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Araştırma Makalesi / Research Article

Determination of Radionuclides and Elemental Concentrations in Black and White Tea Samples from Karadeniz in Turkey

Mirac KAMISLIOGLU[1*], Canel EKE[2], Tugba KAMAN[3], Ismail BOZTOSUN[4]

Abstract

Tea is the second most consumed plant worldwide after water and it can be grown in humid climates. Considering the frequency of consumption, it is inevitable for the tea plant to become a very important field of study both in terms of commercial and public health. The tea plant appears in four different forms which are green tea, black tea, oolong tea and white tea depending on the production method. Especially white tea has started to attract the attention of science in recent years due to its antioxidant effect. The aim of this study is to examine radionuclides and elemental concentrations of black and white tea collected from Karadeniz in Turkey. Natural and artificial radionuclides concentrations were determined using gamma-ray spectrometry. ^{226}Ra, ^{232}Th, ^{40}K, ^{137}Cs were detected in both black and white tea samples. Elemental concentrations of the black and white tea were determined using energy dispersive x-ray fluorescence (EDXRF) spectrometry. Scanning electron microscopy (SEM) micrographs were obtained for black and white tea samples. ^{226}Ra and ^{137}Cs activity concentrations of black tea are higher than white tea however ^{232}Th and ^{40}K activity concentrations of white are higher than black tea. Total dose value of white is higher than black tea.

Keywords: Natural and artificial radionuclides, White tea, HPGe detector.

Türkiye Karadeniz Bölgesinden alınan Siyah ve Beyaz Çay Örneklerinde Radyonüklid ve Elementel Konsantrasyonlarının Belirlenmesi

Öz

Çay, sudan sonra dünyada en çok tüketilen ikinci bitki türüdür ve nemli iklimlerde yetiştirilir. Tüketim sıklığı göz önüne alındığında çay bitkisinin hem ticari hem de halk sağlığı açısından çok önemli bir çalışma alanı haline gelmesi kaçınılmazdır. Çay bitkisi, üretim şekline göre yeşil çay, siyah çay, oolong çayı ve beyaz çay olmak üzere dört farklı biçimde karşımıza çıkar. Özellikle beyaz çay, antioksidan etkisi nedeniyle son yıllarda bilimin ilgisini çekmeye başlamıştır. Bu çalışmanın amacı, Türkiye'de Karadeniz Bölgesi'nden toplanan siyah ve beyaz çayın radyonüklitlerini ve elementel konsantrasyonlarını incelemektir. İncelenen örnek çayların doğal ve yapay radyonüklid konsantrasyonları gama ışını spektrometresi kullanılarak belirlendi. Hem siyah hem de beyaz çay örneklerinde ^{226}Ra, ^{232}Th, ^{40}K, ^{137}Cs konsantrasyon seviyeleri tespit edildi. Siyah ve beyaz çayın elementel konsantrasyonları, enerji ayırımlı x-ışını floresans (EDXRF) spektrometresi kullanılarak belirlendi. Siyah ve beyaz çay örnekleri için taramalı elektron mikroskobu (SEM) mikrografları elde edildi. Siyah çayın ^{226}Ra ve ^{137}Cs aktivite konsantrasyonları beyaz çaydan daha yüksek olduğu, ancak beyaz çayın ^{232}Th ve ^{40}K aktivite konsantrasyonlarının siyah çaydan daha yüksek olduğu görülmüştür. Beyaz çayın toplam doz değeri siyah çaya göre daha yüksek olduğu belirlenmiştir.

Anahtar Kelimeler: Doğal ve yapay radyonüklitler, Beyaz çay, HPGe dedektörü.

[1]Bandırma Onyedi Eylül University, Department of Medical Imaging, Balıkesir, Turkey, mkamislioglu@bandirma.edu.tr
[2]Akdeniz University, Department of Mathematics and Science Education, Antalya, Turkey, ceke@akdeniz.edu.tr
[3]Uskudar University, Department of Medicinal and Aromatic Plants, İstanbul, Turkey, tugba.kaman@uskudar.edu.tr
[4]Akdeniz University, Faculty of Science, Antalya, Turkey, boztosun@akdeniz.edu.tr

[1]https://orcid.org/0000-0002-0666-8832 [2]https://orcid.org/0000-0002-6672-6467 [3]https://orcid.org/0000-0002-5885-0193
[4]https://orcid.org/0000-0001-9668-6307

