with 5 mL of 3 M HNO_3 . Sample dissolved in 3 M HNO_3 –1 M $\text{Al}(\text{NO}_3)_3$ was passed through the resin, followed by washing the resin with 10 mL of 3 M HNO_3 . 5 mL of 9 M HCl was added to convert the resin to chloride system. Interfering elements were separated by addition 20 mL of 0.5 M $\text{H}_2\text{C}_2\text{O}_4$ /5 M HCl. Any neptunium, plutonium or thorium was removed from the resin in this step. Uranium was stripped from the column with 15 mL 1 M HCl and evaporated to dryness.

The sources for alpha spectrometric measurement were prepared by electrodeposition of uranium on 2 mm diameter stainless steel discs. Evaporated actinide solution was dissolved with 2.5 mL 5 wt% NaHSO_4 , 2 mL distilled water and 5 mL 15 wt% Na_2SO_4 . 1 mL of ammonium oxalate was added to the electrolysis cell. Electrodeposition was carried out at 0.75 A for 90 min and ended after addition of 2 mL 25 wt% KOH. The disc was removed from the cell and rinsed with NH_4OH , ethanol and acetone sequentially. The disc was dried 5 min at 200 °C and measured immediately by alpha particle spectrometer. The alpha sources were counted for a period of 3–4 days. The minimum detectable activity (MDA) was 0.42 mBq L^{-1} for both ^{238}U and ^{234}U . Chemical recoveries were typically 70–80 %.

Determination of ^{226}Ra

The radiochemical separation procedure is based on co-precipitation of $\text{Pb}(\text{Ra})(\text{Ba})\text{SO}_4$ from acidified water samples [31–33]. ^{133}Ba radioisotope is added to the water sample as a tracer at the beginning of radium analysis. 10 mL of concentrated H_2SO_4 and 0.5 mL of Pb^{2+} (10 mg mL^{-1}) were added to the sample to co-precipitate Ba and Ra on PbSO_4 . The supernatant was discarded and the precipitate was centrifuged at 3,000 rpm for 5 min. The precipitate was washed with distilled water until neutral pH was achieved. The precipitate was dissolved in 4 mL of 0.1 M EDTA/0.5 M NaOH. The radium was co-precipitated with BaSO_4 by adding 0.3 mL of Ba^{2+} carrier (30 mg mL^{-1}), 1 drop pH indicator, 1.0 mL acetic acid to adjust the pH to 4.5, 4 mL of saturated Na_2SO_4 solution and 0.3 mL of a barium sulphate seeding suspension (10 mg mL^{-1}). The precipitate was left to stand for at least 30 min and then filtered through a 0.1 mm polypropylene filter. The filter was mounted on a stainless steel disc and measured by α -particle spectrometry for determination of

Table 1 Activity concentration of radionuclides A (mBq L^{-1}) in mineral water samples of Turkey with combined standard uncertainties ($k = 2$)

Sample code	^{234}U (mBq L^{-1})	^{238}U (mBq L^{-1})	Activity ratio $^{234}\text{U}/^{238}\text{U}$	^{226}Ra (mBq L^{-1})
MW1	13 ± 1.6	14 ± 1.7	0.93	119 ± 17
MW2	5 ± 0.8	4 ± 0.8	1.25	165 ± 22
MW3	<MDA	<MDA	–	83 ± 10
MW4	<MDA	<MDA	–	<MDA
MW5	34 ± 2.2	36 ± 2.3	0.94	87 ± 13
MW6	30 ± 4	27 ± 3.9	1.11	17.5 ± 2.8
MW7	32 ± 1.9	40 ± 4.5	0.80	<MDA
MW8	2 ± 0.3	2 ± 0.2	1.00	9.5 ± 1.3
MW9	464 ± 21	439 ± 19	1.06	50 ± 7.1
Range	<0.42–464	<0.42–439	0.80–1.25	<0.56–165

Minimum detectable activity (MDA) of ^{238}U , and ^{234}U is 0.42 mBq L^{-1} , ^{226}Ra is 0.56 mBq L^{-1} respectively

^{226}Ra activity concentration. Samples were counted for a period of 3–4 days. The chemical yield was evaluated by means of comparing the net peak area of ^{133}Ba at 356.0134 (7) keV in gamma energy spectrum of sample and standard (disc) source. The mean chemical yield was 85 %. The MDA was 0.56 mBq L^{-1} .

