

THE GROSS ALPHA AND GROSS BETA ACTIVITIES OF HOLOCENE MARINE SEDIMENTS AND THE RELATIONSHIP OF THESE ACTIVITIES WITH RADIONUCLIDES IN THE WESTERN MARMARA SEA, TURKEY

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Abstract. The aim of this study is to analyze the gross alpha and gross beta activities of Holocene marine sediments and to determine the relationship of these activities with radionuclides in the west of Marmara Sea (Turkey). Background and anthropogenic ionizing radiations are the main sources of environmental radioactivity human beings are exposed to. Background radiation occurs as a result of cosmogenic and terrestrial radiation, while anthropogenic refers to man-made radiation. In this context, gross alpha and beta activity of sea sediment samples collected at 29 different stations in the research region were analyzed using the low-background counter (Berthold, LB 770, 10- channel α - β low-level counter). Also, the relation between abundance of radioactive isotopes and alpha and beta activity were discussed in this paper. As a result, the average gross alpha and beta activities were determined as 223.18 ± 14.9 Bq kg⁻¹ and 572.00 ± 23.9 Bq kg⁻¹, respectively. The results obtained can be used as a data base for Holocene marine sediments.

Keywords: radionuclide, radioactivity, radioactivity pollution, Chernobyl accident K-40, Sr-90, Cs-137, Marmara Sea

Introduction

Ionizing radiation pollution which is mainly caused by background and anthropogenic radiations is one of the recent environmental problems. Background radiation originates from cosmogenic and terrestrial radiations, while anthropogenic means man-made. The recent increase in the levels of environmental ionizing radiation pollution is directly related with the industrialization such as the use of fossil fuels, phosphate industry and wrong and excessive use of fertilizers (Topcuoğlu et al., 2010). The radioactive elements are found in magmatic rocks, in phosphate rocks, in sedimentary masses such as clay and shale, and in granitic rocks in high amounts. But

these elements are found in small quantities in Sedimentary rocks such as sand-gravel, sandstone, and limestones with crack (Kumru et al., 2002). The source of radioactivity in the aquatic environment is water pollution which occurs with particles and minerals present in water. The geological structure of the area where the water deposit is located is also a determinant of natural radioactivity. It differs in radioactivity levels according to the quality of the rocks forming the Earth's crust (Karaman, 2003). The knowledge of environmental natural radioactivity is very important in terms of determining where the nuclear accidents and industrialization pollute the environment (Degerlier, 2007). For example, after the Chernobyl accident, anthropogenic radionuclides entered the marine environment. The atmospheric sediments spread from the nuclear weapon tests in the countries around the Black Sea reached the Marmara Sea through the Bosphorus (Topcuoğlu, 2001).

The alpha particle is a helium nucleus with an electric charge (+2). Due to its large mass, it is unlikely to penetrate into matter. Alpha particles are emitted from natural radioactive isotopes such as polonium, thorium, radium, uranium, which are formed in nature and have large atomic weights. The ionization betting that the alpha rays bring into the matter is relatively high. The gross alpha radioactivity consists of U-238, Th-232, Ra-226, Rn-222 and their alpha emitter nuclei. The gross beta radioactivity is due to fission and activation products such as natural long-lived K-40, Sr-90, Cs-137, Co-60 and I-125 (Varol, 2011). Beta particles are negatively charged and have smaller mass than alpha particle. As soon as one of the neutrons in the nucleus of the isotope changes, beta degradation occurs immediately. As it carries ionizing properties, it has harmful effects on living organisms (Varol, 2011). The sediments play an important role in the aquatic environment, because of their geological and chemical structure, which provides the flow system between land, sea and atmosphere (Kurt et al., 2016). Determination of radioactivity in sediment samples provide information on the level of sea water radioactivity the sediment is receiving. One of the most commonly used methods for determining environmental radiation is gross alpha and gross beta measurements, which are the result of quick access and low cost (Bunz and Kracke, 1998). Numerous studies have been conducted in the past years on the identification of environmental radioactivity and the monitoring of pollution using heavy metals and radioactivity in marine environments (Taskin, 2006; Bozkurt et al., 2007; Varol, 2011; Agbalagba et al., 2013; Cam et al., 2013; Kaya et al., 2016; Kurt et al., 2016; Yümün, 2016; Kam et al., 2016; Yümün et al., 2016, 2017; Yümün and Önce, 2017).

