Journal of Radioanalytical and Nuclear Chemistry (2021) 328:805–814
https://doi.org/10.1007/s10967-021-07736-6

Radiological and physicochemical properties of drinking waters consumed in the Western Black Sea Region of Turkey

Aslı Kurnaz[1] · Şeref Turhan[1] · Fatma M. N. S. Alzaridi[2] · Temel Kan Bakır[3]

Received: 22 October 2020 / Accepted: 16 April 2021 / Published online: 27 April 2021

Abstract
In this study, the average activity concentrations of ^{226}Ra, ^{228}Ra and ^{40}K in drinking waters (tap and bottled) consumed in Kastamonu province of Turkey were found as 0.7, 0.5 and 1.3 Bq L^{-1}, respectively. The average value of pH, conductivity, total dissolved solids and total hardness of waters were found as 7.5, 217 µS cm^{-1}, 142 mg L^{-1} and 154 mg L^{-1} for tap waters and 7.4, 94 µS cm^{-1}, 61 mg L^{-1} and 58 mg L^{-1} for bottled waters. The average activity concentration of ^{228}Ra in drinking waters was higher than the WHO recommended concentration of 0.1 Bq L^{-1}.

Keywords Drinking water · Natural radioactivity · Annual effective dose · Gamma-ray spectrometry

Introduction

Drinking water intended for human consumption is obtained from water sources such as lakes, streams, ponds, wells, springs, rainfall, etc. The drinking water of major cities is generally supplied from dams or surface water reservoirs built at main rivers and artificial lakes. However, drinking water sources are contaminated by microbial, chemical, and radioactive contaminants due to industrial and urbanization growth. The pouring of industrial, radioactive, and domestic wastes, and disposal of heavy metals are the main causes of drinking water pollution. The contamination of drinking water resources reduces the quality of drinking water and causes significant risks to human health. Therefore, access to safe and quality drinking water is the most fundamental human right and an essential element of health protection [1]. Drinking water quality can be characterized based on water parameters such as microbial, chemical, physical, and radiological. If values of these parameters exceed the acceptable limits or criteria and then human health is at risk. Although the quality of drinking water is strictly controlled by national and/or international organizations, the safety, cleanliness, and quality of drinking water resources continue to be an important problem all over the world.

Drinking water may include terrestrial radionuclides emitting ionizing radiations that could have radiological hazards, which are small compared to the hazards caused by microorganisms and chemicals found in drinking water, to human health [1]. The radionuclides in drinking water occur from natural processes such as absorption from the soil or technological processes involving naturally occurring radioactive materials (NORMs) such as the mining and processing of mineral sands or phosphate fertilizer production [1]. Ingestion and inhalation are the two main routes by which members of the public can be exposed to the ionizing radiations (α-, β- and γ-rays) emitted from the terrestrial radionuclides in drinking water supplies. Ingestion doses are mainly due to radionuclides in the uranium (^{238}U) and thorium (^{232}Th) series, and radioactive potassium (^{40}K) exist in drinking water intake. Inhalation doses result from the existence in an air of dust particles containing radon (^{222}Rn, half-life 3.8 days) dissolved in water and its the short-lived decay products such as polonium (^{218,214}Po), lead (^{214}Pb), and bismuth (^{214}Bi) emitting primarily α-a and β-rays. The WHO individual dose criterion (IDC) of 0.1 mSv/y associated with the intake of radionuclides from drinking water

✉ Fatma M. N. S. Alzaridi
ftmmnslzd@gmail.com

Aslı Kurnaz
akurnaz@kastamonu.edu.tr

1 Department of Physics, Faculty of Science and Letters, Kastamonu University, 37150 Kastamonu, Turkey

2 Department of Physics, Faculty of Science, Misurata University, Misurata, Libya

3 Department of Chemistry, Faculty of Science and Letters, Kastamonu University, 37150 Kastamonu, Turkey

indicates a very low level of risk that is not expected to give rise to any detectable adverse health effect [1].

The determination of the radioactivity levels of water is the priority to assess the drinking water quality from a radiological point of view. Recently many studies were conducted to research the natural radioactivity in drinking waters (tap, ground, surface, mineral, and spring water samples) around the world [2–20]. The determination of the radioactivity level of tap water provided from surface water is a higher priority because of the most important drinking water source in the majority of cities in Turkey. Until now, a study was conducted to determine the activity concentration of ^{222}Rn in tap water samples consumed in Kastamonu province [21]. However, a detailed study is not performed for the determination of radioactivity levels of the natural radionuclides and measurement of some physicochemical parameters of tap and bottled drinking waters consumed in Kastamonu. Therefore, the aim of the present study is (1) to measure some physicochemical parameters (pH, conductivity, total dissolved solids, and total hardness) of drinking water (tap and bottled drinking water) samples collected from different sampling locations chosen to reflect the distribution of population and purchased from markets in Kastamonu, (2) to determine the activity concentration of ^{226}Ra, ^{228}Ra and ^{40}K, which are radiologically important, in drinking water samples, (3) to assess the radiological hazards due to the internal exposure to adults associated with the ingestion of drinking water samples, and (4) to compare the measured and estimated values with the limits recommended by national and/or international organizations for drinking water and the those obtained for drinking waters consumed by other countries. Therefore, this study can be interesting and will be meaningful for sustainable drinking water quality protection.

Material and methods

Collection and preparation of samples

Kastamonu province is located between 41°21′ N latitude and 33°46′ E longitude in the West Black Sea region of Turkey. The border of Kastamonu province is located in a wide region extending from the Black Sea coast to Central Anatolia and has the largest area of the Black Sea Region of Turkey [22]. Its height above sea level is 775 m. It has an area of 13.108 km^2 and 74.6% of the surface area is mountainous and forested, 21.6% is a plateau and 3.8% is plain. Kastamonu is one of the most important nature tourism destinations of Turkey due to its nature of the landscape and the coast. The population of Kastamonu province as per the 2019 census is 379,405 of which 152,553 live in the urban center. Since Kastamonu province has a long coast to the Black Sea, it can be expected to show marine climate character (humid/temperate). However, it takes on a terrestrial climate structure as it goes from the coast to the inner regions. The average annual rainfall and the average temperature are 488.4 mm and 10.3 °C, respectively.

