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Araştırma Makalesi / Research Article

Radioactivity Levels in Soil And Drinking Water Samples Collected from Andon Region (Rize Province, Turkey)

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Abstract

In the present study, radionuclide and radon concentrations were measured in drinking water samples collected in Andon springs and Rize Andon Drinking Water Treatment Facility located at Rize province in Eastern Black Sea Region in Turkey and radionuclide concentrations were measured in soil samples collected in the same regions. Radionuclide activity concentrations in soil and drinking water samples were measured with gamma ray spectrometry, and ^{238}U, ^{232}Th, ^{40}K and ^{137}C activity concentrations were determined. Annual effective dose equivalents were calculated based on the measured results. Radon concentration measurements for the drinking water samples collected in the region were conducted with a radon gas analyzer (Alpha Guard PQ 2000PRQ). All measured radon gas levels and radionuclide values were found to be among the limit values recommended by international organizations.

Keywords: Radioactivity, Soil, Drinking water, Radon, Rize.

Andon Bölgesi'nden Alınan Toprak ve İçme Suyu Örneklerinde Radyoaktivite Düzeyleri (Rize İli, Türkiye)

Öz

Türkiye'nin Doğu Karadeniz Bölgesi'ndeki Rize iline bağlı Andon içmesi ve aynı bölgede bulunan "Rize Andon İçme Suyu Arıtma Tesisinden" alınan içme suyu örneklerinde ve bu kaynakların çevresinden toplanan toprak örneklerinde radyoaktivite ve radon konsantrasyonları ölçülmüştür. Toprak ve içme suyu örneklerinde radyonüklid aktivite konsantrasyonları gama ışını spektrometresi ile ölçülüp, ^{238}U, ^{232}Th, ^{40}K ve ^{137}C aktivite konsantrasyonları belirlenmiştir. Ölçüm sonuçları dikkate alınarak yıllık etkili doz eşdeğerleri hesaplanmıştır. Bu bölgeden toplanan içme sularındaki Radon konsantrasyonunun ölçümü, radon gazı analiz cihazı (Alpha Guard PQ 2000PRQ) kullanılarak gerçekleştirilmiştir. Ölçülen tüm radon gazı seviyeleri ve radyonüklid değerlerinin uluslararası kuruluşlar tarafından önerilen sınır değerler arasında olduğu bulunmuştur.

Anahtar Kelimeler: Radyoaktivite, Toprak, İçme suyu, Radon, Rize.

1. Introduction

It is important to determine radioactivity levels in the air, water and soil since human beings and other living organisms have been exposed to cosmic rays originated in the universe for millions of years and the radiation emitted from natural radioactive substances on earth. The radioactive materials that are available in air, water, all vegetable and animal nutrients are taken through respiration and digestion mechanisms in small amounts and these materials are accumulated in various organs over time. Since humans are affected by cosmic rays and natural and artificial radioactive materials on earth, the human body is naturally exposed to both internal and external radiation. Approximately 12% (0.29 mSv) of natural radiation dose exposure is induced by radionuclides in nutrients and drinking water. Thus, drinking water and water consumption should be safe for human health. This guarantee could be achieved by precisely limiting the chemical and radioactivity properties in drinking water (UNSCEAR, 2008; Camgöz et al., 2010).

