

## Activity concentrations of $^{226}\text{Ra}$ , $^{232}\text{Th}$ , $^{137}\text{Cs}$ and $^{40}\text{K}$ in *Cystoseira barbata* samples harvested from the Eastern Black Sea coast of Türkiye

*Türkiye'nin Doğu Karadeniz kıyılarından toplanan Cystoseira barbata numunelerindeki  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ ,  $^{137}\text{Cs}$  ve  $^{40}\text{K}$  aktivite konsantrasyonları*

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### Abstract

Macroalgae are important bioorganisms used in fish feed and food industry. In addition, macroalgae are used in studies to determine radioactive pollution levels in the seas. In this study, it was aimed to determine the activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ ,  $^{137}\text{Cs}$  and  $^{40}\text{K}$  in *Cystoseira barbata* which is a brown macroalgae samples obtained from the Eastern Black Sea coast of Turkey. Gamma spectrometry analyzes of the samples were carried out with high purity germanium (HPGe) detector system in the Nuclear Physics laboratory of Recep Tayyip Erdoğan University. The average  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ ,  $^{137}\text{Cs}$  and  $^{40}\text{K}$  activity concentrations in the samples were found to be 11.2, 7.61, 2.74 and 690.7 Bq.kg<sup>-1</sup>, respectively. The average annual effective dose values resulting from these radionuclides were obtained as 3.14, 1.75, 0.04 and 4.28  $\mu\text{Sv.y}^{-1}$ , respectively. Total annual effective dose values calculated with the contribution of all radionuclides were found to be in the range of 1.37-18.83  $\mu\text{Sv.y}^{-1}$ . The calculated annual effective dose values are lower than 0.29 mSv.y<sup>-1</sup>, which is stated by UNSCEAR as the average annual effective dose received through ingestion of natural radionuclides, and the samples examined do not pose any radiological risk.

**Keywords:** Bioindicator, Black sea, *Cystoseira barbata*, Macroalg, Radioactivity

### Öz

Makroalgler, balık yemi ve gıda sanayinde kullanılan önemli biyoorganizmalardır. Ayrıca makroalglerden denizlerdeki radyoaktif kirlilik düzeylerinin belirlenmesi çalışmalarında yararlanılır. Bu çalışmada Türkiye'nin Doğu Karadeniz kıyılarından toplanan kahverengi bir macroalg olan *Cystoseira barbata* numunelerinde  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ ,  $^{137}\text{Cs}$  ve  $^{40}\text{K}$  aktivite konsantrasyonlarının belirlenmesi amaçlandı. Örneklerin gama spektrometre analizleri Recep Tayyip Erdoğan Üniversitesi Nükleer Fizik laboratuvarında bulunan yüksek saflıktaki germanyum (HPGe) dedektör sistemiyle gerçekleştirildi. Numunelerdeki ortalama  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ ,  $^{137}\text{Cs}$  ve  $^{40}\text{K}$  aktivite konsantrasyonları sırasıyla 11.2, 7.61, 2.74 ve 690.7 Bq.kg<sup>-1</sup> olarak bulundu. Bu radyonüklitlerden kaynaklanan ortalama yıllık etkin doz değerleri sırasıyla 3.14, 1.75, 0.04 ve 4.28  $\mu\text{Sv.y}^{-1}$  olarak elde edildi. Tüm radyonüklitlerin katkısı ile hesaplanan toplam yıllık etkin doz değerleri 1.37-18.8  $\mu\text{Sv.y}^{-1}$  aralığında bulundu. Hesaplanan yıllık etkin doz değerleri UNSCEAR tarafından doğal radyonüklitlerin sindirimi yoluyla alınan ortalama yıllık etkin doz değeri olarak belirtilen 0.29 mSv.y<sup>-1</sup>'den düşüktür ve incelenen numuneler radyolojik açıdan herhangi bir risk teşkil etmemektedir.