Instrumentation

The activity concentrations of uranium and radium radioisotopes were determined using alpha particle spectrometer from Canberra. Alpha sources were counted using two Passivated Implanted Planar Silicon (PIPS) detectors with active surface areas of 450 and 600 mm^2 (Canberra). The energy calibration of the spectrometer was performed using two electroplated mixed alpha standard sources containing ^{238}U : 1.67 Bq (±5.2 %), ^{234}U : 1.63 Bq (±4.9 %), ^{239}Pu : 1.91 Bq (±5.5 %), ^{241}Am : 1.81 Bq (±5.3 %) with a diameter of 24.2 mm and ^{238}U : 1.70 Bq (±2.8 %), ^{234}U : 1.69 Bq (±2.8 %), ^{239}Pu : 1.64 Bq (±2.8 %), ^{241}Am : 1.83 Bq (±2.8 %) with a diameter of 17.2 mm supplied from Eckert & Ziegler Isotope Products. The counting efficiency of the detector for radium analyses was determined by calculating the geometry factor (solid angle/ 4π) of the source-detector geometry [31]. The chemical recovery for radium analysis was determined by a High Purity Germanium (HPGe) gamma detector supplied from Ortec Instruments. Thin sources of uranium for alpha particle spectrometry measurement were prepared by electrodeposition on a stainless steel disc using a sulfate system as described by Eichrom ACW03 analytical procedure [29]. Electrodeposition cell assembly consisting of disposable plastic vial, cap assembly containing stainless steel (cathode) and platinum electrode (anode) and a power

supply (model 9611, ITN, Portugal) was used to accumulate uranium on stainless-steel disc of 20 mm in diameter.

Results and discussion

Activity concentration of ^{234}U , ^{238}U and ^{226}Ra in the mineral water samples

The activity concentration of ^{238}U , ^{234}U and ^{226}Ra in the mineral water samples as well as the $^{234}\text{U}/^{238}\text{U}$ activity ratios are presented in Table 1. The reported combined standard uncertainty for individual analyses in the table was multiplied by the statistical coverage factor $k = 2$, which was estimated from the uncertainties associated with tracer activity, the addition of yield tracer to the sample and the counting statistics of the sample and blank.

The results show that the uranium activity concentrations in the mineral water samples are in the range of $<0.42\text{--}439\text{ mBq L}^{-1}$ for ^{238}U and $<0.42\text{--}464\text{ mBq L}^{-1}$ for ^{234}U . The lowest activity concentrations of ^{238}U and ^{234}U were detected in MW3 and the highest activity concentrations of ^{238}U and ^{234}U were observed in the MW9. ^{238}U and ^{234}U activity concentrations of MW9 were found to be 439 ± 19 and $464 \pm 21\text{ mBq L}^{-1}$ respectively. The total activity concentration of uranium radioisotopes in this sample exceeded the existing WHO gross alpha screening level (500 mBq L^{-1}) [4].

High dissolved uranium concentration in underground waters is often associated with carbonate content [34]. This phenomenon can be explained in terms of the soluble carbonate compounds in these waters: These carbonates and bicarbonates make the waters basic and cause to form soluble uranyl complexes enhancing the concentration of dissolved uranium. High carbonate concentration was reported for the ground waters originating from the sampling site of MW9 [35]. This fact may explain high uranium content in mineral waters from this site.