Most of the tap water in the urban center is supplied from the Karaçomak River and the other part is provided from two surface water sources fed from Ilgaz Mountain in Kastamonu. Surface water from this source is stored in the Karaçomak dam and delivered to a treatment site before the final release into the municipal water supply network. Approximately 3×10^6 m^3 of water from this source is distributed to the residents of Kastamonu in a year. Thirty-one water samples were taken directly from the tap at different dwelling sites located in all parts of the province (from its north to south and from east to west) shown in Fig. 1. For tap water samples, the tap was opened, and the water was allowed to run for at least 15 min to ensure that the water was not delayed in the pipes. Bottled drinking water samples belonging to ten different brands consumed as drinking water in Kastamonu were purchased from markets. All water samples were taken in polyethylene bottles and then the samples were transported to the laboratory. The tap and bottled drinking water samples were coded as TW and BDW, respectively. The temperature of each drinking water was measured as 20 °C for the measurement of the physicochemical parameters. For radioactivity measurements, each water sample coded with the information from where they were collected was transferred to a polystyrene sample container a volume of 118 mL whose geometry and size were the same as the reference source prepared for detector efficiency. Then, each cover portion of the sample container was tightly wrapped with Teflon tape to seal the radon gas. The water samples were kept for at least one month to achieve a secular equilibrium between ^{226}Ra and ^{222}Rn, and ^{228}Ra and ^{228}Ac [23].

Experimental

pH, total dissolved solids (TDS), and conductivity measurements were made using a digital multi-parameter (ISOLAB Laborgerate GmbH) instrument with a combined glass and conductivity electrode. Instrument calibrations were checked using analytical purity calibration solutions before each measurement. Deionized water was used at each stage for electrode cleaning. The hardness of water is an expression of the sum of the calcium and magnesium cation concentration in a water sample. The total hardness (TH) of the drinking water samples was determined by complexometric titration using a standard ethylenediaminetetraacetic acid (EDTA) solution. Firstly 100 mL of water sample was taken and then the pH value of the sample was adjusted to 10 with buffer solution (NH_4Cl+NH_4OH). After the addition of 0.1 g total hardness indicator (Eriochrome Black T ground with NaCl),

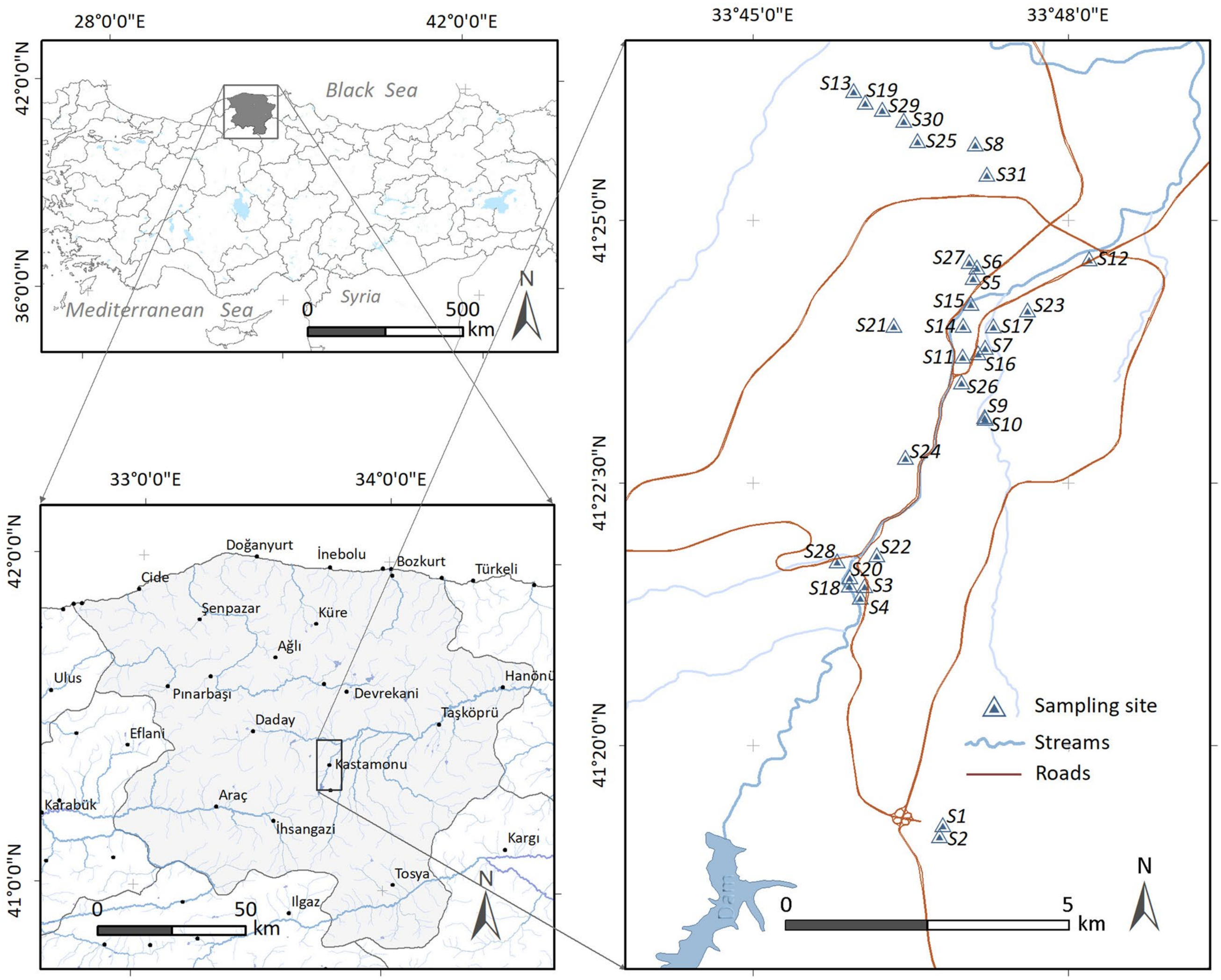

Fig. 1 Sampling sites on the map of Kastamonu province

titrate with 0.01 M EDTA (Tritriplex III) solution until the color changes from wine red to blue. The TH values calculated for each sample are expressed in ppm.