Radioactivity presence in water occurs through two mechanisms. The first is through the contact between the water and the soil and rocks on the pathway of the water, which dissolves the radioactive substances on the soil and rocks, and the second is the mixing of radionuclides into water due to waste discharge or accidents. In particular, the groundwater, otherwise called deep-circulation water, is more active than surface waters since they interact with volcanic and granite rocks that are found in the lower layers. Thus, groundwater radionuclides enter the food chain via the ground or surface water used for irrigation of crops or consumed directly by humans through the ground water used as drinking water. ^{238}U, ^{234}U, ^{232}Th, ^{226}Ra, ^{210}Pb, ^{210}Po, and ^{40}K etc. are radionuclides with long half-lives and could be digested with drinking water. They might lead to the intake of a radiation dose that could threaten human health due to the irradiation of the kidney, liver, bladder, bones and bone marrow. Furthermore, inhalation of radon (^{222}Rn) gas produced by alpha degradation of radium (^{226}Ra) in drinking water leads to lung cancer risk over time through the biological damages caused in the lung tissue by radon and short half-life degradation products (^{218}Po, ^{214}Pb, ^{214}Bi and ^{214}Po). Thus, since the microorganisms in drinking water could lead to risks that could threaten human health due to long exposure to radiation, albeit in small doses, the quality of drinking water should be monitored continuously. Therefore, this necessitates the measurement, monitoring and limitation of radioactivity in drinking water and ground and surface waters, which are used as drinking water sources (El-Arabi, 2007; Janković et al., 2012; Chau and Michalec, 2009; El-Mageed et al., 2013; Kobya et al., 2010; UNSCEAR, 2000).

Natural and artificial radionuclide analyzes of air, soil and plants (Kurnaz et al., 2007; Görür et al., 2012; Keser et al., 2011) were carried out in the Rize Province and are being carried out. In addition, radon concentration studies were performed in home and outside air environments (Özen et

al., 2018). Also, radionuclide activity measurements, total alpha and total beta analyzes were performed in drinking and natural spring waters (Kobya et al., 2010; Kobya et al., 2011) in the same Province.

Andon region is known as an important source for mineral-enriched spring water. Andon springs is visited by a high number of people due to its alleged medicinal value. Rize Andon Drinking Water Network, which provides drinking water for Rize province, is located in the same area.

The present study aimed to determine the radioactivity in water and soil samples collected in Rize Andon Drinking Water Network, which provides drinking water for Rize province, and Andon springs, and whether the radioactivity levels were compliant with the limit values determined by international organizations.

2. Materials and Methods

2.1. Study Area

Rize Andon Drinking Water Network is located in Rize province Andon region. The facility provides drinking water to Rize urban center and other districts. The facility treats the water supplied by Taşlı, Poşut and Ilıca streams (URL-1, 2015).

Spring water is supplied by Andon springs located at Küçükçayır (Andon) village in Rize Province. The region consists of Upper Cretaceous rhyodacite, dacite lava and pyroclastic, basalt, andesite lava-pyroclastic complex, topped by granite-monzonite-granodiorite and microgranite intrusive (Figure 1). The Eastern Pontide is main metallogenetic province in the coastal region of the southern east Black Sea and is subdivided into two different zones, northern and eastern. The studied area, Andon Springs (Küçükçayır, Rize city, Turkey) is located at the northern zone of the eastern Pontides consists of Upper Cretaceous volcanic and igneous rocks. From oldest to youngest, Upper Cretaceous rhyodacite, dacite, basalt, andesite-pyroclastic melange and granite-monzonite-granodiorite-microgranite intrusions are present in the studied area. The Eastern Pontide is main metallogenetic province in the coastal region of the southern east Black Sea and is subdivided into two different zones: northern and eastern. The studied area, Andon Springs (Küçükçayır, Rize city, Turkey) is located at the northern zone of the eastern Pontides consists of Upper Cretaceous volcanic and igneous rocks. From oldest to youngest, Upper Cretaceous rhyodacite, dacite, basalt, andesite-pyroclastic mélange and granite-monzonite-granodiorite-microgranite intrusions are present in the studied area (Göçmez et al., 2007).