In the studies given in the above paragraph, gross alpha and beta activity measurements were performed for groundwater and drinking waters and also radionuclides in sediments were examined. In this study, gross alpha and beta activity of sea sediment samples collected from 29 different stations in the Western Marmara Sea (Recent marine sediments) were analyzed and environmental radioactivity concentration values were determined. Also, the traces of nucleotides, such as Cs-137, which gave rise greatly to the pollution of marine environments, was discussed.

Material and Methods

The sampling stations were between Silivri (Istanbul) - Bandırma (Balıkesir) in Western Marmara Sea which is at the west part of Turkey (*Fig. 1*). The bathymetry of the study area varies between 20.00 m and 35.00 m in core samples. Core samples from 29 locations were used in the study. The samples were taken between 15-20.10.2015

and the coordinates and depth information of the samples are given in *Table 1*. Because the samples are obtained from about 30 m, the samples are in the form of fine-grained, sandy-silty clay (Yümün, 2017). Sediment samples were immediately taken in a nylon bag and protected from atmospheric moisture. Later, the samples were taken to the laboratory, where they were removed from the nylon bags and dried in room temperature.

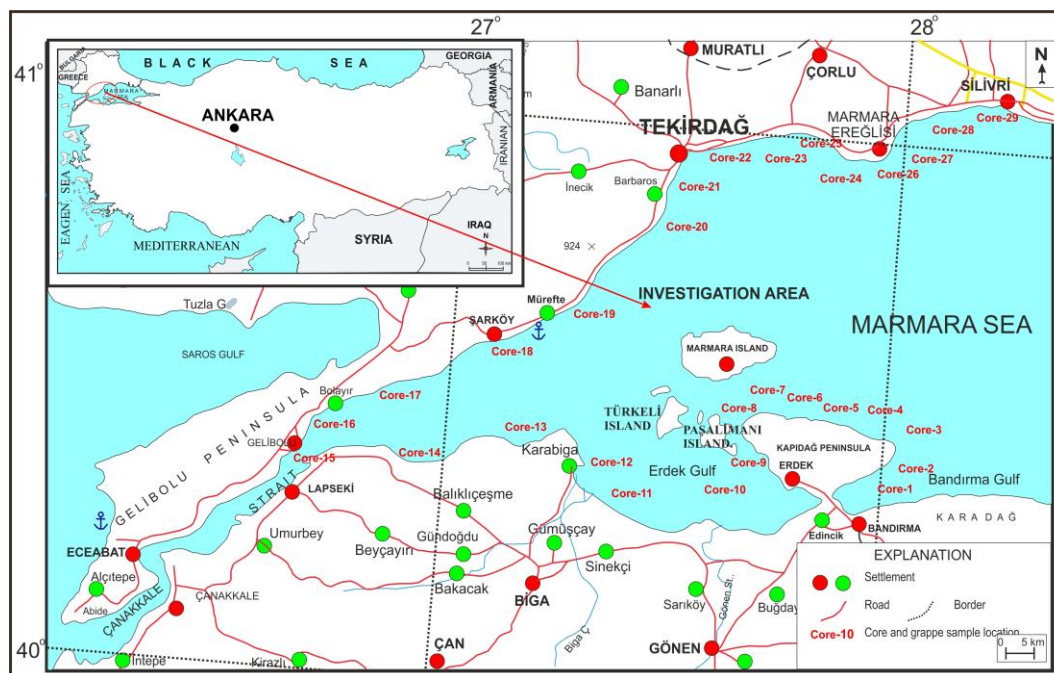


Figure 1. Location map of investigation area and sample's locations

The dried sediment samples were ground in granite bowls and sieved with a stainless-steel sieve to remove the portion that came to the size of the powder. Approximately 100 mg of each sample was weighed on a precision scale and placed in an empty aluminum bowl. Distilled pure water has been used to homogeneously distribute the samples in the bowl. The specimens were dried for 2 hours at 105 °C, then left in the desiccator to cool to room temperature without drawing any moisture. Then, the prepared samples were homogenized with distilled water and placed in the oven. This process is important to determine the amount of residue. If the amount of the residue is more than 400 mg, the self-absorption effect of the alpha particles increases. In this way, fewer particles will reach the detector, causing the results to be erroneous (Taskin, 2006). Finally, the gross alpha and gross beta activity rates of all samples were measured