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Research Article

Determination of the Radioactivity in the Turkish Tea Samples

Türk Çayı Örneklerinde Radyoaktivitenin Belirlenmesi

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Abstract

Natural and artificial radioactivity concentrations of the Turkish tea samples were determined using 20% n-type HPGe gamma-ray spectrometer. ^{238}U, ^{226}Ra, ^{232}Th, ^{228}Ra, ^{228}Th, ^{40}K, ^{210}Pb, ^{7}Be and ^{137}Cs radioactivity concentrations were measured. ^{238}U, ^{226}Ra, ^{232}Th, ^{228}Ra, ^{228}Th radioactivity concentrations were MDA. ^{7}Be, ^{210}Pb, ^{40}K and ^{137}Cs radioactivity concentrations mean values were found 30.8 ± 2.3, 49.4 ± 4.4, 485.4±38.6 and 38.8±2.4 Bq kg^{-1}, respectively.

Keywords: Artificial radionuclides, Gamma spectrometry, Natural radionuclides, Radioactivity concentration, Tea sample

Öz

Türk çayı örneklerinin doğal ve yapay radyoaktivite konsantrasyonları %20 n-tipi HPGe dedektör kullanılarak belirlenmiştir. ^{238}U, ^{226}Ra, ^{232}Th, ^{228}Ra, ^{228}Th, ^{40}K, ^{210}Pb, ^{7}Be ve ^{137}Cs radyoaktivite konsantrasyonları ölçülmüştür. ^{238}U, ^{226}Ra, ^{232}Th, ^{228}Ra, ^{228}Th radyoaktivite konsantrasyonları MDA bulunmuştur. ^{7}Be, ^{210}Pb, ^{40}K ve ^{137}Cs radyoaktivite konsantrasyonlarının ortalama değerleri sırasıyla 30.8 ± 2.3, 49.4 ± 4.4, 485.4±38.6 ve 38.8±2.4 Bq kg^{-1} bulunmuştur.

Anahtar Kelimeler: Yapay radyonüklit, Gama spektrometri, Doğal radyonüklit, Radyoaktivite konsatrasyonu, Çay numunesi

1. Introduction

Turkey is the world's fifth tea producer. According to the Turkish Statistic Institute, tea production in Turkey is 1.327.934 MT at 2015 (TÜİK 2015). At the same time, tea is the major hot and cold beverage consumed by Turkish people. The type of Turkish tea is small-leafed type known as Camelia sinensis. All of the tea consumed by Turkish people grows in Karadeniz region of Turkey. Karadeniz region is located along the coasts of the Black Sea.

The knowledge of radiation levels and radionuclides in the foodstuff is important for assessing the effects of radiation exposure due to both natural and artificial radioactivity. The radiological importance of these radionuclides is due to the gamma-ray exposure of the body via digestion. Radioactivity in the food comes from naturally occurring radionuclides and artificial radionuclides.

Naturally occurring radionuclides are ^{238}U series, ^{232}Th series and ^{40}K (IAEA 2005). Artificial radiations are generated in the past by nuclear tests and the nuclear accidents. In 1986 northeastern part of Turkey has affected by the Chernobyl nuclear power reactor accident. The radionuclide release following the Chernobyl accident caused radionuclide deposition and a significant increase of artificial radionuclide. The radionuclides that caused contamination were ^{137}Cs, ^{134}Cs, and ^{131}I. Among these radionuclides, ^{137}Cs with a half-life of 30.05 y has remained in the ecosystems many years after the accident.

As a result of the spallation reactions of charged particles on oxygen and nitrogen, ^{7}Be is generated in the atmosphere. ^{7}Be is fallout radionuclide. ^{7}Be has 477.6 keV gamma energy and 53.22 days half-life (Gilmore 2008). ^{7}Be radionuclide falls to the ground with wet deposition (Kim et al. 2016, Bourcier et al. 2014). According to the Turkish State Meteorological Service annual average precipitation in the tea production area of the Karadeniz Region are 727.2 mm (MGM 2015). This type of rainy climate enhances deposition probability of the ^{7}Be radionuclide with rainfall.