The activity concentrations of ^{226}Ra, ^{228}Ra, and ^{40}K in the drinking water samples were determined using the gamma-ray spectrometer with a p-type HPGe coaxial detector with a relative efficiency of 50% and energy resolution of 1.9 keV at 1.33 meV gamma-ray photons emitted from ^{60}Co. The details of the gamma-ray spectrometer are given in the study performed by Sultan et al. [23]. Radionuclide specific efficiency calibration of the detector was carried out by using the natural uranium (RGU-1) from the IAEA to reduce the uncertainty in gamma-ray intensities, the influence of coincidence summation, and self-absorption effects of the emitting gamma-ray photons. IAEA RGU-1 reference material used in calibrating laboratory gamma-ray spectrometers for the determination of uranium in geological materials was prepared by dilution of a uranium ore BL-5 (7.09% U) with floated silica powder of similar grain size distribution. 10 g of RGU-1 reference material was taken and put into a polystyrene sample container with 118 mL filled with one third HCl acid. It was kept for 2 weeks. This mixture was then stirred in a magnetic stirrer (Misung MS300HS) for about 2 h by gradually increasing the temperature values to 10, 25, 50 °C and the rotation speed to 10, 20, 30 and 40 rpm, respectively. The white liquid obtained after the mixture was left to rest for 1 day. The mixture is then filtered so that the insoluble fraction that settles to the bottom of the container is removed. Thus, dissolved uranium was obtained as a light yellow and transparent liquid. It was transferred to the same volume of sample container filled with 2/3 ultra-pure water. The standard container was sealed and kept for at least one month.

The standard container was sealed and kept for at least one month. Then the prepared standard was placed on the detector and counted until good statistics were obtained. The gamma-ray photopeaks of daughters (295.2 and 351.9 keV from ^{214}Pb, and 609.3, 1120.3, 1764.5 and 2204.1 keV from ^{214}Bi) in equilibrium with ^{226}Ra were used for the efficiency calibration of the gamma-ray spectrometers. The gamma-ray photopeak efficiencies were determined by fitting the experimental values to the function given below:

$$\varepsilon_\gamma = \frac{1}{(a + b \times E_\gamma^c)} \quad (1)$$

where E_γ is the energy of the gamma-ray photopeak, and *a*, *b* and *c* are 4.64, 0.0973 and 0.899, respectively. Each water sample container placed on the detector was counted for 80,000 s. The background measurements were done for 86,400 s every weekend by using an empty sample container. The activity concentration of ^{226}Ra was measured by using the weighted average activity concentrations of the gamma-ray photopeak at 351.9 keV (^{214}Pb) with an emission probability of 35.1% and the gamma-ray photopeak at 609.3 keV (^{214}Bi) with an emission probability of 44.6%. The activity concentration of ^{232}Th was measured using the activity concentration of gamma-ray photopeak at 911.2 keV (^{228}Ac) with an emission probability of 26.6% while the activity concentration of ^{40}K was measured directly by gamma-ray photopeak at 1460.8 keV with an emission probability of 10.7%. The activity concentrations (*A*) of radionuclides in the drinking water samples were by the following equation:

$$A(Bq\,\mathrm{L}^{-1}) = \frac{NCR}{\varepsilon_\gamma \cdot P_\gamma \cdot V} \quad (2)$$

where NCR is the net count rate of gamma-ray photopeak of interest, ε_γ is the efficiency of the gamma-ray photopeak of interest, $P\gamma$ is the emission probability of the gamma-ray of interest, and *V* is the volume of the water sample (*L*). The net count rate of each gamma-ray photopeak of interest was obtained by subtracting the counts of the gamma-ray photopeak in the background spectrum. The minimum detectable activity for ^{226}Ra, ^{228}Ra and ^{40}K were found as 0.2, 0.2 and 0.8 Bq L^{-1}, respectively.

Annual effective dose due to internal exposure

An annual effective dose was estimated to the radiological hazards due to the internal exposure to adults associated with the ingestion of drinking water samples based on the measured radionuclide activity concentrations. The ingestion annual effective dose (AED$_{\mathrm{ING}}$) was estimated as follows:

$$\mathrm{AED_{ING}}\ (\mathrm{mSv\ y^{-1}}) = A\ (\mathrm{Bq\ L^{-1}}) \times DCF_{ING}\ (\mathrm{Sv\ Bq^{-1}}) \times \mathrm{AWC}\ (\mathrm{L\ y^{-1}}) \times 10^3 \quad (3)$$

where *A* is the activity concentration of the radionuclides (^{226}Ra, ^{228}Ra, and ^{40}K), DCF$_{\mathrm{ING}}$ is the dose conversion factor for ingestion and AWC is an annual water consumption per capita. The value of DCF$_{\mathrm{ING}}$ for adults is taken as 2.8×10^{-7}, 6.9×10^{-7} and 6.2×10^{-9} Sv Bq^{-1} for ^{226}Ra, ^{228}Ra and ^{40}K, respectively [24].