**Anahtar kelimeler:** Biyoindikatör, Karadeniz, *Cystoseira barbata*, Makroalg, Radyoaktivite

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## 1. Introduction

Seas are the important water sources with their great nutritional power. Türkiye is a rich country in terms of seas. Black Sea is located in the north, the Mediterranean Sea in the south, the Aegean Sea in the west and the Marmara Sea in the northwest of Türkiye. The Black Sea is surrounded by six coastal countries, including Bulgaria, Georgia, Romania, Russia, Ukraine and Türkiye. Other countries that are around the Black Sea but do not have a coastline are Albania, Austria, Belarus, Bosnia-Herzegovina, Croatia, Czechia, Germany, Hungary, Italy, Macedonia, Moldova, Poland, Slovakia, Slovenia, Switzerland and Yugoslavia.

The Black Sea, lies between 40°55'- 46°32'N latitudes and 27°27'- 41°32'E longitudes with a water volume of  $5.3 \times 10^5 \text{ km}^3$  (Bakan & Büyükgüngör, 2000). It is connected to the world's seas only through the straits in İstanbul and Çanakkale. This situation has caused the Black Sea to turn into a reservoir that contains oxygen from the surface to a depth of approximately 150 meters, and contains hydrogen sulfide at deeper levels and can accumulate all kinds of organic and inorganic pollutants that reach it (Yılmaz, 2002).

Despite all these adverse conditions, the Black Sea has provided significant benefits to humanity for centuries, both in terms of fishing and other uses. But in recent years, the Black Sea has become an unhealthy ecosystem due to some natural and human induced effects. The main effects are domestic, industrial and agricultural wastes discharged into the Black Sea directly or through rivers and nuclear pollutions. Especially large rivers such as the Danube, Dnieper and Dniester, and streams such as Sakarya, Kızılırmak and Yeşilirmak carry the wastes of the regions they pass through to the Black Sea, and cause it to bear the burden of a basin that is approximately five times larger than its area (Alkan et al., 2008).

One of the main pollutants that cause pollution of the Black Sea is radionuclides. Radionuclides are the radioactive elements that emit gamma rays until they become stable. The main natural radionuclides reaching the marine environment are uranium and thorium decay series products and  $^{40}\text{K}$ , emerged during the formation of the earth. Natural radionuclides can be found in many substances such as rocks, soil, water, agricultural fertilizers, fossil fuels and detergents, and can cause an increase in the level of radioactivity in the seas (Gahrouei et al., 2013; Abojassim et al., 2014).

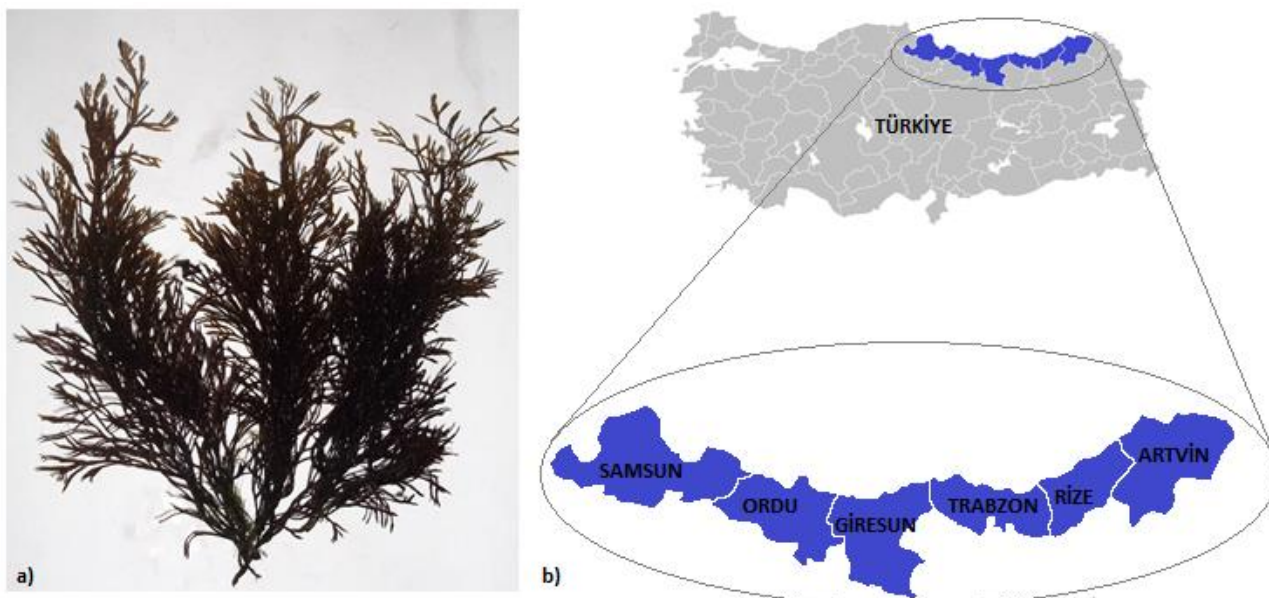
Hundreds of nuclear power plants have been established around the Black Sea and many of them are still active (Kaya & Doğan, 2023). After the Chernobyl Nuclear Power Plant accident occurred on 1986, it has been estimated that the direct deposition due to  $^{137}\text{Cs}$  was around 800 TBq (Sawidis et al., 2003; Kritidis & Florou, 1990). When both  $^{137}\text{Cs}$  and natural radioactive elements ( $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$ ) reach the marine environment, they do not just remain in the water, depending on environmental conditions, they can either settle to the bottom or be taken into the bodies by various organisms and cause bioaccumulation. Radionuclide pollution, which can reach the marine organisms and humans through the food chain, is an important environmental problem for both marine organisms and humans.