[*]**Sorumlu Yazar/Corresponding Author** **Geliş/Received:** 04.04.2021 **Kabul/Accepted:** 25.09.2021

1. Introduction

Tea which is the old timer and widely used up drink in the world is very important for society to determine the amount of radioactivity it contains due to the prevalence of consumption (Silva et al., 2020). It is known that natural and artificial radioactivity triggers or activates cancer in living tissues. Therefore, it is very important to determine the radioactivity concentrations which has been taken from foods. International organizations state the necessity and importance of the determination of the level of the radioactivity amounts received by the population (UNSCEAR, 2000; Scheibel and Appoloni, 2007; Lopes et al.,2020). Turkey is among the leading countries in the world in terms of tea consumption. However, it has been observed that there are very few studies on black and white tea activity concentrations in the literature. Therefore, determining the health risks associated with ionizing radiation in black and white tea may be beneficial in preventing the risks that may arise in terms of public health (Silva et al., 2020).

Tea is a common drink produced from young shoots and leaves of the plant Camellia sinensis L. (O) Kuntze and is served as hot or cold all over the world. There are different varieties such as black tea, green tea, oolong tea, white tea, pu-erh tea according to the processing methods of the tea (Katiyar and Mukhtar, 1997). Tea, flavan-3-ol (catechin) family, e.g., epigallocatechin, epicatechin, epigallocatechin-3-o-gallate, epicatechin3-o-gallate, (+) catechins and (+) contains a large number of polyphenolic compounds such as Gallo catechin (Csupor, 2015).

Tea has been shown to be protective against many diseases as various types of cancers such as coronary heart disease (CHD), stroke, cardiovascular diseases (CVD), hypertension (HT), esophagus, stomach, collateral, lung skin, breast and prostate, and neurological diseases such as Alzheimer's and Parkinson's disease, arthritis, antiviral and anti-inflammatory diseases. It is also stated that it has regulating effects on bone density. It is stated that both green and white tea is an antioxidant drink because of the polyphenolic compounds in its content and it provides its protective effect from chronic diseases in this way (Henning, 2003).

In epidemiological studies, it has been shown that a diet rich in flavonoids can reduce the risk of arteriosclerosis and other heart diseases, prevent cancers and aging and counteract radiation (Zhu, et al., 2006). White tea, unopened buds and leaves covered with silvery hairs It is a non-fermented tea obtained by harvesting once a year in early spring (Hilal and Engelhardt, 2007).White tea extracts has a high antioxidant property (Baublis et al., 2000; Sivritepe, 2000; Tosun and Karadeniz, 2005; Kinsella et al., 1993; Thring et al., 2009; Hajiaghaalipour et al., 2015) and show anti-proliferative activity and inhibit colon cancer cells to the HT-29 cell line (Hajiaghaalipour et al., 2015) also white and green tea have the highest free radical scavenging activity (Almajano et al., 2008) and white tea can act as a protective agent against a wide variety of health conditions caused by oxidative stress due to their strong antioxidant activities and that white tea has significant antioxidant, antibacterial, anticarcinogenic and antimutagenic and antiallergic activities (Hajiaghaalipour et al., 2015; Mandel and Youdim, 2004; Santana-Rios, et al., 2001; Santana-Rios, et al., 2001; Almajano et al., 2008; Gondoin et al., 2010; Unachukwu et al., 2010; Damiani et al., 2014). In addition to the flavanol content of black tea, there are studies showing that black tea also has high antioxidant activity due to these secondary phenolic substances formed in the enzymatic oxidation stage (Tosun and Karadeniz, 2005). These antioxidants help eliminate harmful toxins from the body. As a result, since green tea contains catechin and other polyphenolic components on high levels, it has stronger anticarcinogenic effects than black tea, but it has been reported that the white tea has stronger effects in the fight against cancer because it contains the highest percentage of these components (Salman and Özdemir, 2018).

In addition to aforementioned studies, numerous recent studies have been conducted on the determination of the radioactivity concentration of tea (Baltas et al., 2016; Gökmen et al., 1995; Varinoğlu et al., 1995). In the study has been shown that radioactivity has been detected in the Black Sea region even after years by Varinlioglu et al., Furthermore, this study has been explained that the ^{137}Cs activity has not yet been determined (Varinoğlu et al., 1995).