Radionuclides pass to the human body through the food chain. Root uptake is the initial and significant step of radionuclide transfer from soil to the plant in the food

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chain (IAEA 2009). In the past, translocation and percent distribution of ^{137}Cs in different parts of the tea plants are investigated from the foliar absorption and root uptake. In addition, the natural depuration rates of the radionuclides are investigated (Topcuoğlu et al. 1997).

The aim of this work is to determine the radioactivity levels of ^{238}U, ^{226}Ra, ^{232}Th, ^{228}Ra, ^{228}Th, ^{40}K, ^{210}Pb, ^{7}Be and ^{137}Cs of Turkish market tea samples. For this purpose radioactivity concentrations of the tea samples which produced in the Karadeniz Region were measured. In the literature, there are studies about the natural and artificial radioactivity concentration of the Turkish tea samples (Topcuoğlu et al. 1997, Kılıç et al. 2006, Görür et al. 2011). However, there are no studies concerning the ^{7}Be and ^{210}Pb radioactivity concentrations of the Turkish tea samples.

2. Material and Methods

2.1. Sampling and Sample Preparation

Samples which were produced in the Karadeniz region were collected from the tea and herbal tea markets. The samples which new season and new harvest were preferred. The reason was ^{7}Be radionuclide has relatively short half-life. Samples were separately labeled and brought into the laboratory. In order to remove moisture, tea samples were dried in a drying oven at 80°C until constant mass was obtained. The samples were ground and homogenized in the laboratory. Adequate samples were put into cylindrical plastic analysis containers. Plastic analysis containers had a 6 cm diameter and 5 cm height. Then samples were weighed and sealed with parafilm to prevent the escape of radon gas. Samples were kept for 30 days for secular equilibrium of ^{226}Ra decay products before the measurements. Then each sample was measured and the values were given in Bq kg^{-1} dry weight.

2.2. Radioactivity Measurement

Radioactivity measurements were performed by using a gamma spectrometer. The spectrometer were n-type reverse electrode closed-end coaxial high-purity germanium detector. The detector has 20% relative efficiency and 46:1 peak-to-compton ratio. The energy resolutions of the detector are 1.80 keV for ^{60}Co at 1332.5 keV and 0.97 keV for ^{57}Co at 122 keV. Before the measurements, energy calibration was done by using peaks of ^{241}Am, ^{137}Cs and ^{60}Co radionuclides of the standard point radioactive source.

The efficiency calibration of gamma spectrometry system was performed using 79829-839 coded certified standard volume source. The efficiency calibration source had vegetation matrix and 13 radionuclides that have the energy range of 59.5–1836.1 keV. Before the measurements an empty plastic sample container was counted in the same manner as the samples for the determination of the background effects. After measurements and subtraction of the background, the activity concentrations were determined. The activity concentrations of the samples were determined from radionuclide's own energies or gamma-ray photopic of their decay products.

- The activity concentration of ^{238}U was calculated from 63.3 keV gamma-ray energy.
- The activity concentration of ^{226}Ra was calculated from 295.2, 351.9 keV gamma-ray energies of ^{214}Pb and 609.3 keV of ^{214}Bi.
- The activity concentration of ^{232}Th, ^{228}Th and ^{228}Ra were calculated from 238.63 keV of ^{212}Pb, 583.2 keV of ^{208}Tl and 338.4, 911.2 keV of ^{228}Ac gamma-ray energies.
- The activity concentration of ^{40}K was determined by using its own energy of 1460.8 keV.
- The Activity concentration of ^{137}Cs was calculated from 661.7 keV photopic energy.
- The activity concentration of ^{210}Pb was calculated from 46.5 keV photopic energy.
- The activity concentration of ^{7}Be was calculated from 477.60 keV gamma-ray energy. ^{7}Be has 53.3 days half-life. Therefore activity correction factors during the measurement period applied.

A sample spectrum is shown in Figure 1. The activity concentrations of the samples are calculated by the following formula:

$$A = \frac{N}{\varepsilon_\gamma P_\gamma t M} \quad (1)$$

where N corresponds the net peak area of gamma-ray energy, ε_γ denotes the absolute efficiency, P_γ is the gamma-ray yield per decay, t and M denotes the counting time and sample mass, respectively. Minimum detectable activity (MDA) calculations were performed by the following formula (Currie 1968):

$$MDA = \frac{(1.64)\sigma_n}{\varepsilon P t w} \quad (2)$$

where σ_n stands for standard deviation of the background in the region, ε is absolute efficiency, P is the emission probability of gamma decay, t and w denote measurement time and weight of the dried sample, respectively.