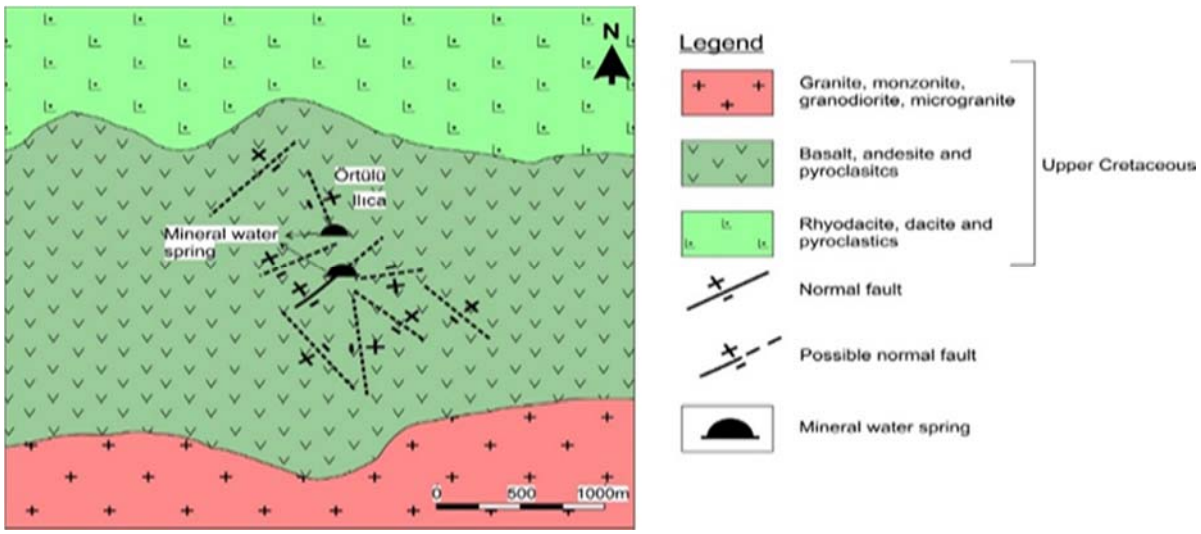

Figure 1. Geological map of Andon and its surroundings.

2.2. Gamma Spectrometric Analyses of Soil and Water Samples

The soil samples were collected at a depth of 15-20 cm. The samples were placed in pre-labeled, clean, sealed nylon bags and transferred to the laboratory. The collected samples were initially classified and dried at room temperature. Then, the samples were grinded and sieved. Soil samples were sieved through a 80 mesh screen. Each sample was put in a gas-tight, radon-impermeable cylindrical polyethylene plastic container (6 cm diameter and 5 cm height) for the concentration measurements of radionuclides. Thus, the radioactive balance was ensured between the ^{238}U and ^{232}Th and their decay products and the samples were prepared for counting.

Ten liters of water was obtained from each water source and placed in pre-sterilized, labeled plastic bottles and transferred to the laboratory. Water samples were transferred into 10-liter clean glass beakers and allowed to evaporate on a 50-60°C heater. Evaporation was maintained until the water volume reached 100 ml. During evaporation, a few drops of HCl were added into the beaker to prevent the substances to adhere to beaker walls. The remaining volume was then transferred into tared Marinelli vessels and stored for about one month to reach internal equilibrium. Then, the samples were counted in a gamma spectrometer device for 80,000 s.

Gamma spectrometry was employed for determining the radioactivity of the studied samples. The spectrometry system consisted of an HPGe detector (CANBERRA model GC 1519) with a relative efficiency of 15%. To acquire data for analysis, a multichannel analyser (MCA) with an inbuilt power supply, preamplifier, and amplifier, was installed on a personal computer. The resolution of the system was 1.9 keV at the 1332.5 keV peak of ^{60}Co. Spectral analysis was performed using the Genie 2000 software that was obtained from CANBERRA.