Macroalgae are prokaryotic or eukaryotic primitive plant organisms that have photosynthetic pigments and do not have complex reproductive systems (Sze, 1998). Thanks to their ability to absorb pollutants from water, macroalgae are used as bioindicator in the researches of radioactive pollution in the seas (Mihai et al., 1999; Sawidis et al., 2003; Sam et al., 1998). Additionally, macroalgae are also used in fish feed and food industry (Mwendwa et al., 2023). *Cystoseira barbata* is a brown macroalgae (*Phaeophyceae*) that can survive by clinging to the hard surfaces and their thallus can reach up to 150 cm in length (Ribera et al., 1992; İrkin & Erduğan, 2014). Brown macroalgae constitute the feeding, resting, protection and breeding areas of marine organisms. The products of photosynthesis in brown macroalgae are mannitol, laminarin, algin and oils (Yeşilova, 2014). Algin is a type of colloidal carbohydrate and used in the food industries thanks to its properties such as gelling and suspension (Taşkın, 1999; Kurt, 1999).

Pollution of the Black Sea is an important problem that concerns dozens of states and millions of people. A few of the studies using *Cystoseira barbata* samples to determine the level of radioactive pollution in the Black Sea were carried out by Mihai et al. (1999), Strezov & Nonova (2005), Güven et al. (1993) and Topçuoğlu et al. (2001). However, no further studies were found for following years. In this study, it was aimed to determine the activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ ,  $^{137}\text{Cs}$  and  $^{40}\text{K}$  in *Cystoseira barbata* samples obtained from the Eastern Black Sea coast of Türkiye. Additionally, annual effective radiation dose values resulting from the ingestion of these samples were determined.

## 2. Material and method

*Cystoseira barbata* samples shown in Figure 1a were harvested from forty three stations near the coastal areas of Artvin, Rize, Trabzon, Giresun, Ordu and Samsun provinces shown in Figure 1b. The samples were washed in sea water at the sampling stations and removed from foreign substances such as gravel, sand and seaweed. The samples were brought to the laboratory into polyethylene bags and dried in an oven at 80°C for 12 hours. The dried samples were ground into small pieces and placed into 100 ml plastic containers. After radioactive equilibrium gamma analyzes of the samples were carried out with high purity germanium detector (ORTEC Gem55P4-95) system in the Recep Tayyip Erdoğan University nuclear physics laboratory.



**Figure 1.** a) One of the *Cystoseira barbata* samples examined in this study b) Sampling area

To determine the activity concentrations of radionuclides ( $A_r$ ), Equation 1 was used, where  $N_r$ ,  $\epsilon$ ,  $Y$ ,  $m$  and  $t$  indicate the net counting area of the relevant peak, detector efficiency, branching rate (%), sample mass and counting time, respectively (Baykara & Doğru, 2009). For the  $^{226}\text{Ra}$  activity concentration 295.2 keV, 352 keV ( $^{214}\text{Pb}$ ) and 609.4 keV ( $^{214}\text{Bi}$ ) were taken into account.  $^{232}\text{Th}$  activity was calculated by using peaks at energies of 238.6 keV ( $^{212}\text{Pb}$ ), 583.1 keV ( $^{208}\text{Tl}$ ) and 911.1 keV ( $^{228}\text{Ac}$ ).  $^{137}\text{Cs}$  and  $^{40}\text{K}$  activity concentrations were found from peaks at energies of 661.6 keV and 1460.7 keV, respectively (Akçay, 2013).