3. Results

The activity concentrations of ^{238}U, ^{214}Pb, ^{214}Bi, ^{226} Ra, ^{40}K and ^{210}Pb in Turkish tea samples are given in the Table 1. A less than sign (<) was used to indicate the below MDA values of the detector. As can be seen in the Table 1 ^{238}U, ^{214}Pb, ^{214}Bi and ^{226}Ra activity concentrations are below MDA value.

^{210}Pb activity concentration ranges were found to be 11.1 ± 0.9 – 81.9 ± 6.6 Bq kg^{-1}. Sample 10 has highest value of the ^{210}Pb activity concentration. Sample 4 has smallest value of the ^{210}Pb activity concentration. Harb (2007) found that ^{210}Pb activity concentrations ranged from 20.6 ± 1.5 to 30.5 ± 2.9 Bq kg^{-1} for the Egyptian market tea samples. In another study Desideri et al. (2011) found that activity concentrations ranged from <10.0 and 58.9 Bq kg^{-1}.

^{40}K activity concentrations ranges were found to be 428.7 ± 32.2 – 681.9 ± 51.4 Bq kg^{-1}. Sample 11 has highest value of the ^{40}K activity concentration. Sample 8 has smallest value of the ^{40}K activity concentration. Kılıç et al. (2006) found that ^{40}K activity concentrations ranged from 446 ± 12 to 523 ± 14 Bq kg^{-1}. Görür et al. (2011) found that

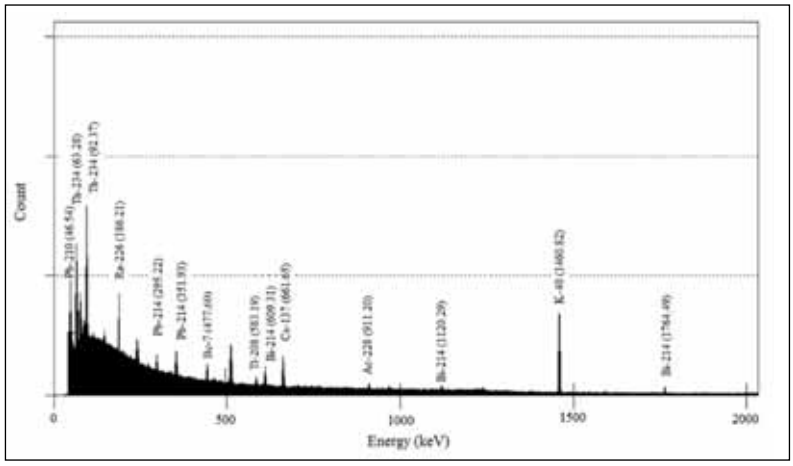

Figure 1. Gamma spectrum of a tea sample.

Table 1. The activity concentrations of ^{238}U, ^{214}Pb, ^{214}Bi, ^{226} Ra, ^{40}K and ^{210}Pb in Turkish tea samples (Bq kg^{-1})

Sample No	^{238}U	^{214}Pb	^{214}Bi	^{226}Ra	^{40}K	^{210}Pb
1	<2.8	<3.2	<2.6	<3.0	506.6±40.5	41.5±3.3
2	<3.1	<3.6	<2.9	<3.1	435.2±34.8	44.1±3.5
3	<3.6	<3.7	<3.3	<3.4	484.0±38.7	58.3±4.6
4	<4.1	<4.4	<4.0	<4.2	586.4±46.9	11.1±0.9
5	<3.7	<3.8	<3.5	<3.6	476.2±38.1	47.2±3.8
6	<3.2	<3.5	<3.0	<3.3	432.5±34.6	73.4±5.9
7	<2.7	<3.2	<2.9	<4.0	520.6±39.1	55.7± 4.5
8	<3.1	<3.5	<3.3	<3.4	428.7± 32.2	29.8± 2.4
9	<3.5	<3.8	<3.6	<2.8	452.0±33.9	34.8±2.8
10	<2.8	<3.3	<3.0	<3.4	514.0±38.6	81.9±6.6
11	<3.7	<4.3	<4.1	<3.1	681.9±51.4	47.0±3.8
12	<3.4	<4.1	<3.8	<3.8	449.0±33.7	50.0±4.1
13	<3.3	<3.9	<3.5	<4.1	448.1±33.6	41.1±3.3
14	<3.8	<4.4	<4.0	<3.0	432.4±32.4	73.1±5.9
15	<4.1	<4.6	<4.4	<4.6	433.0±32.5	52.5±4.2