using two low-level LLB 770-PC 10-channel alpha-beta counting systems in two cycles of 500 minutes each. LLB 770-PC 10-channel alpha-beta counting systems have been produced by Contact Berthold Technologies GmbH & Co. in Germany. Each analysis was repeated twice and the arithmetic mean of these measurements was used. The samples were taken from the sea using a boat and the gravity core method. The diameter of the samples (54 mm) and the sampling depths are given in *Table 1*.

Table 1. Gross alpha and beta activity concentration of sediments collected from Western Marmara Sea

Sample No	Gross Alpha (Bq/kg)	Uncertainty (±)	Gross Beta (Bq/kg)	Uncertainty (±)	Geographic Position (WGS-84)		Sample Depth (m)
					Y	X	
CORE-1	311.76	17.6	900.41	30	0581459	4474677	30,
CORE-2	259.45	16.1	585.45	24.1	0584985	4476692	29
CORE-3	135.2±11	11	338.19	18.3	0588105	4481639	30
CORE-3b	265.58	16.2	768.23	27.7	0586229	4482780	35
CORE-4	240.88	15.4	38.38	6.1	0582393	4483551	30
CORE-4b	286.8	16.9	848.05	29.1	0582260	4483937	32
CORE-4c	119.12	10.9	456.56	21.3	0579835	4485462	35
CORE-5	282.76	16.7	640.28	25.2	0577401	4485335	38
CORE-6	309.42	17.5	875.06	29.5	0566840	4486334	40
CORE-7	149.43	12.2	344.29	18.5	0562101	4486810	48
CORE-8	32.23	5.6	154.99	12.4	0558197	4484862	39,5
CORE-9	184.99	13.5	658.76	25.6	0558282	4480129	28
CORE-9b	269.92	16.4	667.47	25.8	055 83 61	4479791	32
CORE-10	253.39	15.9	613.51	24.7	0556281	4463687	19
CORE-11	152.24	12.3	475.71	21.7	0542436	4464634	26
CORE-12	608.41	24.6	1320.14	36.3	0528796	4470648	18
CORE-13	121.04	11	496.28	22.7	0514452	4478784	46
CORE-14	307.22	17.5	753.74	27.4	0490282	4473347	29
CORE-15	137.53	11.7	563.11	23.7	0473914	4469971	41
CORE-16	178.87	13.3	150.33	12.2	0472908	4472487	30
CORE-17	158.37	12.5	591.22	24.3	0488099	4481025	33
CORE-18	166.42	12.8	374.49	19.3	0511008	4494809	32
CORE-19	134.78	11.5	459.08	21.4	0540197	4522435	30
CORE-20	347.03	15.6	970.66	31.1	0542591	4530386	32
CORE-21	164.15	11.3	447.84	21.1	0546815	4533986	33
CORE-22	106.34	18.1	117.65	11.8	0560139	4536953	33
CORE-23	104.22	14.6	212.23	12.2	0569767	4535792	32
CORE-24	127.64	14.7	307.82	26.5	0579050	4532627	30
CORE-25	419.99	20.4	321.19	28.6	0581005	4534924	33
CORE-26	66.34	8.1	115.65	10.8	0585198	4539137	32
CORE-27	114.3	10.6	252.93	11.8	0588549	4541739	30
CORE-28	167.64	12.9	707.82	16.5	0596407	4543970	31
CORE-29	419.99	20.4	821.19	18.6	0604571	4544155	35