The activity concentrations for the natural radionuclides in the measured samples were computed using the Equation 1;

$$C_s(\text{Bq kg}^{-1}) = N_a/\varepsilon P_r M_s t \quad (1)$$

where N_a is the net counting rate (peak area) of the gamma ray, ε is the detector efficiency of the specific gamma ray, P_r is the absolute transition probability of gamma decay for related radionuclide, M_s is the mass of the sample, M_s is taken as a volume (l) for water samples, M_s is the mass of the sample (kg) for soil samples, and t is the counting time (Çevik et al., 2007).

2.3. Absorbed dose rate and annual effective dose for soil

If the radionuclide activities of soil are known, the rate of absorption at a height of one meter from the ground could be determined. Using the radionuclide activities obtained in soil sample analysis, Gamma Dose (D) values in the samples were calculated with Equation (2) in the present study (UNSCEAR 2000).

$$D\ (\text{nGy/s}) = (0.462 \times {}^{238}U) + (0.604 \times {}^{232}Th) + (0.0417 \times {}^{40}K) \quad (2)$$

The annual effective dose equivalent (AEDE), that is, the that individuals are exposed to annually, the dose that they are exposed to the radiation emitted by different radiation sources could be found using the calculated gamma doses. Equation (3) was used to calculate the annual effective dose equivalent

$$\text{AEDE}(\mu\text{Sv.y}^{-1}) = \text{Dose ratexDCCxOccupation factorxTime} \quad (3)$$

The coefficients in Equation (2) were obtained from UNSCEAR (UNSCEAR, 2000). The dose conversion coefficient (DCC) in Equation (3) was taken to be 0.7 $Sv.Gy^{-1}$. The occupation factor was taken to be 0.2, assuming that people spend 20% of their time outdoors and 80% indoors. The time was taken to be 8760 seconds per year.

2.4. Annual effective dose for water

In order to calculate the harmful biological effects on an individual exposed to radiation, the equivalent of a measurable radiation dose is needed. Thus, the standards for the radiation dose exposure by the public and those work with radiation are expressed for certain periods. The annual effective dose equivalents in water samples are found using Equation 4 (Amrani, 2002).

$$\mathrm{AEDE}(\mu\mathrm{Sv\ year}^{-1}) = \mathrm{C_R}.\mathrm{I_A}.\mathrm{E_D} \quad (4)$$

Where, AEDE is the annual effective dose equivalent ($Sv.y^{-1}$), C_R is the annual water consumption ($l.y^{-1}$), I_A is the radionuclide activity ($Bq.l^{-1}$), and E_D is the dose conversion factor ($Sv.Bq^{-1}$). The dose conversion factors used for the calculation were taken from the World Health Organization publication (WHO, 2006) and were equal to 2.8×10^{-4}, 2.3×10^{-4} and 1.3×10^{-5} $mSv.Bq^{-1}$ for ^{226}Ra, ^{232}Th and ^{137}Cs, respectively. And for ^{40}K, intake was calculated using the conversion factor of 6.2×10^{-6} $mSv.Bq^{-1}$. Assuming that every adult drinks 2 $l.day^{-1}$, the annual effective doses caused by intake of the ^{226}Ra, ^{232}Th, ^{137}Cs and ^{40}K isotopes were calculated (Kobya et al., 2011). The overall annual effective dose from all measured radionuclides was much below the WHO (2011) recommended reference level of 0.1 $mSv.y^{-1}$ for all water samples.

2.5. Measurement of Water Radon Concentration with AlphaGUARD

For radon measurement, the water samples were obtained by dipping 500 ml glass bottles with a rubber stopper into the pool and the bottles were closed while in the pool and taken out and the bottles were wrapped with aluminum foil and transferred to the laboratory. Then, the iron cage above the filter of the device was removed and the apparatus required to attach the hoses to the device was mounted for water measurements. Then, the glass water measuring cups in the AquaKit box of the device were connected to the device with AlphaPump plastic hoses. The water was removed from the sample beaker using the plastic syringes in the aqua kit and transferred to glass measurement containers. After the connection was established, FLOW was adjusted to 1 minute using the AlphaGUARD menu to record the radon concentration measured in a period of 1 minute.