Radioactive sources that reach nature in some way are first transmitted directly through plants. Moreover, plants absorb radioactive sources through their roots by indirect contamination from the soil. In plants consumed daily by humans, there are generally ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs and radionuclides come from their roots. Since natural radionuclides are taken into the human body through plants, similar activity is expected in plants used for medicinal purposes (Gökmen, 1995). As a result of the Chernobyl disaster that occurred in April 1986, an increase in ^{137}Cs activity was determined in nature and especially in soil-derived nutrients (plants). In this study, we will calculate natural and artificial radionuclides concentration of black and white tea also elemental concentrations of black and white tea will be determined.

2. Materials and Methods

The black and white tea samples of this study were taken from the Black Sea in Rize region. The coordinates of the region chosen as the study area are given as for black tea (41,1602 latitude -40,9473 longitude) and for white tea (41,1590 latitude- 40,9456 longitude). White tea was obtained by drying at 90 ^{0}C for approximately 15 minutes after withering in the shade for 3 days. Black tea was obtained by curling after 12 hours of withering, followed by oxidation for 55 minutes at 25 ^{0}C, followed by drying at 90 C^{0} for 20 minutes (Carloni et al., 2012).

Activity concentration of natural radionuclides and artificial radionuclides were determined using gamma-ray spectrometry system. The black and white tea samples were weighed and put into

100 ml plastic containers. Black and white tea samples kept 30 days before the counted at air impermeable case to come secular equilibrium between ^{232}Th and ^{226}Ra and their decay products (Korkmaz et al.,2011).

Black and white tea samples were counted using high purity germanium (HPGe) detector from Akdeniz University, Department of Physics which is electrical cooled, p type, energy resolution and relative efficiency of the HPGe detector are 1.85 keV and 40%, respectively at 1332.5 keV of 60Co. Details of the HPGe detector can be found in Ref. (Eke and Boztosun, 2015). The detector efficiency calibration was carried out using mixed reference material from Cekmece Nuclear Research and Training Center which was called IAEA 1364-43-2 in the energy range from 47 to 1836 keV by Ozmen et al. (Ozmen et al. 2014a, 2014b). Black and white tea samples were counted about a day using high purity germanium (HPGe) detector and a background count was taken from the detector one day without a sample to reduce the background effect of the tea samples. Obtained gamma-ray spectra of black and white tea samples were analyzed using Maestro-32 software (Maestro 2008). Background spectrum was subtracted from spectrum of black and white tea samples to obtain accurate counts. Minimum detectable activities (MDA) of each nuclide were calculated at the 95% confidence level using well known as follow Curie's equation (Chalmers, 1970);

$$MDA = \frac{2.71+4.66\sigma}{t.\varepsilon.I_{\gamma}.m} \tag{1}$$

Where σ is the background standard deviation, t is counting time, ε is the efficiency of the of the interested gamma-ray energy, I_{γ} is emission probability of interested gamma-ray energy and m is mass of sample. Activity of the radionuclides (A) were calculated using following equation (Canbazoğlu and Doğru, 2013);

$$A(Bq/kg) = \frac{N}{m.t.\varepsilon.I_{\gamma}} \tag{2}$$

Where N is the number of gamma ray counts after background spectrum subtracted from tea sample spectrum, m is the mass of the sample, t is counting time, ε is efficiency of the interested gamma-ray energy and I_{γ} is emission gamma-ray probability.

To forecast of health effect of radionuclides on the humans, intake total dose from food should be calculated. Total ingestion dose is proportional to consumption ratio of food and activity concentration of the radionuclides (Korkmaz et al.,2011; Canbazoğlu and Doğru, 2013). Total ingested dose ($H_{T,r}$) from foods are calculated using following equation (ICRP, 1996; Abojassim et al., 2014; Van et al., 2019):

$$H_{T,r} = \sum (U^i C_r^i) g_{T,r} \tag{3}$$

where i represents food category, $U^i (kg/y)$ is annual consumption rate parameter, C_r^i $(Bq/kg)$is activity concentration of the interested radionuclide in sample, $g_{T,r}$ is dose transformation paramater for interested radionuclide in food sample. In this study annual consumption rate of tea is taken 3 kg per year (Kilic et al., 2012). Dose transformation parameters for adults are 4.5×10^{-8} (Sv/Bq), 2.3×10^{-7}(Sv/Bq), 6.2×10^{-9} (Sv/Bq) and 1.3×10^{-8} (Sv/Bq) for ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs, respectively (ICRP, 1996).