$$A_r (\text{Bq.kg}^{-1}) = \frac{N_r}{m \cdot t \cdot \epsilon \cdot Y} \quad (1)$$

$^{152}\text{Eu}$  source was used for calibration of the detector. The minimum detection limits (mdl) of the detection system for radionuclides were calculated with Equation 2 where  $\epsilon$ ,  $Y$ ,  $m$  and  $t$  indicate the detector efficiency, branching rate (%), sample mass and counting time, respectively (IAEA, 1989). Minimum detection limits for  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ ,  $^{137}\text{Cs}$  and  $^{40}\text{K}$  were obtained as 2.9, 1.2, 0 and 8.4  $\text{Bq.kg}^{-1}$ , respectively.

$$\text{mdl} (\text{Bq.kg}^{-1}) = \frac{4.66 \sqrt{\text{Background}}}{m \cdot t \cdot \epsilon \cdot Y} \quad (2)$$

Since macroalgae are used in many areas such as in fish feed and food industry, they can be included in human nutrition directly or indirectly (Tejera et al., 2019). Therefore, the annual effective dose ( $D_r$ ) values that could occur due to consumption of the samples were calculated using Equation 3 where  $U_i$ ,  $A_r$  and  $g_r$  denotes the annual consumption rate (assuming one  $\text{kg.y}^{-1}$ ), activity concentration of the radionuclide and dose conversion coefficient for ingestion for radionuclide, respectively (Canbazoglu & Doğru, 2013; ICRP, 1996).

$$D_r (\mu\text{Sv.y}^{-1}) = U_i \cdot A_r \cdot g_r \quad (3)$$

### 3. Results and discussions

The  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ ,  $^{137}\text{Cs}$  and  $^{40}\text{K}$  activity concentrations of the samples harvested from sampling stations were indicated in Table 1. According to Table 1, the detected activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ ,  $^{137}\text{Cs}$  and  $^{40}\text{K}$  were found in the range of 3.3-33.4, 1.7-24.1, 0.9-4.3 and 48.6-2003.1 Bq.kg<sup>-1</sup>, respectively. The highest  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ ,  $^{137}\text{Cs}$  and  $^{40}\text{K}$  activity concentrations were measured in the samples coded with C18 (Trabzon-Araklı), C20 (Trabzon, Arsin), C10 (Rize, Çiftekavak) and C15 (Trabzon-Of), respectively.

**Table 1.**  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ ,  $^{137}\text{Cs}$  and  $^{40}\text{K}$  activity concentrations of *Cystoseira barbata* samples (Bq.kg<sup>-1</sup>)