Results and discussion

Some physicochemical parameters of drinking water samples

The values of pH, conductivity, total dissolved solids (TDS), and total hardness (TH) measured in the studied TW and BDW samples are given in Table 1. In assessing the drinking water quality, it is important to determine the pH level, which is one of the most important operational water quality parameters, because it can affect chemical reactions such as solubility [1, 25]. According to the World Health Organization (WHO) Guidelines for drinking water quality there is no health concern at pH levels found in drinking waters [1]. However, Turkish regulation [26] and Turkish standard (TS) [27], and European Union (EU) directive [28] recommend a range of 6.5–9.5 for pH levels ($6.5 \le \mathrm{pH} \le 9.5$) in drinking water while WHO [1] and the United States Environmental Protection Agency (US EPA) [29] recommend a range of 6.5–8.5 ($6.5 \le \mathrm{pH} \le 8.5$). The pH values of the TW and BDW samples varied from 6.7 to 7.9 with an average of 7.5 and 6.9–8.1 with an average of 7.4, respectively. The pH levels found in tap and bottled drinking water samples are within recommended limits. Conductivity related to the dissolved mineral solids in water can be defined as a measure of the ability of water to conduct an electrical current [1]. The electrical conductivity values of the TW and BDW samples varied from 5 to 327 μS cm^{-1} with an average of 217 μS cm^{-1} and 8–259 μS cm^{-1} with an average of 94 μS cm^{-1}, respectively. The conductivities of all tap and bottled drinking water samples are lower than the maximum limit of 2500 μS cm^{-1} set for drinking waters by the Turkish regulation and standard, and EU directive [26–28]. The TDS contains inorganic salts (magnesium, calcium, potassium, sodium, sulfates, chlorides, bicarbonates), and a small quantity of organic matter dissolved in water. There is no risk of possible health effects caused by the ingestion of TDS in drinking waters (WHO, 2011). The national and international organizations have not set a mandatory TDS limit, only the EPA recommends a maximum level of 500 mg L^{-1} [29]. The TDS contents of the TW and BDW samples varied from 3 to 216 mg L^{-1} with an average of 142 mg L^{-1} and from 5 to 171 mg L^{-1} with an average of 61 mg L^{-1}, respectively. The

Table 1 Physicochemical values measured for drinking waters

Sample Code	pH	Conductivity ($\mu S\ cm^{-1}$)	TDS ($mg\ L^{-1}$)	TH ($mg\ L^{-1}$)
TW-1	7.3	223	147	134
TW-2	7.6	165	109	143
TW-3	7.7	258	169	152
TW-4	7.0	22	15	27
TW-5	7.5	292	193	215
TW-6	7.4	136	90	86
TW-7	7.6	166	109	143
TW-8	7.5	293	193	188
TW-9	7.6	169	110	143
TW-10	7.5	200	130	125
TW-11	7.8	316	203	242
TW-12	7.7	311	205	206
TW-13	7.4	273	179	204
TW-14	6.9	12	7	27
TW-15	7.9	327	216	224
TW-16	7.5	160	106	143
TW-17	6.7	5	3	18
TW-18	7.6	210	138	143
TW-19	7.6	227	149	143
TW-20	7.6	295	192	197
TW-21	7.5	156	100	118
TW-22	7.6	233	151	166
TW-23	7.6	164	107	125
TW-24	7.4	278	182	193
TW-25	7.7	308	203	209
TW-26	7.6	287	190	202
TW-27	7.8	326	214	233
TW-28	7.8	277	182	190
TW-29	7.8	321	212	215
TW-30	7.1	22	14	29
TW-31	7.5	286	188	197
BDW-1	7.3	97	65	79
BDW-2	7.2	44	29	54
BDW-3	8.1	74	49	63
BDW-4	7.4	131	85	72
BDW-5	7.1	8	5	27
BDW-6	7.7	259	171	54
BDW-7	7.5	92	59	47
BDW-8	7.2	55	35	54
BDW-9	6.9	73	48	48
BDW-10	7.4	103	67	81

total hardness (TH) of water caused by calcium (Ca) and magnesium (Mg) cation is usually represented by the precipitation of soap scum and the requirement for excess use of soap to accomplish cleaning [1]. There is no health-based guideline value for hardness in drinking water. However, the public acceptability of the TH degree of drinking water may vary significantly from one community to another [1]. The taste thresholds for the Ca ion vary from 100 to300 mg L^{-1} and the taste threshold for Mg is likely below that for Ca. In some cases, consumers tolerate water hardness above 500 mg L^{-1} [1]. The TH contents of the TW and BDW samples varied from 18 to 242 mg L^{-1} with an average of 154 mg L^{-1} and from 27 to 81 mg L^{-1} with an average of 58 mg L^{-1}, respectively.

Radionuclide concentration of drinking water samples

The activity concentrations of ^{226}Ra, ^{228}Ra, and ^{40}K measured in the studied drinking water samples are given in Table 2. Descriptive statistical data on the activity concentration of radionuclides in tap, bottled drinking, and all water samples is given in Table 3. The comparison of the average activity concentrations of these radionuclides in the drinking water samples with those measured for drinking water samples consumed in different countries and guidance levels recommended by the WHO and EPA is presented in Table 4. The frequency distributions of the activity concentrations of ^{226}Ra, ^{228}Ra, and ^{40}K are shown in Fig. 2.