The activity concentration of ^{238}U and ^{234}U in other mineral water samples were well below 50 mBq L^{-1} . The presented ^{238}U and ^{234}U levels for mineral waters can be considered low except MW9 sample and comparable with the data from many other countries [36–40].

If there are no chemical (leaching) or physical disturbing effects (alpha recoil), the activity ratio of $^{234}\text{U}/^{238}\text{U}$ is around 1.00 which states the radioactive secular equilibrium [36]. Equilibrium or close to equilibrium state between ^{234}U and ^{238}U activity concentrations was observed in most of the investigated mineral water samples. The $^{234}\text{U}/^{238}\text{U}$ activity ratio was found to vary between 0.80 and 1.25. MW7 sample with a high ^{238}U activity concentration (40 mBq L^{-1}) registered the lowest activity ratio (0.80). MW2 sample with 4 mBq L^{-1} of ^{238}U

Table 2 Dose coefficients of the relevant radionuclides (D_{coeff}) in mSv Bq^{-1} and water consumption rate (year^{-1}) for three age groups

Radionuclide	Dose coefficients (mSv Bq^{-1})		
	7–12 years	12–17	>17
^{238}U	6.80×10^{-5}	6.70×10^{-5}	4.50×10^{-5}
^{234}U	7.40×10^{-5}	7.40×10^{-5}	4.90×10^{-5}
^{226}Ra	8.00×10^{-4}	1.50×10^{-3}	2.80×10^{-4}
Water consumption rate (L year^{-1})	350	600	730

and 5 mBq L^{-1} of ^{234}U activity concentrations registered the highest level of $^{234}\text{U}/^{238}\text{U}$ activity ratio (1.25). The mean value for $^{234}\text{U}/^{238}\text{U}$ activity ratio was 1.01 ± 0.14 .

The ^{226}Ra activity concentrations in the mineral water samples are in the range of $<0.56\text{--}165\text{ mBq L}^{-1}$ as shown in Table 1. The measured ^{226}Ra activity concentrations of the samples were in most cases below 90 mBq L^{-1} , however the activity concentration of ^{226}Ra in MW1 and MW2 samples with 119 and 165 mBq L^{-1} respectively exceeded this value. The presented ^{226}Ra activity concentrations are comparable with the data for many other countries [36, 41–44]. The activity concentration of ^{226}Ra in MW8 sample was found to be lower than the previous results reported for the same region [5].

Estimation of the committed effective dose for different age groups due to mineral water intake

Activity concentrations of ^{234}U , ^{238}U and ^{226}Ra in mineral water samples were used to estimate the committed effective dose for three different age groups due to mineral water intake. Three age groups of the population investigated are children (ages between 7 and 12), teenagers (ages between 12 and 17) and adults (older than 17 years). Dose coefficients (Table 2) recommended by the International Commission of Radiological Protection and International Atomic Energy Agency were used for this purpose [45, 46]. The annual average consumption rate of water is also presented in Table 2 [1, 4].

Equations (1) and (2) and the data from Tables 1 and 2 were used for the annual committed effective dose calculation.

$$\text{CD}_{\text{Annual}} = \sum_i A_i F_i \quad (1)$$

where $\text{CD}_{\text{Annual}}$ is the annual committed effective dose (mSv a^{-1}), A_i is the activity concentration of nuclide i , F_i is a proportionality constant for nuclide i with units of (mSv a^{-1}) per (Bq L^{-1}). F_i is calculated using following equation:

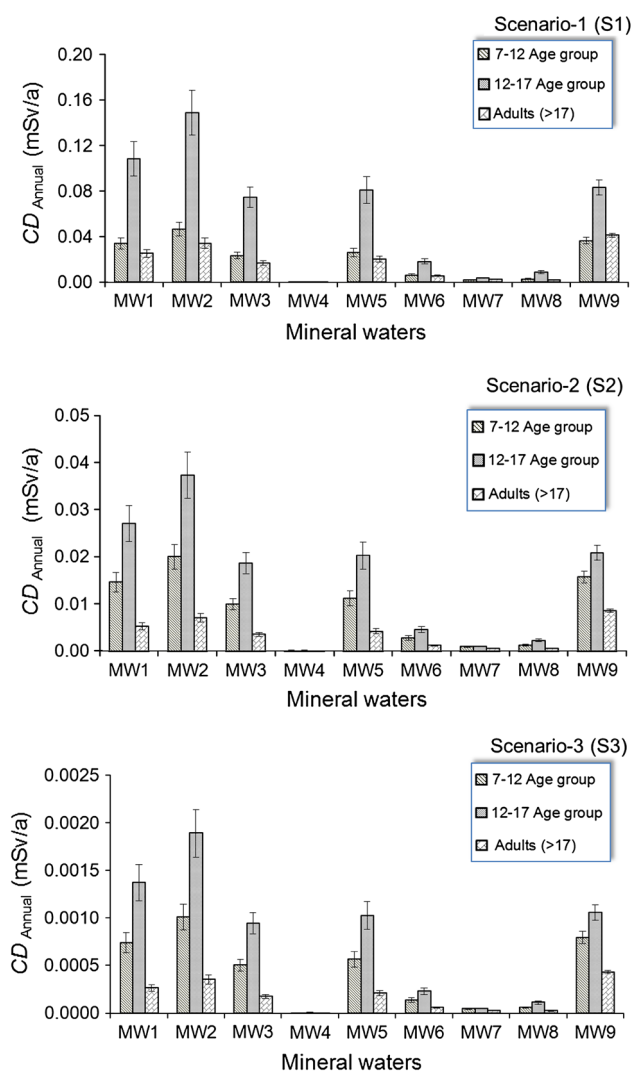


Fig. 2 Annual dose rates from mineral water samples for three different scenarios based on mineral water consumption

$$F_i = \sum_j (D_{\text{coeff}})_{ij} * CA_j * W_j \quad (2)$$

where CA_j is annual water consumption rate for an age group j (L a^{-1}), $(D_{\text{coeff}})_{ij}$ is the dose coefficient for radionuclide i and age group j (mSv Bq^{-1}) and W_j is a weighting factor for age group j . Different D_{coeff} values are given for different age groups due to changing chemical behaviour of the radionuclides in the body during the time period. This time period is given as 50 years for adult age group (>17) and $70-t_0$ for smaller age groups where t_0 is the average age in the age group assuming a total lifespan of 70 years for an average person [44]. The weighting factor for each age group is taken as unity (1) for all age groups for age specific committed effective dose. The weighting factors used in the calculation of lifetime annual committed effective dose for the children, teenagers and adults were

calculated by dividing 50 by 70 for adult age group and $(70-t_0)$ by 70 for all other age groups [45]. Lower limit of detection values were used for committed effective dose calculations where activity concentration values are below the lower limit of detection.

Three different scenarios were planned for dose assessment. In the first scenario (S1) it was assumed that the mineral water is consumed as drinking water with consumption rate of 730 L year^{-1} for adults, 600 L year^{-1} for teenagers and 350 L year^{-1} for children. The annual consumption rate of natural mineral water in European Countries (150 L year^{-1}) was assumed as the second scenario (S2) and this consumption rate was used for all age groups. The annual consumption rate of natural mineral water in Turkey (7.6 L year^{-1}) was assumed as the third scenario (S3) and used for all age groups [17, 18].

The annual committed doses for the three critical age groups of the population mentioned above in three different scenarios are presented in Fig. 2. In the most extreme scenario (S1), the committed effective dose for teenagers from the intake of ^{234}U , ^{238}U and ^{226}Ra isotopes in mineral waters varied between 0.0005 and $0.15 \text{ mSv year}^{-1}$ with an average value of $0.06 \text{ mSv year}^{-1}$. All water samples except MW1 and MW2 cause committed effective doses below the reference level ($0.1 \text{ mSv year}^{-1}$) recommended by WHO. Higher radium activities in these samples cause higher committed effective doses for teenagers due to the accumulation of radium in the body during the growth period in which the bone mass increases rapidly. Committed effective doses in S1 varied between 0.0002–0.047 and 0.0001–0.04 mSv year^{-1} for 7–12 and >17 age groups, respectively.