Code	Province	Station	Activity concentration (Bq.kg <sup>-1</sup> )			
			$^{226}\text{Ra}$	$^{232}\text{Th}$	$^{137}\text{Cs}$	$^{40}\text{K}$
C1	Artvin	Sarp	mdl	mdl	2.8 ± 0.4	1287.6 ± 18.5
C2	Artvin	Kemalpaşa	mdl	mdl	2.3 ± 0.3	1602.7 ± 26.3
C3	Artvin	Hopa	mdl	14.7 ± 2.9	2.6 ± 0.3	1211.8 ± 15.3
C4	Artvin	Arhavi	16.9 ± 2.6	20.2 ± 5.1	3.6 ± 1.1	555.7 ± 30.1
C5	Rize	Fındıklı	mdl	5.6 ± 1.0	3.9 ± 0.9	1225.3 ± 34.2
C6	Rize	Ardeşen	mdl	9.2 ± 1.5	3.4 ± 0.7	1608.7 ± 33.3
C7	Rize	Pazar	mdl	mdl	4.1 ± 0.4	710.5 ± 8.7
C8	Rize	Çayeli	mdl	mdl	nd	221.3 ± 8.2
C9	Rize	Merkez	mdl	2.4 ± 0.4	2.7 ± 0.3	1255.9 ± 18.1
C10	Rize	Çiftekavak	6.7 ± 1.2	1.9 ± 0.2	4.3 ± 0.5	1631.1 ± 23.1
C11	Rize	Derepazarı	6.8 ± 1.6	2.8 ± 0.5	2.8 ± 0.5	1748.5 ± 24.6
C12	Rize	İyidere	23.2 ± 1.9	13.7 ± 2.2	3.4 ± 0.9	557.6 ± 28.2
C13	Trabzon	Eskipazar	6.0 ± 0.9	5.7 ± 1.1	2.4 ± 0.4	1574.8 ± 21.9
C14	Trabzon	Kıyıcık	15.4 ± 1.3	15.7 ± 3.2	nd	514.32 ± 27.2
C15	Trabzon	Of	9.6 ± 1.8	5.9 ± 1.1	3.6 ± 0.6	2003.1 ± 28.8
C16	Trabzon	Yeniay	7.8 ± 0.9	7.6 ± 1.4	2.1 ± 0.5	49.4 ± 4.4
C17	Trabzon	Sürmene	24.2 ± 3.2	8.2 ± 1.6	3.1 ± 0.5	1633.1 ± 27.6
C18	Trabzon	Araklı	33.4 ± 4.1	10.3 ± 1.7	1.8 ± 0.7	913.3 ± 29.2
C19	Trabzon	Yeşilyalı	6.5 ± 0.9	5.5 ± 1.1	3.3 ± 0.5	135.1 ± 7.9
C20	Trabzon	Arsin	8.9 ± 0.9	24.1 ± 3.8	nd	281.3 ± 12.9
C21	Trabzon	Yomra	9.3 ± 0.9	3.5 ± 0.5	2.7 ± 0.4	395.3 ± 11.3
C22	Trabzon	Yalıncağ	6.9 ± 0.9	9.6 ± 1.5	2.6 ± 0.6	446.1 ± 18.1
C23	Trabzon	Merkez	6.5 ± 0.8	5.7 ± 1.1	2.4 ± 0.4	913.9 ± 20.8
C24	Trabzon	Beşirli	mdl	2.9 ± 0.2	2.5 ± 0.8	107.4 ± 8.5
C25	Trabzon	Akyazı	9.5 ± 1.4	8.5 ± 1.6	1.8 ± 0.5	233.5 ± 11.7
C26	Trabzon	Akçaabat	7.8 ± 1.0	7.5 ± 1.3	3.2 ± 0.9	91.8 ± 7.1
C27	Trabzon	Salacık	mdl	20.8 ± 4.3	3.9 ± 0.9	333.3 ± 25.4
C28	Trabzon	Akçakale	mdl	1.7 ± 0.2	1.2 ± 0.3	389.8 ± 12.5
C29	Trabzon	Mersin	4.4 ± 0.6	3.6 ± 0.1	nd	136.8 ± 7.5
C30	Trabzon	Gülbahçe	6.8 ± 1.0	6.7 ± 1.2	1.7 ± 0.5	111.6 ± 7.9
C31	Trabzon	Çarşıbaşı	mdl	2.7 ± 0.5	nd	234.5 ± 9.8
C32	Trabzon	Beşikdüzü	mdl	5.2 ± 1.3	2.4 ± 0.4	429.2 ± 12.0
C33	Giresun	Eynesil	mdl	4.7 ± 0.8	0.9 ± 0.2	152.6 ± 6.2
C34	Giresun	Çavuşlu	mdl	5.0 ± 0.9	nd	195.5 ± 10.1
C35	Giresun	Görele	mdl	4.5 ± 1.0	3.7 ± 0.5	856.2 ± 18.3
C36	Giresun	Yalıköy	mdl	19.0 ± 3.6	nd	99.1 ± 7.7
C37	Giresun	Tirebolu	3.3 ± 0.9	5.3 ± 0.9	3.6 ± 0.6	175.8 ± 8.1
C38	Giresun	Keşap	10.0 ± 1.1	3.0 ± 0.4	2.7 ± 0.5	48.6 ± 3.8
C39	Ordu	Gülyalı	3.3 ± 0.6	5.8 ± 1.1	2.0 ± 0.5	648.6 ± 17.4
C40	Ordu	Fatsa	mdl	2.4 ± 0.4	1.6 ± 0.5	790.0 ± 17.4
C41	Samsun	Merkez	6.2 ± 1.5	2.9 ± 0.3	nd	606.7 ± 0.3
C42	Samsun	Atakent	mdl	3.6 ± 0.5	1.3 ± 0.2	785.2 ± 14.9
C43	Samsun	Yakakent	29.6 ± 3.4	8.6 ± 1.4	3.6 ± 0.9	796.3 ± 35.6
minimum			mdl	mdl	nd	48.6
maximum			33.4	24.1	4.3	2003.1
average			11.2	7.61	2.74	690.7