Gross Alpha and Beta concentration obtained from analysis results have been analyzed by Kriging method (*Figures 2 and 3*). The Kriging method is an interpolation method that estimates the optimal values of the data at one point using the data from the other known points (Inal et al., 2002). Kriging method is a technique in which the estimation of spatial changes in not sampled points is made optimally using semi variogram structural properties (Trangmar et al., 1985). The activities were obtained using the following formulas:

$$Activity(a) = \frac{NetCounting}{60 \cdot Efficiency \cdot Mass(kg)} \quad (Eq.1)$$

$$Activity(b) = \frac{N(a) - B(crosstalk)}{60 \cdot Efficiency \cdot Mass(kg)} \quad (Eq.2)$$

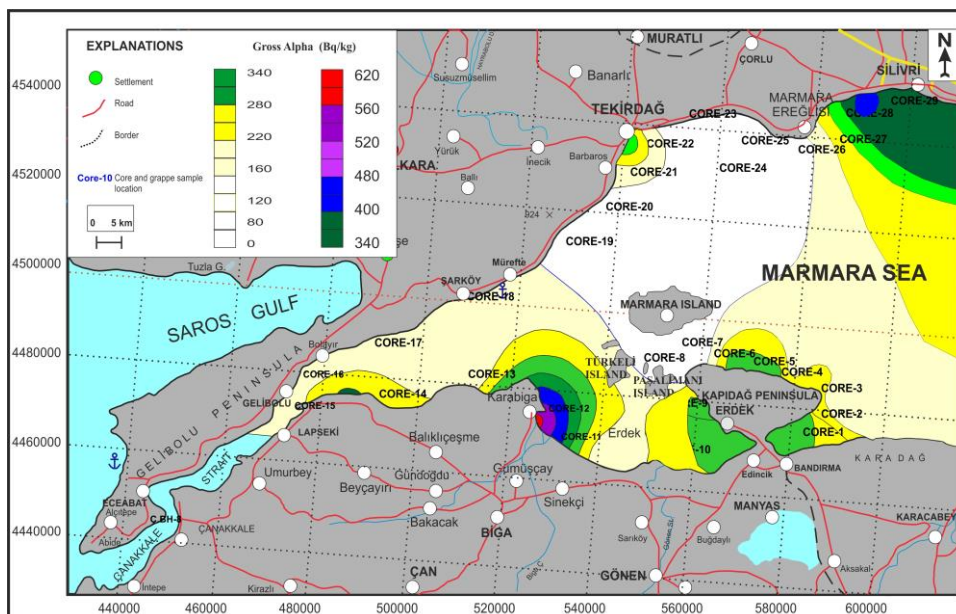


Figure 2. Map of gross alpha distribution of investigation area (Western Marmara Sea)