In scenario two (S2) and scenario three (S3) the committed effective dose for all age groups were found to be well below the reference level recommended by the WHO.

The lifetime annual committed effective doses are between 0.0002–0.04, 0.00004–0.0094 and 0.000002–0.0005 mSv a^{-1} for S1, S2 and S3, respectively. These doses are all below the WHO guidelines.

Conclusions

Nine mineral water samples from three different regions of Turkey were examined for their radiochemical content. The activity concentrations of ^{234}U , ^{238}U and ^{226}Ra were determined by alpha particle spectrometry.

The activity concentrations of ^{238}U and ^{234}U range from <0.42 to 439 mBq L^{-1} and from <0.42 to 464 mBq L^{-1} respectively. The activity concentration of ^{226}Ra ranges from <0.56 to 165 mBq L^{-1} . In one sample elevated level of uranium ($>400 \text{ mBq L}^{-1}$) and in two samples elevated level of radium ($>100 \text{ mBq L}^{-1}$) were obtained with a

maximum of 464 mBq L⁻¹ for ²³⁴U, 439 mBq L⁻¹ for ²³⁸U and 165 mBq L⁻¹ for ²²⁶Ra respectively. Secular equilibrium between ²³⁴U and ²³⁸U activity concentrations was observed in most of the mineral water samples.

The potential committed effective doses for an average person with a life span of 70 years were calculated for three different scenarios according to the mineral water consumption. In the most extreme scenario (S1) where mineral water is consumed as drinking water, the lifetime annual committed effective doses are in the range of 0.0002–0.04 mSv year⁻¹ with an average of 0.018 mSv year⁻¹. In all scenarios none of the mineral water samples exceeded the reference level recommended by WHO (0.1 mSv year⁻¹).

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Conflict of interest The authors declare that they have no conflict of interest.