mdl: under minimum detection limit, nd: not detected

According to provinces calculated average  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ ,  $^{137}\text{Cs}$  and  $^{40}\text{K}$  activity concentrations were shown in Table 2. As seen in Table 2, the highest average activity concentration of  $^{232}\text{Th}$  and  $^{40}\text{K}$  were found for Artvin,  $^{226}\text{Ra}$  and  $^{137}\text{Cs}$  were found for Samsun and Rize, respectively.

In this study, average  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ ,  $^{137}\text{Cs}$  and  $^{40}\text{K}$  activity concentrations were found as 11.2, 7.61, 2.74 and 690.7  $\text{Bq.kg}^{-1}$ , respectively. In the [UNSCEAR \(2000\)](#) report, reference values for  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  activities in vegetables were stated as 50 and 15  $\text{mBq.kg}^{-1}$ , respectively. The average  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  activities determined in this study are well above than these values. However, the activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ ,  $^{137}\text{Cs}$  and  $^{40}\text{K}$  in the samples are comparable to the results obtained in similar studies.

In Table 3, the average  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ ,  $^{137}\text{Cs}$  and  $^{40}\text{K}$  activity concentrations calculated for this study were compared with the similar results obtained in different researches. According to Table 3, it can be seen that the average  $^{226}\text{Ra}$  concentration for this study was higher than the samples obtained from Spain by [Tejera et al. \(2019\)](#), [Sudan by Sam et al. \(1998\)](#), and [Syria by Al Masri et al. \(2003\)](#), and lower than obtained from İskenderun by [Varinlioğlu et al. \(1997\)](#). However, the average  $^{226}\text{Ra}$  activity concentration in this study is compatible with the  $^{226}\text{Ra}$  activities determined for samples obtained from Bulgaria ([Strezov & Nonova, 2005](#)) and Romania ([Mihai et al., 1999](#)).

As seen in Table 3, the average  $^{232}\text{Th}$  activity in this study is greater than the  $^{232}\text{Th}$  activity in the samples obtained from Romania ([Mihai et al., 1999](#)) and Sudan ([Sam et al., 1998](#)), and is lower than the samples from Spain ([Tejera et al., 2019](#)) and İskenderun ([Varinlioğlu et al., 1997](#)).

**Table 2.** Average  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ ,  $^{137}\text{Cs}$  and  $^{40}\text{K}$  activity concentrations and annual effective ingestion dose values for *Cystoseira barbata* samples according to provinces

Province	Activity concentration ( $\text{Bq.kg}^{-1}$ )				Annual effective dose ( $\mu\text{Sv.y}^{-1}$ )				
	$^{226}\text{Ra}$	$^{232}\text{Th}$	$^{137}\text{Cs}$	$^{40}\text{K}$	$\text{D}_{\text{Ra-226}}$	$\text{D}_{\text{Th-232}}$	$\text{D}_{\text{Cs-137}}$	$\text{D}_{\text{K-40}}$	$\text{D}_{\text{Total}}$
Artvin	16.9	17.5	2.83	1164.5	4.73	4.03	0.04	7.22	10.45
Rize	12.2	5.9	3.51	1119.9	3.42	1.36	0.05	6.94	9.29
Trabzon	10.9	8.1	2.54	546.4	3.05	1.86	0.03	3.39	7.55
Giresun	6.7	6.9	2.72	254.6	1.86	1.59	0.04	1.58	3.81
Ordu	3.3	4.1	1.80	719.3	0.92	0.94	0.02	4.46	5.89
Samsun	17.9	5.0	2.45	729.4	5.01	1.15	0.03	4.52	9.04
minimum	3.3	4.1	1.80	254.6	0.92	0.94	0.02	1.58	3.81
maximum	17.9	17.5	3.51	1164.5	5.01	4.03	0.05	7.22	10.45