Scanning electron microscope (SEM) images of the acquired composites were saved on Philips XL30 ESEM-FEG/EDAX. Elemental contents of components of black and white tea samples were determined using energy dispersive x-ray fluorescence (EDXRF) spectrometry in Marmara University.

3. Results and Discussion

Black and white tea samples were examined using gamma-ray spectrometry and EDXRF spectrometry. MDAs were calculated 2.365 Bq/kg for ^{214}Pb, 1.476 Bq/kg for ^{214}Bi, 1.936 Bq/kg for ^{228}Ac, 0.441 Bq/kg for ^{137}Cs and 6.997 Bq/kg for ^{40}K using background spectrum. ^{226}Ra activity concentration was determined using mean value of ^{214}Pb (351 keV) and ^{214}Bi (609 keV), ^{232}Th activity concentration was determined using ^{228}Ac (911 keV). Activity concentrations of ^{40}K and ^{137}Cs were determined directly using 1460 keV and 662 keV photon energies, respectively. Gamma-ray spectrum of black and white tea sample are given in Figure 1. EDXRF spectrum of black tea and white tea samples are presented in Figure 2.

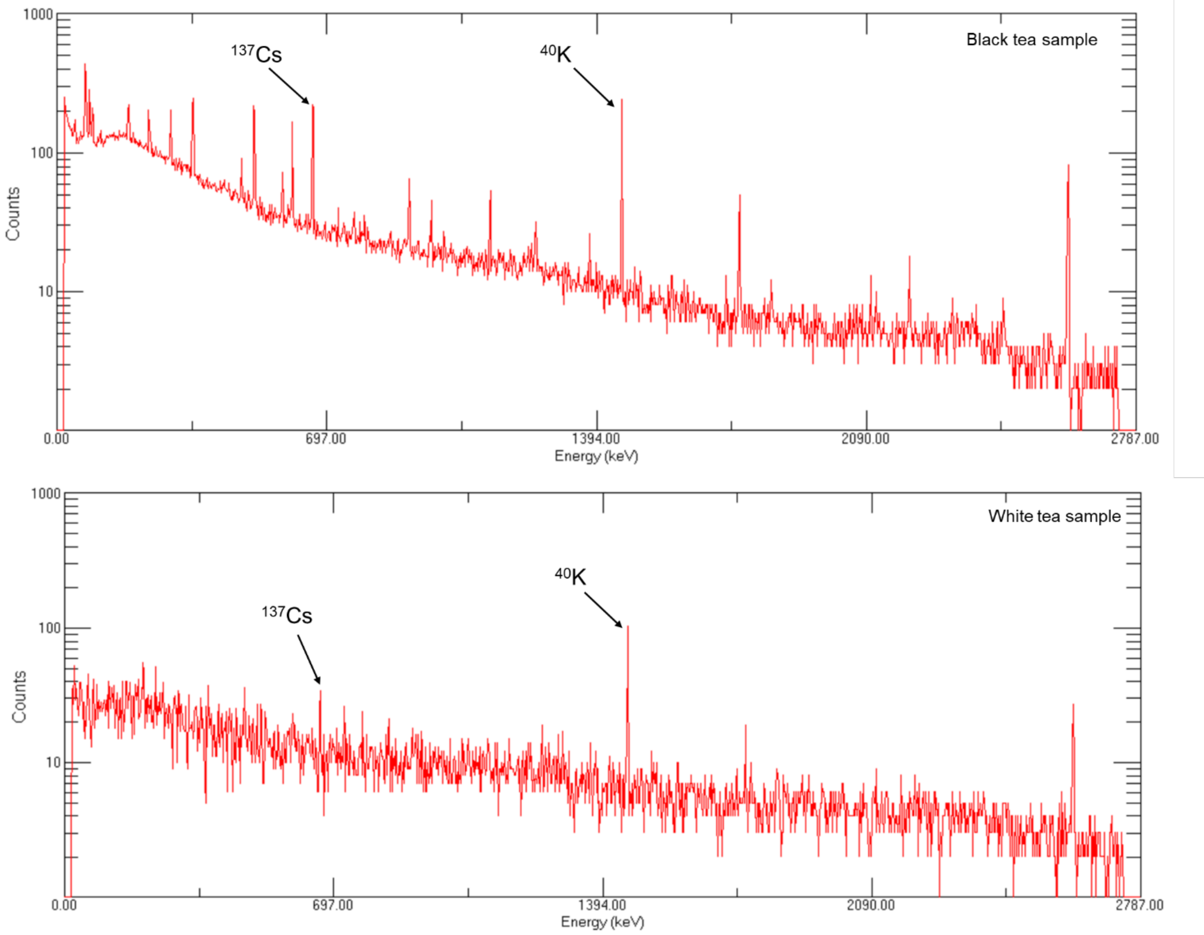


Figure 1. Gamma-ray spectrum of black and white tea sample

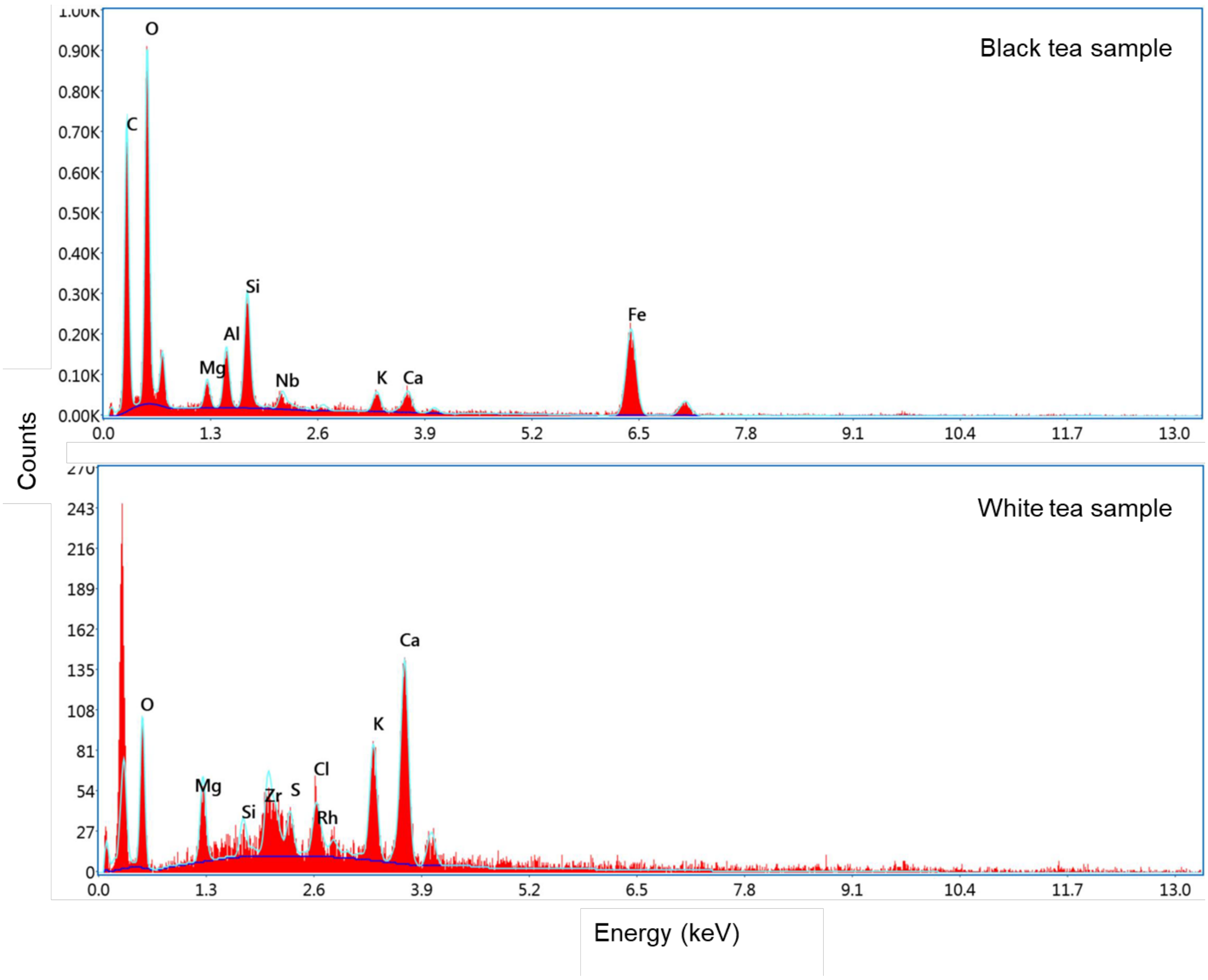


Figure 2. EDXRF spectrum of black tea and white tea samples.