References

- United Nations Scientific Committee on the Effects of Atomic Radiation, UNSCEAR (2000) Report to the general assembly, with scientific annexes, vol. I. Sources and effects of ionizing radiation. United Nations Publication, Austria
- European Commission Council Directive 98/83/EC of 3 November 1998 on the quality of water intended for human consumption. Official Journal of the European Communities L 330, 5.12.1998
- World Health Organization (1998) Guidelines for drinking water quality, 2nd ed. vol. 1. Geneva, Switzerland
- World Health Organization (2004) Guidelines for drinking water quality, 3rd ed. vol. 1. Geneva, Switzerland
- Kobya Y, Damla N, Cevik U, Kobya AI (2011) Radiochemical characterization of mineral waters in the Eastern Black Sea Region, Turkey. *Environ Monit Assess* 182:415–422
- Wallner G, Wagner R, Katzlberger C (2008) Natural radionuclides in Austrian mineral water and their sequential measurement by fast methods. *J Environ Radioact* 99:1090–1094
- Kozłowska B, Walencik A, Dorda J, Przylibski TA (2007) Uranium, radium and ⁴⁰K isotopes in bottled mineral waters from outer Carpathians, Poland. *Radiat Meas* 42:1380–1386
- Wrenn ME, Durbin PW, Howard B, Lipsztein J, Rundo J, Still Edwin T, Willis DL (1985) Metabolism of ingested U and Ra. *Health Phys* 48(5):601–633
- World Health Organization (2011) Guidelines for drinking water quality, 4th ed. vol. 1. Geneva, Switzerland
- United States Environmental Protection Agency (EPA). <http://water.epa.gov/drink/contaminants/basicinformation/radionuclides.cfm#What%20are%20EPA%E2%80%99s%20drinking%20water%20regulations%20for%20radionuclides>
- Bundesgesetzblatt Nr. 56 Seite 2762:Deutsche mineral and tafelwasserverordnung, 12.12.2006
- Benedik L, Repinc U, Strok M (2010) Evaluation of procedures for determination of Ra-226 in water by alpha-particle spectrometry with emphasis on the recovery. *Appl Radiat Isot* 68:1221–1225
- Lawrie WC, Desmond JA, Spence D, Anderson S, Edmondson C (2000) Determination of radium-226 in environmental and personal monitoring samples. *Appl Radiat Isot* 53:133–137
- Barut İF, Erdoğan N, Başak E (2004) Hydrogeochemical evaluation of Western Anatolian mineral waters. *Environ Geol* 45: 494–503
- Tosun M (2005) İçme ve maden suyu sektör araştırması. Türkiye Kalkınma Bankası A.Ş., SA-05-01-01, Ankara, Türkiye
- Cemek M, Akkaya L, Birdane YO, Seyrek K, Bulut S, Konuk M (2007) Nitrate and nitrite levels in fruit and natural mineral waters marketed in western Turkey. *J Food Compos Anal* 20: 236–240
- Maden suyu potansiyeli kullanılmıyor. <http://www.gidagundemi.com/turkiye/madensuyu-potansiyeli-kullanilmiyor-h1375.html>. Accessed 17 Oct 2013
- Maden suyu üreticileri hedef büyüttü. <http://www.hurriyet.com.tr/ekonomi/23325802.asp> Accessed 20 Sep 2013
- Sahin L, Çetinkaya H, Saç MM, İçhedef M (2013) Determinations of radon and radium concentrations in drinking water samples around the city of Kütahya. *Radiat Prot Dosim* 155: 472–474
- Tabar E, Kumru MN, Saç MM, İçhedef M, Bolca M, Özen F (2013) Radiological and chemical monitoring of Dikili geothermal waters, Western Turkey. *Radiat Phys Chem* 91:89–97
- Turhan S, Özcıttak E, Taşkın H, Varinlioglu A (2013) Determination of natural radioactivity by gross alpha and beta measurements in ground water samples. *Water Res* 47:3103–3108
- Çevik U, Damla N, Karahan G, Çelebi N, Kobya AI (2006) Natural radioactivity in tap waters of Eastern Black Sea region of Turkey. *Radiat Prot Dosim* 118:88–92
- Saç MM, Aydemir S, İçhedef M, Kumru MN, Bolca M, Özen F (2014) Natural radioactivity levels of geothermal waters and their influence on soil and agricultural activities. *Radiat Prot Dosim* 158:148–155
- Korkmaz Görür F, Keser R, Dizman S, Okumuşoğlu NT (2011) Annual effective dose and concentration levels of gross α and β in various waters from Samsun, Turkey. *Desalination* 279: 135–139
- Taskin H, Asliyukse H, Bozkurt A, Kam E (2013) Natural radioactivity in bottled mineral and thermal spring waters of Turkey. *Radiat Prot Dosim* 157:575–578
- Türkiye Atom Enerjisi Kurumu, Teknik Raporu (2012) Türkiye’deki doğal mineralli (maden suları) içmece sularının toplam alfa/beta, ²²⁶Ra, ²³⁴U, ²³⁸U, ²¹⁰Po ve ²²⁸Ra aktivitelerinin belirlenmesi ve toplam gösterge dozlarının hesaplanması TAEK TR-2012-06
- United States Environmental Protection Agency (EPA) (2007) Gross alpha and gross beta radioactivity in drinking water method 900.0 http://water.epa.gov/scitech/methods/cwa/Bioindicators/upload/2007_07_10_methods_method_900.0.pdf
- http://d-maps.com/carte.php?num_car=15375&lang=en Accessed 14 Feb 2014
- Eichrom Technologies, Inc (2001) Uranium in Water ACW02 Analytical Procedures Rev. 1.3. Eichrom Technologies, Inc, Lisle
- Amoli HS, Barker J, Flowers A (2006) Electrodeposition and determination of nano-scale uranium and plutonium using alpha spectroscopy. *J Radioanal Nucl Chem* 268:497–501
- Dirican A, Erden PE, Seferinoğlu M, Pommé S (2012) The use of solid angle for alpha detector efficiency in ²²⁶Ra analyses of soil samples. *Appl Radiat Isot* 70:2260–2262
- Lozano JC, Fernandez F, Gomez JMG (1997) Determination of radium isotopes by BaSO₄ co-precipitation for the preparation of alpha-spectrometric sources. *J Radioanal Nucl Chem* 22:133–137
- Benedik L, Spasova Y, Vasile M, Watjen U (2008) Determination of ²³⁴U, ²³⁸U and ²²⁶Ra in bottled drinking water by alpha spectrometry. In: CD proceedings of the seventh international conference on nuclear and radiochemistry (NRC-7), Budapest, Hungary
- Jobbágy V, Kávási N, Somlai J, Máté B, Kovács T (2010) Radiochemical characterization of spring waters in Balaton