The sample radon concentration is calculated the Equation 5.

$$C_{water} = [(C_{air}(V_{system} - V_{sample})/V_{sample}) + k) - C_o]/1000 \quad (5)$$

where C_{water} is the radon concentration in the water sample ($Bq.l^{-1}$), C_{air} the radon concentration in the set-up after expelling radon from water ($Bq.m^{-3}$), C_0 the background radon concentration ($Bq.m^{-3}$). V_{system} the interior volume of the measurement set-up (ml), V_{sample} the volume of the water sample (ml) and k, the radon distribution coefficient (Duran et al., 2017).

2.6. Estimated Doses Due to The Radon

Radon and its decomposition products create an additional radiation dose in the digestive tract when individuals drink water with high radon concentrations. The annual effective dose for the population due to drinking water with radon is calculated with Equation 6 (Nikolov et al., 2012).

$$\mathrm{AEDE} = \mathrm{K.C.KM.t} \quad (6)$$

Where, the annual effective dose (AEDE) equivalent due to drinking water with radon is ($Sv.y^{-1}$), dose conversion factor for K, ^{222}Rn is 10^{-8} $Sv.Bq^{-1}$ for adults and 2.10^{-8} $Sv.Bq^{-1}$ for children, C is ^{222}Rn concentration in water ($Bq.l^{-1}$), KM is the daily water consumption (2 $l.day^{-1}$), and t is the consumption period (365 days for l year) (Todorovic et al., 2012).

3. Results and Discussion

3.1. Radionuclide Activities in Soil and Estimated Doses

The activity concentrations of radionuclides in soil samples from the Andon vicinity of Turkey were determined by gamma spectrometry. Totally 13 soil samples were collected from Andon vicinity. Activity concentrations of ^{238}U, ^{232}Th, ^{40}K, and ^{137}Cs were measured, and the corresponding mean values are listed in Table 1. Using the measured activity concentrations, dose rates and annual effective doses were estimated and are listed in the same table.

As can clearly be seen from the table, the mean concentrations found in this study are 30 $Bq.kg^{-1}$ for ^{238}U, 14 $Bq.kg^{-1}$ for ^{232}Th and 404 $Bq.kg^{-1}$ for ^{40}K. The world's mean values of ^{238}U, ^{232}Th and ^{40}K in soil are 32, 45 and 420 $Bq.kg^{-1}$, respectively (UNSCEAR 2000).

Table 1. Activity concentration of radionuclides in soil of Andon vicinities and estimated dose rates.

Sample	^{238}U ($Bq.kg^{-1}$)	^{232}Th ($Bq.kg^{-1}$)	^{137}Cs ($Bq.kg^{-1}$)	^{40}K ($Bq.kg^{-1}$)	D ($nGy.s^{-1}$)	AEDE ($\mu Sv.y^{-1}$)
A- 1	24±5	5±4	5±2	608±42	39	48
A- 2	17±4	12±3	10±2	384±29	31	38
A- 3	14±3	13±5	43±7	439±38	33	41
A- 4	31±3	10±4	29±2	402±39	37	46
A- 5	50±7	12±5	101±4	491±44	35	43
A- 6	54±5	19±9	74±4	505±43	57	70
A- 7	13±5	18±5	67±3	533±44	39	48
A- 8	28±3	9±4	14±2	406±34	35	43
A- 9	55±6	17±5	64±4	486±44	56	69
A- 10	34±2	17±4	0	207±7	35	43
A- 11	24±2	12±2	81±4	220±8	27	34
A- 12	18±2	16±3	28±3	304±10	31	38

A- 13	26±2	21±3	17±2	276±10	36	44
Average	30±4	14±5	41±3	404±30		

3.2. Radionuclide Activities in Waters and Estimated Doses

Gamma spectrometric analyzes conducted on the water samples collected in Andon Spring water (A_1), any drinking water in Andon (A_2) and the drinking water (Main Water Network) (A_3) determined the radionuclide activity concentrations and the results are presented in Table 2. Also, the calculated effective doses are given in Table 2.