Table 1. Activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K, ^{137}Cs and total annual ingested dose value of tea samples.

Samples	^{226}Ra (Bq/kg)	^{232}Th (Bq/kg)	^{40}K (Bq/kg)	^{137}Cs (Bq/kg)	H(μSv/y)
Black tea	17.63±2.47	15.25±6.73	902.88±59.60	76.18±6.64	33.67
White tea	9.70±2.14	28.27±9.51	1240.78±81.73	13.44±7.99	44.42

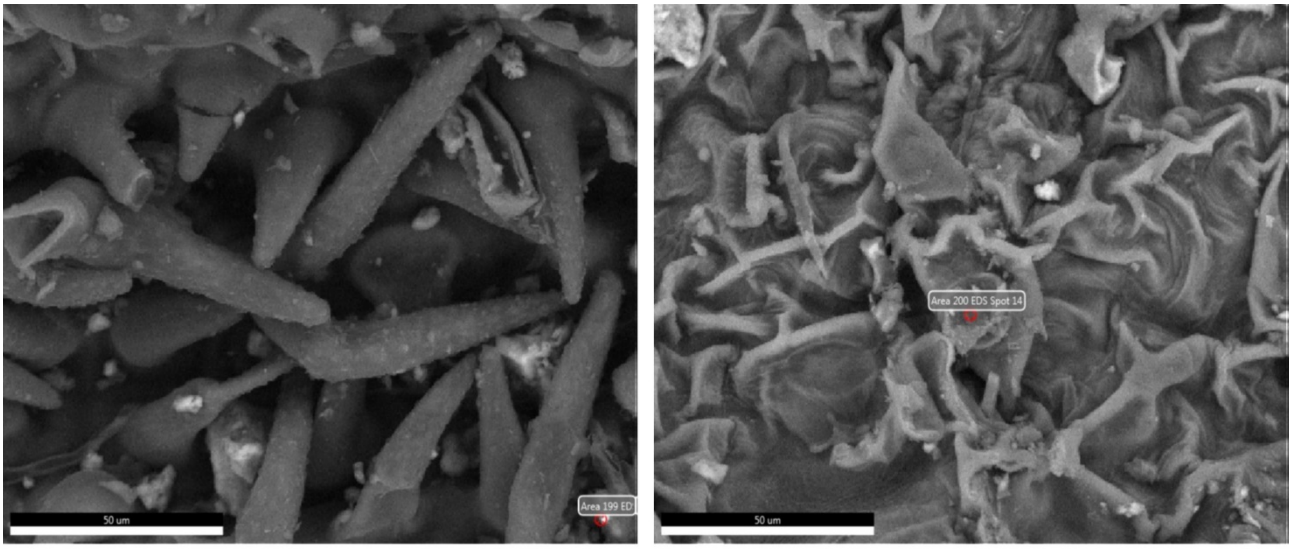
Table 2. Elemental components of black and white tea samples (wt.%).

Element	Black tea	White tea
Carbon (C)	25.25	n.d.
Oxygen (O)	39.9	40.58
Magnesium (Mg)	1.43	6.14
Aluminium (Al)	2.47	n.d
Silicon (Si)	4.0	1.94
Niobium (Nb)	1.4	n.d
Potassium (K)	1.08	10.44
Calcium (Ca)	1.38	22.96
Iron (Fe)	13.09	n.d.
Zirconium (Zr)	n.d.	9.16
Sulfur (S)	n.d.	2.82
Chlorine (Cl)	n.d.	3.83
Rhodium (Rh)	n.d.	2.13

* n.d: not detected

The activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K, ^{137}Cs and total annual ingested dose values (H) of tea samples are presented in Table 1. ^{226}Ra and ^{137}Cs activity concentrations of black tea samples are higher than white tea samples while ^{232}Th and ^{40}K activity concentrations of the white tea sample is higher than black tea sample. ^{226}Ra, ^{232}Th, ^{40}K, activity concentrations of the tea samples lower than reported limit values by UNSCEAR (UNSCEAR, 2000). Total annual ingested dose values of white tea sample are higher than black tea sample.