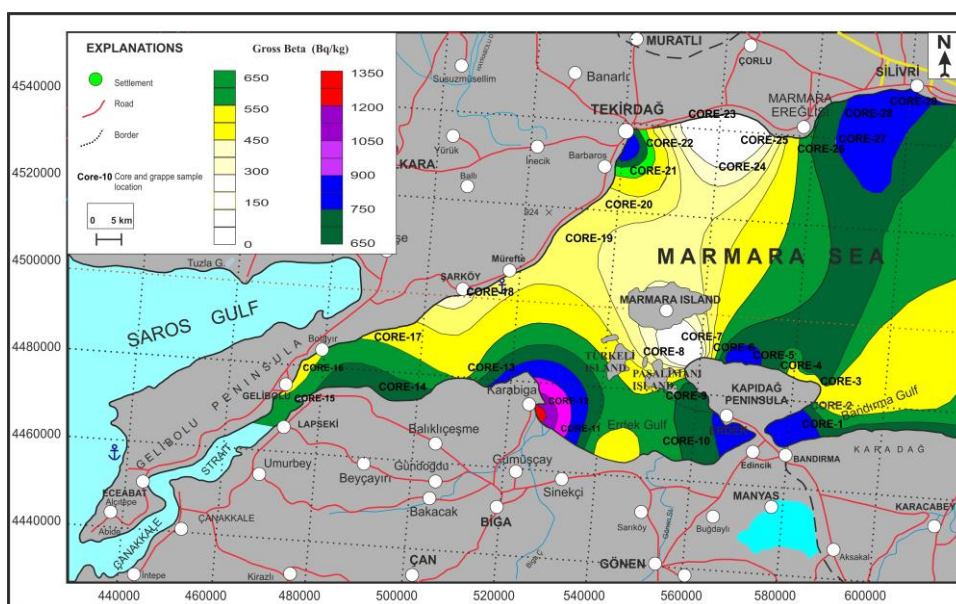


Figure 3. Map of Gross Beta distribution of investigation area (Western Marmara Sea)

Low level counting system is commonly used for measuring environmental samples with low natural radiation. Its calibration has been carried out with standard solutions, which contained known activities of ^{241}Am for alphas and ^{90}Sr for betas which are similar to the sample geometry (Currie, 1968). Gross alpha radioactivity is composed of radionuclides (U-238, Th-232, Ra-226, Rn-222) and their alpha emitting nuclei, Gross beta radioactivity is of natural long-lived radionuclides (K-40, Sr-90, Cs-137, Co-60 and I-125) are known to originate from fission and activation products (Table 2). For this reason, the causes of Gross alpha and Gross Beta activities were evaluated by taking the radionuclide values of the samples of this study.

Table 2. Results of gamma spectrometric analyses of core samples (Yümin and Kam, 2020)