**Table 3.** Comparison of activity concentrations in *Cystoseira* samples with various studies

Location	Activity concentration ( $\text{Bq.kg}^{-1}$ )				Reference
	$^{226}\text{Ra}$	$^{232}\text{Th}$	$^{137}\text{Cs}$	$^{40}\text{K}$	
Bulgaria	2.5-17	-	3-10	1025-2040	<a href="#">Strezov &amp; Nonova, 2005</a>
Greece	-	-	20.3-250.5	-	<a href="#">Sawidis et al., 2003</a>
Romania	5.7-11.8	0.83-1.40	3.9-8.9	-	<a href="#">Mihai et al., 1999</a>
Spain	9	10.2	-	940	<a href="#">Tejera et al., 2019</a>
Sudan	3.7	0.61	0.85	827.5	<a href="#">Sam et al., 1998</a>
Syria	1.2	-	<0.47	2260	<a href="#">Al Masri et al., 2003</a>
Amasra	-	-	<3	328	<a href="#">Topçuoğlu et al., 2001</a>
Beşikdüzü	-	-	15	340	<a href="#">Güven et al., 1993</a>
Çayeli	-	-	15	430	
İğneada	-	-	<3	869	<a href="#">Topçuoğlu et al., 2001</a>
İskenderun	19-33	15-25	1-1.7	740-1100	<a href="#">Varinlioğlu et al., 1997</a>
Pazar	-	-	<3	1180	
Rize	-	-	<3	1122	<a href="#">Topçuoğlu et al., 2001</a>
Sarp	-	-	7	1579	<a href="#">Güven et al., 1993</a>
Şile	-	-	13	826	<a href="#">Güven et al., 1990</a>
Ünye	-	-	<3	543	<a href="#">Topçuoğlu et al., 2001</a>
Eastern Black Sea	11.2	7.61	2.74	690.7	Present study

According to Table 3, it can be seen that the average  $^{40}\text{K}$  activity concentration determined in this study is lower than the  $^{40}\text{K}$  activity in the samples obtained from Bulgaria (Strezov & Nonova, 2005), Spain (Tejera et al., 2019), Sudan (Sam et al., 1998), Syria (Al Masri et al., 2003), İğneada, Pazar, Rize (Topçuoğlu et al., 2001), İskenderun (Varinlioğlu et al., 1997), Sarp (Güven et al., 1993) and Şile (Güven et al., 1990) and higher than from Amasra, Ünye (Topçuoğlu et al., 2001), Beşikdüzü and Çayeli (Güven et al., 1993).

As seen in Table 3, the average  $^{137}\text{Cs}$  activity concentration determined for this study is higher than the  $^{137}\text{Cs}$  activity in the samples obtained by Sam et al. (1998) from Sudan, Al Masri et al. (2003) from Syria and Varinlioğlu et al. (1997) from İskenderun and lower than the all remaining studies in Table 3 (Strezov & Nonova, 2005; Sawidis et al., 2003; Mihai et al., 1999; Topçuoğlu et al., 2001; Güven et al., 1990; Güven et al., 1993). However, the average  $^{137}\text{Cs}$  activity concentration calculated for this study is significantly lower than the value of  $1000 \text{ Bq.kg}^{-1}$  allowed for  $^{137}\text{Cs}$  in food samples by ICRP (1996).

According to UNSCEAR (2000) report the average annual effective dose value that people are exposed to from natural radiation sources through ingestion is  $0.29 \text{ mSv.y}^{-1}$ . The average annual effective dose from samples due to  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ ,  $^{137}\text{Cs}$  and  $^{40}\text{K}$  were found as 3.14, 1.75, 0.04 and  $4.28 \text{ }\mu\text{Sv.y}^{-1}$ , respectively. Total annual effective ingestion dose values from the samples were found to be in the range of  $1.37\text{-}18.83 \text{ }\mu\text{Sv.y}^{-1}$  with average value as  $7.65 \text{ }\mu\text{Sv.y}^{-1}$ . The highest total annual effective dose was determined for the sample coded with C17 (Trabzon-Sürmene) and the lowest for both C8 (Rize-Çayeli) and C24 (Trabzon-Beşirli).