Radium with an atomic number of 88 is an alkaline earth metal and has more than 20 radioactive isotopes [35]. However, ^{226}Ra and ^{228}Ra are significant in terms of radiological protection due to their presence in nature, their relatively long half-lives, and high dose conversion factors [35]. ^{226}Ra (half-life: 1600 y and alpha-particle emitter) and ^{228}Ra (half-life: 5.75 y and beta-particle emitter) can be easily incorporated into bones due to having similar properties to other elements in Group II of the periodic table such as Ca and produce daughters with a short half-life and high activity concentration [35]. It can be seen from Table 2 and 3 that the activity concentrations of ^{226}Ra and ^{228}Ra measured in the studied drinking water (tap and bottled) samples varied from 0.3 to 1.5 Bq L^{-1} with an average of 0.7 Bq L^{-1} and 0.3 to 1.3 Bq L^{-1} with an average of 0.5 Bq L^{-1}. At the same time, the average activity concentrations of ^{226}Ra and ^{228}Ra measured in TW and BDW samples are close to each other. The activity concentration distribution of ^{226}Ra and ^{228}Ra

Table 2 Radionuclide concentrations measured in drinking waters

Sample code	Activity concentration (Bq L^{-1})		
	^{226}Ra	^{228}Ra	^{40}K
TW-1	1.05 ± 0.10	0.35 ± 0.05	1.90 ± 0.15
TW-2	1.25 ± 0.10	0.35 ± 0.05	1.00 ± 0.10
TW-3	0.45 ± 0.05	0.40 ± 0.05	1.65 ± 0.15
TW-4	0.90 ± 0.05	0.35 ± 0.05	1.25 ± 0.10
TW-5	0.95 ± 0.10	0.40 ± 0.05	1.20 ± 0.10
TW-6	0.80 ± 0.05	0.35 ± 0.05	1.50 ± 0.15
TW-7	0.75 ± 0.05	1.25 ± 0.10	1.15 ± 0.10
TW-8	0.95 ± 0.10	0.60 ± 0.05	1.45 ± 0.10
TW-9	0.85 ± 0.05	0.55 ± 0.05	1.10 ± 0.10
TW-10	0.70 ± 0.05	0.25 ± 0.05	1.05 ± 0.10
TW-11	1.50 ± 0.10	0.30 ± 0.05	1.55 ± 0.15
TW-12	0.45 ± 0.05	0.40 ± 0.05	1.05 ± 0.10
TW-13	0.45 ± 0.05	0.30 ± 0.05	1.65 ± 0.15
TW-14	0.70 ± 0.05	0.35 ± 0.05	1.10 ± 0.10
TW-15	0.40 ± 0.05	0.85 ± 0.10	1.20 ± 0.10
TW-16	0.45 ± 0.05	0.75 ± 0.05	0.95 ± 0.10
TW-17	1.00 ± 0.10	0.80 ± 0.05	1.10 ± 0.10
TW-18	1.25 ± 0.10	0.60 ± 0.05	1.55 ± 0.15
TW-19	0.50 ± 0.05	0.55 ± 0.05	1.40 ± 0.10
TW-20	0.45 ± 0.05	0.35 ± 0.05	1.20 ± 0.10
TW-21	0.30 ± 0.00	0.45 ± 0.05	1.50 ± 0.15
TW-22	0.50 ± 0.05	0.45 ± 0.05	1.40 ± 0.10
TW-23	0.45 ± 0.05	0.40 ± 0.05	1.30 ± 0.10
TW-24	0.45 ± 0.05	0.40 ± 0.05	1.20 ± 0.10
TW-25	0.45 ± 0.05	0.30 ± 0.05	1.35 ± 0.10
TW-26	1.00 ± 0.10	0.45 ± 0.05	0.95 ± 0.10
TW-27	0.80 ± 0.05	0.60 ± 0.05	0.90 ± 0.10
TW-28	0.40 ± 0.05	0.45 ± 0.05	0.85 ± 0.05
TW-29	0.60 ± 0.05	0.50 ± 0.05	0.85 ± 0.05
TW-30	0.70 ± 0.05	0.65 ± 0.05	1.00 ± 0.10
TW-31	1.00 ± 0.75	0.75 ± 0.05	0.95 ± 0.10
BDW-1	0.10 ± 0.05	0.75 ± 0.05	1.90 ± 0.15
BDW-2	0.65 ± 0.05	0.55 ± 0.05	1.70 ± 0.15
BDW-3	0.75 ± 0.05	1.10 ± 0.10	1.20 ± 0.10
BDW-4	0.45 ± 0.05	0.45 ± 0.05	2.05 ± 0.15
BDW-5	0.60 ± 0.05	0.40 ± 0.05	1.00 ± 0.10
BDW-6	0.55 ± 0.05	0.40 ± 0.05	1.60 ± 0.15
BDW-7	0.90 ± 0.05	0.40 ± 0.05	1.05 ± 0.10
BDW-8	0.70 ± 0.05	0.70 ± 0.05	1.25 ± 0.10
BDW-9	0.85 ± 0.05	0.75 ± 0.05	1.70 ± 0.15
BDW-10	0.85 ± 0.05	0.45 ± 0.05	1.45 ± 0.10

shows the near-normal distribution shown in Fig. 2. 78% and 17% of the activity concentrations of ^{226}Ra measured in all drinking water samples are in the range of 0.3–0.9 Bq L^{-1} and 1.0–1.5 Bq L^{-1} respectively. 95% and 5% of the activity concentrations of ^{228}Ra measured in all drinking water samples are in the range of 0.3–0.9 Bq L^{-1} and 1.0–1.3 Bq L^{-1} respectively. As can be seen in Table 4, the average activity concentration of ^{226}Ra in the drinking water samples is higher than those measured in drinking water samples except for the water samples consumed in Canada. The average activity concentration of ^{228}Ra in the drinking water samples is higher than those measured in drinking water samples except for the water samples consumed in Iran (Kermanshah). The activity concentrations of ^{226}Ra measured in only four tap water samples (TW-1, TW-2, TW-11, and TW-18) are greater than the maximum concentration of 1 Bq L^{-1} set by the WHO, while the ^{226}Ra activity concentrations measured in the bottled drinking water samples are smaller than the quoted limit value. The activity concentrations of ^{228}Ra measured in all drinking water samples are higher than the maximum concentration of 0.1 Bq L^{-1} set by the WHO while the activity concentrations of ^{226}Ra and ^{228}Ra measured in all drinking water samples are higher than the maximum contaminant level of 0.185 Bq L^{-1} set by the EPA.