Sample	Radionuclide	Activity ± Bq/kg	Gross Alpha (Bq/kg)	Gross Beta (Bq/kg)	Sample	Radionuclide	Activity ± Bq/kg	Gross Alpha (Bq/kg)	Gross Beta (Bq/kg)		
CORE-1	⁴⁰ K	490	311.76	900.41	CORE-13	²³² Th	45	269.92	667.47		
	¹³⁷ Cs	2.5				²²⁶ Ra	25.7				
	²²⁶ Ra	30				¹³⁷ Cs	2.2				
	²³² Th	42				⁹⁵⁺ Zr	0.27				
CORE-2	¹³⁷ Cs	1.9	259.45	585.45	CORE-14	⁴⁰ K	470	253.39	613.51		
	²²⁶ Ra	19.8				²³² Th	36				
	²³² Th	27.1				²²⁶ Ra	25.2				
CORE-3	²²⁶ Ra	25	135.2	338.19	CORE-15	¹³⁷ Cs	0.90	152.24	475.71		
	²³² Th	28				²²⁶ Ra	19.5				
CORE-4	⁴⁰ K	24.4	265.58	768.23	CORE-16	²³² Th	36	608.41	1320.14		
	¹³⁷ Cs	1.4				⁴⁰ K	600				
	²³² Th	21.6				¹³⁷ Cs	4.7				
	⁵⁴ Mn	0.71				²²⁶ Ra	24.9				
CORE-5	⁴⁰ K	470	240.88	38.38	CORE-17	²³² Th	37	121.04	496.28		
	¹³⁷ Cs	1.8				¹³⁷ Cs	1.6				
	²²⁶ Ra	21.5				²²⁶ Ra	18.9				
	²³² Th	30				²³² Th	26				
CORE-6	⁴⁰ K	450	286.8	848.05	CORE-18	¹³⁷ Cs	4.8	307.22	753.74		
	¹³⁷ Cs	1.3				²²⁶ Ra	29				
	²²⁶ Ra	27				²³² Th	36				
	²³² Th	26				⁴⁰ K	650				
CORE-7	¹³⁷ Cs	1.1	119.12	456.56	CORE-19	¹³⁷ Cs	2.3	137.53	563.11		
	²²⁶ Ra	24				²²⁶ Ra	27.9				
	²³² Th	39				²³² Th	39				
	⁹⁵⁺ Zr	0.2				⁴⁰ K	630				
CORE-8	¹³⁷ Cs	1.1	282.76	640.28	CORE-20	¹³⁷ Cs	9.4	178.87	150.33		
	²²⁶ Ra	18.6				²²⁶ Ra	20.5				
	²³² Th	30				²³² Th	35				
CORE-9	⁴⁰ K	470	309.42	875.06	CORE-21	¹³⁷ Cs	3.7	158.37	591.22		
	¹³⁷ Cs	0.9				²²⁶ Ra	20.6				
	²²⁶ Ra	19				²³² Th	30				
	²³² Th	33				¹³⁷ Cs	5.1				
CORE-10	¹³⁷ Cs	3.5	149.43	344.29	CORE-22	²²⁶ Ra	27	166.42	374.49		
	²²⁶ Ra	50				²³² Th	20.7				
	²³² Th	86				¹³⁷ Cs	1.7				
	¹⁵⁵ Eu	3.2				²²⁶ Ra	13.7				
CORE-11	⁴⁰ K	670	32.23	154.99	CORE-23	²³² Th	18.9	134.78	459.08		
	²³² Th	57				CORE-24	²²⁶ Ra			10	
	²²⁶ Ra	34					²³² Th			21	
	¹³⁷ Cs	3.8					CORE-25			⁴⁰ K	550
	⁵⁴ Mn	1.1								¹³⁷ Cs	1
²³² Th	49	²²⁶ Ra	22								
CORE-12	²²⁶ Ra	28.2	184.99	658.76	CORE-26	²³² Th	38	164.15	447.84		
	¹³⁷ Cs	4.9				⁹⁵⁺ Zr	0.19				
	⁴⁰ K	480				CORE-28	⁴⁰ K			580	
²³² Th	24	¹³⁷ Cs	1								
²²⁶ Ra	19.2	²²⁶ Ra	18.1								
¹³⁷ Cs	0.8	²³² Th	24.8								
CORE-27	¹³⁷ Cs	1.3	114.3	252.93	CORE-29	²²⁶ Ra	20.9	419.99	821.19		
	²²⁶ Ra	16.1				²³² Th	31				
	²³² Th	21.7									

Results and Discussion

The gross alpha and gross beta concentration of the obtained sediments are given in *Table 1* and the position of the sample stations are given in *Fig. 1*. According to the analysis results, gross beta values were found higher than gross alpha values at all locations except M5. The lowest and highest gross alpha and gross beta values are 32, 23-608, 41 Bq/kg (average value: 223, 18 Bq/kg) and 38, 38-1320, 14 Bq/kg (average value: 572, 00 Bq/kg), respectively. The lowest concentration of gross alpha is in M11, M26, while Beta is in M5. However, M16 is the region with the highest gross alpha and beta intensities.

Since there is no regulatory standard for radiological pollutants in sediments, the results obtained in this study are compared with those in different countries in *Table 3* and gross alpha and gross beta activity concentration of sediment samples are shown in *Figure 4*. According to *Table 3*, the gross alpha concentration of the study is higher than that in Los Alamos and Sir Dam Pond. The gross beta concentration of the western Marmara sea samples is considerably lower than the values for the Bendimahı River, but higher than the data for other locations referenced in the manuscript. Although the gross alpha and gross beta concentrations in studies conducted in other countries are close to the average value, the concentrations obtained in this study have very large ranges. This is due to the high concentration of environmental pollutants in some locations in this region. According to the locations where the samples are taken, there are differences in the analysis results. Core samples have a fine-grained, sandy-silty clayey composition, because they have been taken from areas, where higher than 30 m depth. Concentrations of radionuclides vary depending on sediment particle size, mineral composition, local pollution and organic matter content (Papaeftymiou et al., 2007).