^{40}K mean activity concentrations were 445.63 ± 17.83. Harb found that ^{40}K activity concentrations ranged from 470 ± 20 to 691 ± 27 Bq kg^{-1}. D. Desideri et al. found that ^{40}K activity concentrations ranged from 463 and 936 Bq kg^{-1}. Di Gregorio et al. (2004) found that ^{40}K activity concentrations ranged from 404 ± 80 to 877 ± 180 Bq kg^{-1}. ^{210}Pb and ^{40}K radioactivity concentrations diagram are shown in the Figure 2.

The activity concentrations of ^{232}Th, ^{228}Th, ^{228}Ra, ^{7}Be and ^{137}Cs in Turkish tea samples are given in the Table 2. A less than sign (<) was used to indicate the below MDA values of the detector. ^{232}Th, ^{228}Ra and ^{228}Th activity concentrations are below MDA value.

^{137}Cs activity concentrations ranges were found to be 12.6±1.0 - 60.5 ± 4.6 Bq kg^{-1}. Sample 13 has highest value of the ^{137}Cs activity concentration. Sample 15 has smallest value of the ^{137}Cs activity concentration. The results were within acceptable limits for human consumption according to European Council Regulation No 737/90 for the reported radionuclides (European Council 1990). Görür et al. found that ^{137}Cs mean activity concentrations were 42.00 ± 1.45 for the Turkish market tea samples. Kılıç et. al.

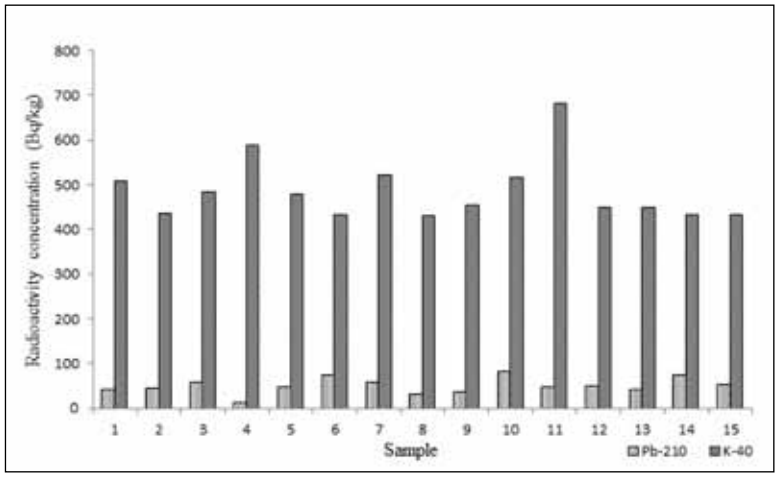

Figure 2. ^{210}Pb and ^{40}K radioactivity concentrations.

Table 2. The activity concentrations of ^{232}Th, ^{228}Th, ^{228}Ra, ^{137}Cs and ^{7}Be in Turkish tea samples (Bq kg^{-1}).

Sample No	^{232}Th	^{228}Th	^{228}Ra	^{137}Cs	^{7}Be
1	<3.8	<3.5	<4.0	39.5±3.2	<4.1
2	<4.2	<4.0	<3.9	12.9±1.0	<3.8
3	<4.8	<4.4	<4.2	35.3±2.8	<2.5
4	<4.3	<3.9	<4.1	26.1±2.1	<3.8
5	<3.8	<3.9	<4.0	22.8±1.8	<3.4
6	<2.7	<2.5	<2.9	56.6±4.5	<2.9
7	<3.2	<3.5	<3.4	53.6±4.1	<2.5
8	<5.4	<5.0	<5.6	45.5±3.4	<3.7
9	<4.2	<3.9	<4.5	35.9±2.7	<4.1
10	<3.2	<3.1	<3.6	40.0±3.1	<3.7
11	<3.7	<3.5	<3.8	44.5±3.4	25.6±1.9
12	<2.8	<2.9	<3.2	40.4±3.1	18.9±1.4
13	<4.6	<4.2	<4.7	60.5±±4.6	9.1±0.7
14	<3.8	<3.5	<3.6	56.7±4.3	42.6±3.2
15	<3.4	<3.2	<3.5	12.6±1.0	57.9±4.3