Elemental components of black and white tea samples are shown in Table 2 and Fig.2. Carbon (C), Oxygen (O), Magnesium (Mg), Aluminium (Al), Silicon (Si), Niobium (Nb), Potassium (K), Calcium (Ca) and Iron (Fe) were detected in black tea sample. Oxygen (O), Magnesium (Mg), Sulfur (S), Chlorine (Cl), Potassium (K), Calcium (Ca), Zirconium (Zr), Rhodium (Rh) were detected in white tea sample. Zirconium (Zr), Rhodium (Rh) were not detected in black tea sample. Carbon (C), Phosphate (P) and Molybdenum (Mo) were not detected in white tea sample. Scanning electron microscope (SEM) micrographs of black tea and white tea samples were indicated in Figure 3.



a) Black tea sample b) White tea sample

Figure 3. SEM micrographs of black and white tea samples.

Generally, studies conducted in Turkey have been mostly about measuring the radioactivity in the Black Sea region, but not on the tea plant. In study, conducted by Puchkova and Bogdanova in 2015, he took the tea plant as a reference plant when examining the characteristics of the medicinal plant, as it resembles a medicinal plant structurally. He made radioactivity analysis for ^{210}Po and ^{210}Pb (Puchkova and Bogdanova, 2015). After the Chernobyl disaster, it has been reported in the studies conducted by TAEK that the radiation activity increased in the Black Sea region especially for tea plants which are located in very high regions (Yeltepe et al., 2018).

In a study conducted by Mishra for white teas in 2006, he found that the amount of 10 kGy radiation due to radiation therapy did not affect the tea radioactively. It is important to know the level of radioactivity concentrations in tea to ensure consumer safety. The activity concentration in tea was evaluated in areas of high background radiation (Hamzah et al., 2011; Mitrovi´c et al., 2014). Similarly, it is important to know the concentration of activity in the tea grown in risky regions for the necessity to assess radiological hazard indexes and estimate soil-plant transfer factors (Espiona et al.,2016; Aktar et al., 2018). The changes in activity concentrations of tea samples occur due to nuclear accidents and it is very important to determine the environmental effects (Jevremovic et al., 2011). In some cases, radioactivity in tea has been detected in places that have been affected and/or unexpectedly by the accident. Regular measurements of radioactivity are used as a monitor to help identify possible contaminant sources (Di Gregorio, et al., 2004). Dose estimates are also included in the determination of radiological hazards, therefore annual radionuclide distributions should be examined. Determination of total amount of radioactivity is very important for effective radioactivity analysis.

4. Conclusions and Recommendations

Activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K, ^{137}Cs and total annual ingested dose value for black and white tea samples were calculated using gamma-ray spectrometry. MDAs of interested radionuclides were determined. Activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K, ^{137}Cs were compared with other studies from scientific literature. Elemental concentrations of black and white tea samples were determined using EDXRF spectrometry. SEM micrographs were obtained for black and white tea samples. The obtained results indicate that ^{226}Ra and ^{137}Cs activity concentrations of black tea are higher than white tea however ^{232}Th and ^{40}K activity concentrations of white are higher than black tea. Total annual ingested dose value of white is higher than black tea. Also, we can deduce that in tea samples which are grown in the Black sea region can still observe the Chernobyl nuclear accident effect after 34 years because ^{137}Cs activity concentration which is artificial radionuclide was detected in both the black and white tea samples. According to the research made by Gökmen et al. (1995), it has been determined that the products most affected by the Chernobyl disaster in the Eastern Black Sea region are tea, hazelnuts and tobacco. This study has been shown that the most radioactive concentration belongs to the tea plant, as it has no protective shell (Gökmen et al., 1995). Compared to other studies, activity concentration values were found that to be within low limits (Zhu 2006, Hamzah 2011, Yeltepe 2018). In the next process, it has been revealed that in order to distinguish the basic level of radioactivity from the contamination, the measurements and analyzes should be made for the plants at the background level.

Authors' Contributions

All authors contributed equally to the study.

Statement of Conflicts of Interest

There is no conflict of interest between the authors.

Statement of Research and Publication Ethics

The author declares that this study complies with Research and Publication Ethics.

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