- Upland, Hungary, estimation of radiation dose to members of public. *Microchem J* 94:159–165
35. Dilsiz C, Marques JM, Carreira PMM (2004) The impact of hydrological changes on travertine deposits related to thermal springs in the Pamukkale area (SW Turkey). *Environ Geol* 45:808–817
36. Jobbágy V, Dirican A, Wätjen U (2013) Radiochemical characterization of mineral waters for a European interlaboratory comparison. *Microchem J* 110:675–680
37. Benedik L, Jeran Z (2012) Radiological of natural and mineral drinking waters in Slovenia. *Radiat Prot Dosim* 151:306–313
38. Labidi S, Mahjoubi H, Essafi F, BenSalah R (2010) Natural radioactivity levels in mineral, therapeutic and spring waters in Tunisia. *Radiat Phys Chem* 79:1196–1202
39. Gharbi F, Baccouche S, Abdelli W, Samaali M, Oueslati M, Trabelsi A (2010) Uranium isotopes in Tunisian bottled mineral waters. *J Environ Radioact* 101:50–589
40. Díaz-France's I, Mantero J, Manjón G, Díaz J, García-Tenorio R (2013) ^{210}Po and ^{238}U isotope concentrations in commercial bottled mineral water samples in Spain and their dose contribution. *Radiat Prot Dosim* 156:336–342
41. Janković MM, Todorović DJ, Todorović NA, Nikolov J (2012) Natural radionuclides in drinking waters in Serbia. *Appl Radiat Isot* 70:2703–2710
42. Wätjen U, Benedik L, Spasova Y, Vasile M, Altitzioglou T, Beyermann M (2010) EC comparison on the determination of ^{226}Ra , ^{228}Ra , ^{234}U and ^{238}U in water among European monitoring laboratories. *Appl Radiat Isot* 68:1200–1206
43. Rožmarić M, Rogić M, Benedik L, Štok M (2012) Natural radionuclides in bottled drinking waters produced in Croatia and their contribution to radiation dose. *Sci Total Environ* 437:53–60
44. Wallner G, Jabar T (2010) Natural radionuclides in Austrian bottled mineral waters. *J Radioanal Nucl Chem* 286:329–334
45. International Commission on Radiological Protection (ICRP) (1996) Age-dependent doses to members of the public from intake of radionuclides: part 5. Compilation of ingestion and inhalation dose coefficient. ICRP Publication 72. Pergamon Press, Oxford, p 1996
46. International Atomic Energy Agency (IAEA) (1996) International basic safety standards for protection against ionizing radiation and for the safety of radiation sources. Safety Report Series, No. 115. International Atomic Energy Agency (IAEA), Vienna