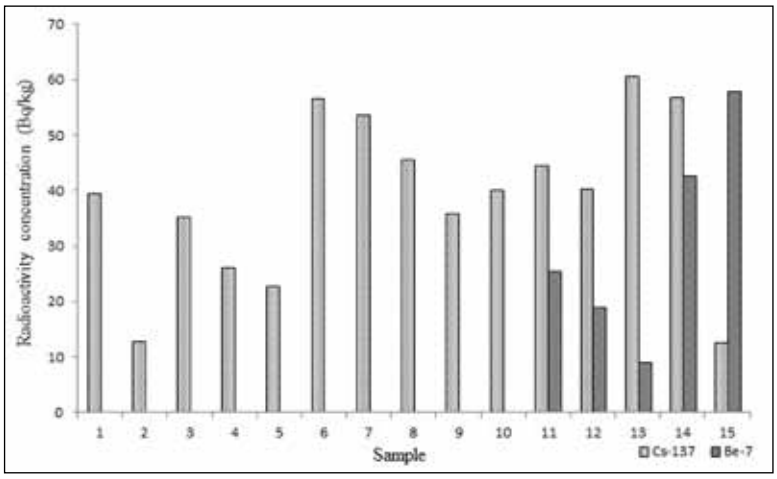

Figure 3. ^{137}Cs and ^{7}Be radioactivity concentrations.

found that ^{137}Cs activity concentrations ranged from 29.5 ± 0.5 to 100.7 ± 1.6 Bq kg^{-1}. Harb found that ^{137}Cs activity concentrations ranged from 0.4 ± 0.2 to 1.3 ± 0.2 Bq kg^{-1}. D. Desideri et al. found that ^{137}Cs activity concentrations ranged from 0.3 and 2.6 Bq kg^{-1}. In another study for the Argentinian tea samples Di Gregorio et al. found that ^{137}Cs activity concentrations ranged from 1.1 ± 0.5 to 10.3 ± 2.1 Bq. kg^{-1}. The eastern Karadeniz Region of Turkey which the tea plantation area has been affected from the Chernobyl nuclear accident. As a result of that ^{137}Cs activity levels were much higher in the present study than the values of other studies.

^{7}Be activity concentrations ranges were found to be 9.1 ± 0.7 – 57.9 ± 4.3 Bq kg^{-1}. Sample 15 has highest value of the ^{7}Be radioactivity concentration. Sample 13 has smallest value of the ^{7}Be activity concentration. Samples which have radioactivity concentrations are new season harvested. ^{7}Be and ^{137}Cs radioactivity concentrations diagram are shown in the Figure 3.

4. Conclusion

In this paper, the natural and artificial radioactivity concentrations of tea samples which produced in Turkey are investigated. Present study showed that ^{137}Cs activity concentrations are much higher than the values of the other studies. The reason was tea plantation area of Turkey (The Eastern Karadeniz Region) was effected by the Chernobyl nuclear accident. In the EU, the limit for radiocaesium was established at 600 Bq kg^{-1} for agricultural production. According to European Council Regulation No 737/90 for the reported radionuclides, ^{137}Cs activity values are not hazardous for public health. The natural radionuclide concentrations of ^{238}U and ^{232}Th series were found MDA in the tea samples.

On the other hand, ^{40}K and ^{210}Pb radionuclide activity concentrations are in accordance with other studies. Due to the radionuclide properties of the ^{7}Be and climate conditions of the tea production area of the Turkey, ^{7}Be radioactivity was observed. ^{7}Be radioactivity encountered samples were newly harvested. There are no studies about the ^{7}Be and ^{210}Pb radioactivity concentrations of the Turkish tea samples in the literature.

It is very important to determine the level of radioactivity concentrations in tea products to ensure consumer safety. The obtained results provide useful information to carry out a dose assessment due to ingestion of these products. Also dose assessment may carry out for workers in the tea production industries.

5. Acknowledgement

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