For provinces the highest annual effective dose values due to  $^{232}\text{Th}$  and  $^{40}\text{K}$  were found for Artvin,  $^{226}\text{Ra}$  and  $^{137}\text{Cs}$  were found for Samsun and Rize, respectively. Additionally, the lowest annual effective dose values due to  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{137}\text{Cs}$  were calculated for Ordu, and  $^{40}\text{K}$  for Giresun. Total annual effective dose values resulting from all radionuclides were found to be in range between 3.81 (Giresun) and  $10.45 \text{ }\mu\text{Sv.y}^{-1}$  (Artvin). All the annual effective ingestion dose values calculated for this study are much lower than  $0.29 \text{ mSv.y}^{-1}$  (UNSCEAR, 2000).

#### 4. Conclusions

In this study, average  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  activity concentrations of *Cystoseira barbata* samples obtained from the Eastern Black Sea coast of Türkiye were found as 11.2, 7.61 and  $690.7 \text{ Bq.kg}^{-1}$ , respectively. The highest  $^{137}\text{Cs}$  activity concentration was calculated as  $4.3 \text{ Bq.kg}^{-1}$  in C10 (Rize-Çiftেকavak). It was observed that all detected  $^{137}\text{Cs}$  activities were significantly lower than the limit value of  $1000 \text{ Bq.kg}^{-1}$  which was allowed by ICRP (1996) for  $^{137}\text{Cs}$  in food samples.

$^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  are the main radionuclides that found naturally in the earth's crust, including marine environments. Soil transport to the seas as a result of erosion, waste from the construction industry and household and the use of fertilizers contribute to the increase in the amount of these radionuclides in the seas. Nevertheless, in studies conducted to determine radioactive pollution in the Black Sea, activity concentrations of artificial radionuclides originating from the Chernobyl accident were generally examined. Therefore,  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  levels were not determined in many studies conducted in Turkey related with *Cystoseira barbata* samples. This study provides important data about  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ ,  $^{137}\text{Cs}$  and  $^{40}\text{K}$  levels in *Cystoseira barbata* samples harvested from the Eastern Black Sea coast of Türkiye with its wide sampling locations.

The average  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  activity concentrations in this study were found to be higher than the reference values stated by UNSCEAR (2000) as  $50 \text{ mBq.kg}^{-1}$  and  $15 \text{ mBq.kg}^{-1}$  for  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  in vegetable samples, respectively. Despite this, it was observed that all detected  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ ,  $^{137}\text{Cs}$  and  $^{40}\text{K}$  activity concentrations were at a level comparable to the results obtained in similar studies conducted in various regions of the world.

Total annual effective dose values resulting from the samples examined in this study were calculated in the range of  $1.37\text{-}18.83 \text{ }\mu\text{Sv.y}^{-1}$ . According to the provinces, the highest total annual effective dose value was determined for Artvin as  $10.45 \text{ }\mu\text{Sv.y}^{-1}$  and the lowest for Giresun as  $3.81 \text{ }\mu\text{Sv.y}^{-1}$ . All the calculated annual effective dose values were found to be well below  $0.29 \text{ mSv.y}^{-1}$  stated by UNSCEAR (2000) as the average

annual effective ingestion dose received through natural radiation sources worldwide. For this reason, it can be said that the samples examined in this study do not pose any radiological risk for human health.

*Cystoseira barbata* is a brown macroalgae that can grow to many meters, cling to rocks and withstand to strong waves. In this study, it is seen that *Cystoseira barbata* is an useful bioindicator used in determining of radioactive pollution in the seas and comparing the results obtained from similar studies conducted in various parts of the world. Additionally, it can be stated that the differences between the detected activity concentrations can be due to the concentrations of radionuclides in the environments where the samples were obtained and the radionuclide retention capacity of the macroalgae.

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### Author contribution

The first author contributed to the literature research, providing of samples, preparation and writing of the article. The second author contributed to the consultancy process during the writing of the thesis and the article.

### Declaration of ethical code

The authors of this article declare that the materials and methods used in this study do not require ethical committee approval or legal-specific permission.

### Conflicts of interest

The authors of this article declare that no conflict of interest.

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