Table 2. Results of drinking-water measurements.

Radionuclides	Water Samples A_1 Spring water		A_2 Drinking water		A_3 Water Network	
	Radionuclides ($mBq.l^{-1}$)	AEDE ($\mu Sv.y^{-1}$)	Radionuclides ($mBq.l^{-1}$)	AEDE ($\mu Sv.y^{-1}$)	Radionuclides ($mBq.l^{-1}$)	AEDE ($\mu Sv.y^{-1}$)
^{238}U	57±7	0.08	412±20	0.56	71±5	0.09
^{232}Th	152±20	1	35±6	0.24	31±6	0.2
^{137}Cs	20±2	0.08	34±7	0.013	24±9	0.09
^{40}K	3980±310	0.7	186±7	0.034	78±3	0.013

3.3. Radon Concentrations in Waters and Estimated Doses

The radon gas was measured in spring water with AlphaGUARD radon monitor water probe. Radon gas measurements were conducted twice on Andon Spring water in the summer and winter. The measurement results are presented in Table 3.

Table 3. ^{222}Rn activity concentrations and the effective doses from Andon drinking waters

Water type	C ($Bq.l^{-1}$)	Effective Dose ($\mu Sv.y^{-1}$)
A_1 Spring water (summer)	2.21±0.60	0.198
A_1 Spring water(winter)	3.84±1.20	0.346
A_2 Drinking water	3.72±0.80	0.334
A_3 Main Water Network	0.78±0.20	0.070

4. Conclusions and Recommendations

Radionuclide analysis, radon gas measurement water samples collected from Andon springs and the drinking water network located in Andon and supplied drinking water for Rize province and radionuclide analysis was conducted on soil samples collected in the area.

Comparison of drinking water network and Andon spring water radon gas concentrations obtained in the present study with the findings of other studies conducted both in Turkey and abroad are presented in Table 4.

Table 4. Water Radon Concentrations in Certain Drinking Water Networks in Turkey and Abroad

Province	Water	Radon $Bq.l^{-1}$	References
Turkey(Zonguldak)	Drinking water	0.32-21.30	(Koray et al., 2014)
Turkey (Bursa)	Tap water	0.91-12.58	(Tarım et al., 2011)
Turkey(Seydişehir)	Spring water	1.85-99.27	(Erdoğan et al., 2017)
Turkey(Kastamonu)	Tap water	0.31-13.14	(Yalçın et al., 2011)
India	Drinking water	0.85-60.74	(Srinivasa et al., 2015)
China	Drinking water	3.68-29	(Wu et al., 2014)
Rize (Andon)	Spring water	2.21-3.84	**Present Study**
Rize (Andon)	Drinking water	0.78-3.72	**Present Study**

Analysis of the results in Table 4 shows that the highest radon activity concentrations are observed in the water of Turkey (Seydişehir), India. Measurements revealed that drinking water radon gas levels were lower than EPA limits 11 $Bq.m^{-3}$ (EPA, 1991).

Based on the drinking water and soil sample radionuclide analysis results, it was determined that the annual effective dose calculated with ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs activities were below the global average and did not pose any danger to human health. Furthermore, the annual effective dose equivalent calculated with the radon concentrations obtained with the calculations conducted in the present study was compared to the annual effective dose equivalent limit of 0.1 $mSv.y^{-1}$ determined by the WHO (World Health Organization), and it was observed that the values determined for all drinking water networks were below that limit and did not pose any danger to human health.

The traces of artificial radioactivity product ^{137}Cs were determined in the collected drinking water and soil samples which might be as a result of the radioactive leakage caused by Chernobyl disaster in 1986, albeit in low amounts.

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