The activity level of ^{40}K (half-life: 1.28×10^9 y) is generally not considered in assessing radiological hazards on health from radionuclides in drinking water because potassium is a key element in regulating many body functions and the potassium content of the body is retained constant by a range of physiological processes [1]. It can be seen from Tables 2 and 3 that the activity concentrations of ^{40}K measured in the studied drinking water (tap and bottled) samples varied from 0.9 to 2.1 Bq L^{-1} with an average of 1.3 Bq L^{-1}. The average activity concentration of ^{40}K measured in TW is approximately 20% higher than that measured in the bottled drinking water samples. The activity concentration distribution of ^{40}K shows the near-normal distribution shown in Fig. 2. 93% and 7% of the activity concentrations of ^{40}K measured in all drinking water samples are in the range of 0.9–1.7 Bq L^{-1} and 1.8–2.1 Bq L^{-1} respectively. As can be seen in Table 4, the average activity concentration of ^{40}K in the drinking water samples is higher than that measured in drinking water samples consumed in Jordan (Irbid) while the average activity concentration of ^{40}K is lower than those measured in drinking waters consumed in Iran (Kermanshah) and Nigeria (Ogun). The activity concentrations of ^{40}K

Table 3 Descriptive statistical data on the activity concentration of radionuclides in drinking waters

	Activity concentration (Bq L^{-1})								
	TW			BDW			All drinking waters		
	^{226}Ra	^{228}Ra	^{40}K	^{226}Ra	^{228}Ra	^{40}K	^{226}Ra	^{228}Ra	^{40}K
Average	0.72	0.50	1.23	0.71	0.60	1.49	0.72	0.52	1.30
Standard error	0.05	0.04	0.05	0.05	0.07	0.11	0.04	0.03	0.05
Median	0.70	0.45	1.20	0.73	0.50	1.53	0.70	0.45	1.20
Standard deviation	0.30	0.21	0.27	0.14	0.23	0.36	0.27	0.22	0.31
Kurtoses	−0.17	4.09	−0.33	−0.67	1.33	−1.23	0.36	2.57	−0.40
Skewness	0.71	1.76	0.55	−0.37	1.26	0.07	0.76	1.52	0.59
Min	0.30	0.25	0.85	0.45	0.40	1.00	0.30	0.25	0.85
Max	1.50	1.25	1.90	0.90	1.10	2.05	1.50	1.25	2.05
Number of sample	31	31	31	10	10	10	41	41	41

Table 4 Comparison of the average concentration of radionuclides in drinking waters with the literature values and guidance levels

Country	Activity concentration (Bq L^{-1})			Reference
	^{226}Ra	^{228}Ra	^{40}K	
Jordan (Amman)	0.096	0.251	–	[30]
Egypt (Qena)	0.015	–	–	[31]
Italy	0.089	0.0048	–	[32]
Jordan (Irbid)	0.132	0.0494	0.194	[3]
Croatia	0.0013	0.0068	–	[33]
Iran (Kermanshah)	0.53	1.07	7.17	[12]
Canada	2	–	–	[5]
Nigeria (Ogun)	–	0.3	7.4	[34]
Turkey (Kastamonu)	0.72	0.52	1.3	This study
Guidance level of the WHO	1	0.1	10	[1]
Combined ^{226}Ra and ^{228}Ra	0.185		–	[29]

Table 5 Annual effective doses due to the consumption of water samples

Scenario	Water type	Annual effective dose (mSv y^{-1})			
		^{226}Ra	^{228}Ra	^{40}K	Total
First	TW	0.030	0.052	0.001	0.083
	BDW	0.030	0.062	0.001	0.093
	All Water samples	0.030	0.054	0.001	0.086
Second	TW	0.101	0.173	0.004	0.278
	BDW	0.099	0.205	0.005	0.309
	All Water samples	0.101	0.180	0.004	0.285
Third	TW	0.148	0.252	0.006	0.406
	BDW	0.144	0.300	0.007	0.451
	All Water samples	0.147	0.264	0.006	0.417

measured in all drinking water samples are on average five times lower than the maximum concentration of 10 Bq L^{-1}.

Assessment of radiological hazards

The annual effective doses due to ingestion were estimated for adults in three different scenarios according to the intake of the drinking waters. In the first scenario, the annual water consumption was taken as yearly consumption of bottled drinking water in Turkey (150 L) [36]. In the second scenario, the annual water consumption was taken as 500 L given in the UNSCEAR report [37]. In the third scenario, the annual water consumption was taken as 730 L recommended by the WHO (assuming consumption of 2 L per day) [1]. The average values of the ingestion annual effective doses estimated for different scenarios are presented in Table 5. For the first, second and third scenarios, the average of the total annual effective dose was estimated as 0.086, 0.285 and 0.417 mSv y^{-1}, respectively. The contribution of ^{226}Ra, ^{228}Ra and ^{40}K to the average total annual effective dose is 35%, 63% and 1%, respectively. ^{228}Ra, which is one of the most radiotoxic naturally occurring radionuclides, is the highest contributor to the total annual effective dose for all drinking water (tap and bottled) samples. It can be seen from Table 5 that total annual effective doses estimated for the second and third scenarios are higher than the individual dose criterion (IDC) or the total indicative dose of 0.1 mSv y^{-1} recommended by the WHO, Turkish legislation, Turkish standards, and EU directive. However, all annual effective doses are lower than the reference level of 1 mSv y^{-1} recommended for drinking water in the International Basic Safety Standard [38].