Table 3. Gross alpha and gross beta activity concentration values compared to similar studies (1: (Pentreath, 1984), 2: (Wallove et al., 2016), 3: (Zorer, 2009), 4: (Pentreath, 1984))

Stations	Gross α (Bq/kg)	Gross β (Bq/kg)	References
<i>Pentreath, 1984</i> : Western Marmara, Turkey	32.23-608.41 (average: 223,18)	38.38-1320.14 (average: 572,00)	Present study
Los Alamos, New Mexico	55,5	55.5	<i>Wallove et al., 2016</i>
Bosna River, Bosnia	215-610 (average: 460,7)	495-628 (average: 539,1)	<i>Wallove et al., 2016</i>
Bendimahı River (May)	782-4596 (average: 2000)	482-10372 (average: 4635)	<i>Zorer, 2009</i>
Sir Dam Pond (May)	34.8-229 (average: 101)	144.1-419.3 (average: 234,3)	<i>Pentreath, 1984</i>

In this study the results generally have high values compared to similar studies which are generally referred to. K-40 is the most abundant natural radioisotope in the earth's crust and soluble in abundant amounts in seawater (Peterson, 2007). The areas where the Nilüfer and Gönen Streams flow into the Marmara Sea and the sea areas adjacent to the coasts where the settlements are located are the most polluted areas of the Marmara Sea. It has been observed that K40 values in these locations are higher than the world average values.

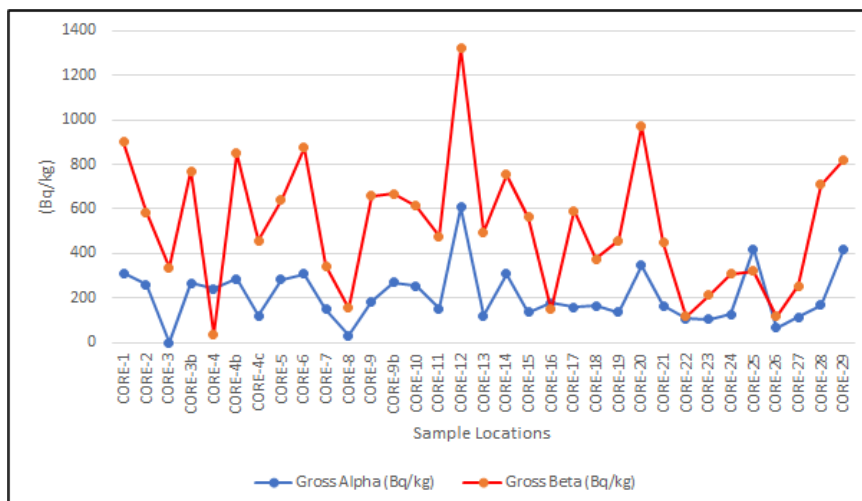


Figure 4. The gross alpha and gross beta activity concentration of sediment specimens in the Western Marmara Sea

In the graph given in *Figure 4*, it is seen that gross alpha and gross beta concentrations decreased and increased similarly in each location. In *Table 2*, gross alpha and gross beta activities were also compared with radionuclides (^{40}K , ^{137}Cs , ^{226}Ra , ^{232}Th , ^{95}Zr , ^{54}Mn) of the same samples. It is seen that gross alpha and gross beta values increase in locations where radionuclide concentrations increase, and decrease in locations where radionuclide concentrations decrease. It is thought that domestic and industrial wastes, agricultural activities and ship wastes are among the reasons for the very high concentrations of gross alpha and gross beta, especially in settlements, agricultural areas, piers and ports. Another important reason why the gross alpha and beta concentrations are generally high is that the fertilizers used in agriculture contain uranium, thorium and their decomposition products and natural potassium-40 (NCRP, 1987).