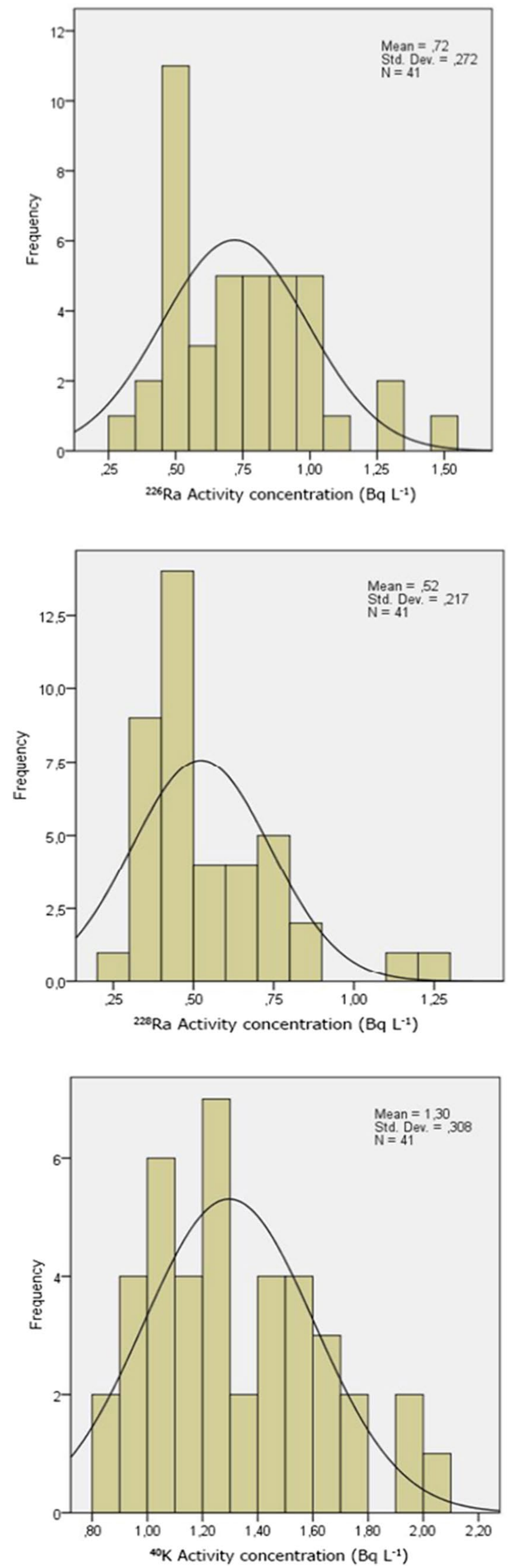

Fig. 2 Frequency distributions of the activity concentrations of ^{226}Ra, ^{228}Ra, and ^{40}K in water samples

Conclusions

This study provides the first detailed information on the determination of the natural radioactivity together with some physicochemical values of the drinking water in Kastamonu, which has been making efforts to become an important tourism destination in nature and religious tourism in recent years. The values of the pH, conductivity, TDS, TH, and activity of ^{226}Ra, ^{228}Ra, and ^{40}K of the total of forty-one drinking water (tap and bottled) were determined. The values of the measured physicochemical parameters are appropriate in quality according to national and international regulations. The average values of the measured physicochemical parameters for tap water are greater than those of the most consumed bottled drinking water in Kastamonu province. The activity concentration of ^{228}Ra measured in the studied drinking water samples is five times higher than the WHO recommended value of 0.1 Bq L^{-1}.

The total annual effective dose estimated for adults in the most extreme condition based on the drinking water consumption recommended by WHO was found as 0.42 mSv y^{-1}. This value is above the individual dose criterion of 0.1 mSv y^{-1} but it is below the reference level of 1 mSv y^{-1}. Both individual dose criterion and the reference level should not be interpreted as an acceptable dose or a dose limit where drinking water is not safe for consumption. Drinking water is a main requirement of life and the hazards of not having a drinking water source will be much bigger than consuming drinking water that does not meet the dose criterion. However, attempts should be made to reduce any exposure above the reference level to a lower level if possible [39].

Acknowledgements This study is carried out within the framework of a master thesis conducted at Kastamonu University and supported by The Scientific Research Projects Coordinator of Kastamonu University (Research Project codded of KÜ-BAP01/2018-23).

Declaration

Conflict of interest The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

References

1. WHO (World Health Organization), Guidelines for Drinking-water Quality, 4th edition (Incorporating the First Addendum), WHO Library Cataloguing-in-Publication Data NLM classification: WA 675, Geneva, 2017
2. Hossain I, Yussuf NM, Wagiran H (2016) Radioactive contamination in tap, rain and mine water in Malaysia by gamma-ray spectroscopy. J Optoelectron Biomed Mater 8(2):39–43
3. Al-Shboul KF, Alali AE, Batayneh IM et al (2017) Radiation hazards and lifetime risk assessment of tap water using liquid scintillation counting and high-resolution gamma spectrometry. J Environ Radioact 178:245–252
4. Kale AB, Bandela NN, Kulkarni JA (2018) Radiological and chemo-toxicological risk assessment of naturally occurred uranium in ground water from Aurangabad District of Maharashtra. Int J Environ Sci 3:49–55
5. Chen J, Cooke MW, Mercier JF (2018) Review of natural radionuclides in Canadian drinking water (1975–16). Radiat Prot Dosim 179(1):26–36
6. Karabıdak SM, Kaya A, Kay S (2019) Cancer risk analysis of some drinking water resources in Gümüşhane, Turkey. Sustain Water Resour Manag 5:1939–1949
7. El-Gamal H, Sefelnasr A, Salaheldin G (2019) Determination of natural radionuclides for water resources on the West Bank of the Nile River, Assiut Governorate. Egypt Water 11(311):1–13
8. Alomari AH, Saleh MA, Hashim S et al (2019) Measurement of gross alpha and beta activity concentration in groundwater of Jordan: groundwater quality, annual effective dose and lifetime risk assessment. J Water Health 17(6):957–970
9. Opoku-Ntim I, Gyampo O, Andam AB (2019) Risk assessment of radon in some bottled water on the Ghanaian market. Environ Res Commun 1:1–8
10. Turhan Ş, Zriba NAEM, Taşkın H et al (2019) Radiochemical analysis of bottled drinking waters consumed in Turkey and a risk assessment study. Microchem J 149:1–7
11. Sekudewicz I, Gąsiorowski M (2019) Determination of the activity and the average annual dose of absorbed uranium and polonium in drinking water from Warsaw. J Radioanal Nucl Chem 319:1351–1358
12. Parhoudeh M, Khoshgard K, Zare MR et al (2019) Natural radioactivity level of ^{226}Ra, ^{232}Th, and ^{40}K radionuclides in drinking water of residential areas in Kermanshah province, Iran using gamma spectroscopy. Iran J Med Phys 16:98–102
13. Semerjian L, Alrajaby H, Naaz N et al (2020) Age-dependent effective ingestion dose estimations and lifetime risk assessment for selected radionuclides (^{40}K and ^{3}H) in bottled waters marketed in United Arab Emirates. Chemosphere 249:1–5
14. Todorović N, Nikolov J, Stojković I et al (2020) Radioactivity in drinking water supplies in the Vojvodina region, Serbia, and health implication. Environ Earth Sci 79(162):1–10
15. Turhan Ş (2020) The natural radioactivity in drinking water by gross alpha and beta measurements and radiological quality assessment. Radiochim Acta 108:491–498
16. El-Badry BA, Al-Naggar TI, Khouqeer GA (2020) Monitoring the levels of radon and toxic elements pollutants in bottled drinking water. Int J Radiat Res 18(3):427–435
17. Kinahan A, Hosoda M, Kelleher K et al (2020) Assessment of radiation dose from the consumption of bottled drinking water in Japan. Int J Environ Res Public Health 17:1–12
18. Nagabhushana SR, Sunilkumar SS et al (2020) Study on activity of radium, radon and physicochemical parameters in ground water and their health hazards around Tumkur industrial area. J Radioanal Nucl Chem 323:1393–1403
19. Öge TÖ, Özdemir FB (2020) Assessment of radioactivity from groundwater samples from selected areas of Western Black Sea Region. Turkey Radiochim Acta 108(6):499–508
20. Khaled SD, Khaled AM, Shaban RH et al (2020) Measurement of ^{222}Rn concentration levels in drinking water samples from Qena city (Egypt) and evaluation of the annual effective doses. Int J Radiat Res 18(2):227–233
21. Kurnaz A, Çetiner M (2016) Exposure assessment of the radon in residential tap water in Kastamonu. Int J Radiat Res 14(3):245–250
22. Duran C (2017) Local distribution of the temperature and precipitation in Kastamonu province and surrounding. J Int Soc Res 10(52):509–517 (**in Turkish**)
23. Sultan DAO, Turhan Ş, Kurnaz A et al (2020) Investigation of natural radionuclide and essential metal contents of ancient wheat einkorn (Triticum monococcum L.) grown in Turkey. Radiochim Acta (2020).
24. ICRP (International Commission on Radiological Protection). Age-dependent doses to members of the public from intake of radionuclides: Part 5, compilation of ingestion and inhalation dose coefficients. ICRP Publication 72 (1996), Oxford, United Kingdom: Pergamon Press.
25. Adewoye SO, Adewoye AO, Opasola OA et al (2013) Physicochemical parameters and heavy metal analyses of water samples from Hand Dug wells In Gambari, Ogbomoso, Oyo State. IOSR J Environ Sci Toxicol Food Technol 5(1):22–30
26. Journal O (2013) Regulation on water intended for human consumption 28580:07 (**in Turkish**)
27. Turkish Standard (TS) 266. Water intended for human consumption. Turkish Standards Institute, Ankara (2005) (**in Turkish**).
28. European Union (EU) Directive. EU Council directive 98/83/EC of 15 November 1998 on the quality of water intended for human consumption. Off. J. Eur. Communities L 330/32, 05.12.1998.
29. United States Environmental Protection Agency (US EPA). Drinking Water Regulation, Code of Federal Regulations, 40 Part 141, USA, (1986).
30. Al-Amir SM, Al-Hamarneh IF, Al-Abed T et al (2012) Natural radioactivity in tap water and associated age-dependent dose and lifetime risk assessment in Amman, Jordan. App Radiat Isot 70:692–698
31. Shweikani R, Raja G (2015) Natural radionuclides monitoring in drinking water of Homs city. Radiat Phys Chem 106:333–336
32. Jia G, Torri G, Magro L (2009) Concentrations of ^{238}U, ^{234}U, ^{235}U, ^{232}Th, ^{230}Th, ^{228}Th, ^{226}Ra, ^{228}Ra, ^{224}Ra, ^{210}Po, ^{210}Pb and ^{212}Pb in drinking water in Italy: reconciling safety standards based on measurements of gross α and β. J Environ Radioact 100:941–949
33. Rozamarić M, Rogić M, Benedik L et al (2014) Radiological characterization of tap waters in Croatia and the age dependent dose assessment. Chemosphere 111:272–277
34. Usikalu MR, Olawole CO, Joel E (2016) Assessment of natural radionuclides levels in drinking water from Ogun State, Nigeria. Jurnal Teknologi (Sciences & Engineering) 78:25–29
35. IAEA Technical Report. Analytical methodology for the determination of radium isotopes in environmental samples. IAEA Analytical Quality in Nuclear Applications No. IAEA/AQ/19. International Atomic Energy Agency, Vienna (2010).
36. SUDER, 2020. Packaged Water Manufacturers Association (Ambalajlı su Üreticileri Derneği) Web page: https://suder.org.tr/. (**in Turkish**)
37. UNSCEAR 2000. Sources and effects of ionizing radiation. United Nations Scientific Committee on the Effects of Atomic Radiation, United Nations Publication, New York, USA (2000).
38. International Atomic Energy Agency (IAEA) (2014). Radiation protection and safety of radiation sources: International Basic Safety Standards. IAEA Safety Standards Series No. GSR Part 3, (2014) Vienna.

39. WHO (World Health Organization), Management of radioactivity in drinking-water. Licence: CC BY-NC-SA 3.0 IGO. Geneva, (2018).