This study shows that fertilizer and agricultural pesticide residues from the soil carried by Kocasu, Gönen and Biga streams in the areas where agricultural activities are concentrated cause radioactive pollution (Kam et al., 2016). Another contribution to gross beta results is the Chernobyl nuclear reactor accident and nuclear weapon trials. Radioactive nuclei of ^{137}Cs , ^{90}Sr , ^{131}I , ^{132}Te and ^{239}Pu are spread from the nuclear reactor accident and nuclear weapon tests. The points where Gross alpha and Gross Beta activity values are high are the locations close to Çanakkale. The locations with low activity values were obtained in the samples taken near the Kapıdağ Peninsula. The high activity values may be related to the geological structure and pollution. In part, it could be the effects of the Chernobyl accident. Especially high Cs values may be due to weapon trials. After the Chernobyl accident, the vertical velocity of ^{134}Cs and ^{137}Cs in the soil was found to be 0.2-0.3 cm/hour (Bunz and Kracke, 1988).

Especially ^{137}Cs (half-life 30 years) and ^{90}Sr (half-life 29, 12 years) radionuclides have quite long half-lives. For this, background levels should be determined for the individual radionuclides that may create the potential pollution cause of the regions. The radionuclide potential of marine sediments from the same locations where this study was conducted was determined (Yümün and Kam, 2020). International average values of radionuclides are ^{232}Th : 30 Bq/kg, ^{226}Ra : 35 Bq/kg, ^{40}K : 400 Bq/kg (UNSCEAR,

2000). The radionuclide potential obtained from the study is given in *Table 2*. In *Table 2*, gross alpha and gross beta values are presented together with radionuclides and correlations are made. When the values given in *Table 2* are examined, it is seen that the gross alpha radioactivity consists of radionuclides (U-238, Th-232, Ra-226, Rn-222) and their alpha emitter cores. Again in *Table 2* it was seen that gross beta radioactivity resulted from fission and activation products such as natural long-lived radionuclides (K-40, Sr-90, Cs-137, Co-60 and I-125). However, in most of the sediment samples taken in this study, it is seen that the radionuclides are above the world average values. The high radionuclide concentrations seen here are responsible for the high gross alpha and gross beta activities. These analyses can be applied to water and sediment samples, as well as to foodstuffs (Pintilie et al., 2017) such as bread (Pintilie et al., 2018).

Conclusion

The main purpose of this study was to determine the natural radioactivity of the region by examining the gross alpha and gross beta concentrations of sediment samples from the Western Marmara Sea. In addition, studies have been carried out to define the relationship of gross alpha and gross beta activities with radionuclides. As a result, beta activity has been found to be significantly higher than alpha activity at all locations. Natural radioactivity depends directly on the regional geology, weather conditions and physicochemical variability of the water. The fact that the agricultural activities of the study area is high and is in the leading region of the country in terms of population and industry, strongly influences radioactive pollution.

Gross alpha radioactivity consists of U-238, Th-232, Ra-226, Rn-222 and their alpha emitting nuclei. The gross beta radioactivity is due to fission and activation products of natural long-lived radionuclides (K-40, Sr-90, Cs-137, Co-60 and I-125) (Varol, 2011). According to the values given in *Table 2*, Gross alpha and gross beta concentrations are also high in locations where K40 and other radionuclides are high. In addition, the presence of Cs-137, which cannot occur in the natural environment, in many locations in this study area, is thought to be a result of the Chernobyl Nuclear accident. In addition, there may have been nuclear weapon tests carried out by some countries recently. This is an important issue and needs to be taken seriously.

The data published in this paper can be the base data which also will be used to evaluate future changes. In future studies to determine the radioactive pollution of the Marmara Sea, sea water and sea sediment samples should be sampled at more frequent intervals.

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