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Radionuclide Potential of Holocene Sediments in the West of Marmara Sea (Turkey)

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Abstract: Radionuclides that cause radioactive pollution descend to the bottom in marine and water environments such as heavy metals and accumulate in bottom sediments. It is useful to determine the radionuclides in these environments to control the radionuclide release and its damage. Radioactive pollution can harm people's life directly or through the food chain. In this study, natural and artificial radionuclide values were measured in Recent sediment samples taken from the seabed in the western part of the Marmara Sea. Gamma spectrometry method was used in radionuclide examinations. In gamma spectrometry studies of sediments, values of radionuclides (^{40}K, ^{137}Cs, ^{226}Ra, ^{54}Mn, ^{95}Z, and ^{232}Th) were determined. Sea depths where 18 analyzed seafloor sediments are taken vary between 15-50 m. The determined radionuclide concentration activity values of the study area are ^{137}Cs (0.9 - 9.4 (Bq / kg)), ^{232}Th (18.9 - 86 (Bq / kg)), ^{226}Ra (10 - 50 (Bq / kg)), ^{40}K (24.4 - 670 (Bq / kg)), ^{54}Mn (0.71-0.9 (Bq / kg)) and ^{95}Zr (0.18 - 0.19 (Bq / kg)). These values were correlated with the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR). The ^{226}Ra series, ^{232}Th series, and 40K radionuclides accumulate naturally, and their concentrations increase gradually due to anthropogenic impurities. ^{226}Ra values obtained across the study areas are within normal limits according to UNSCEAR values. ^{40}K and ^{232}Th values were higher than UNSCEAR values in all locations. ^{137}Cs (0.9 - 9.4 (Bq / kg)) from almost all locations reveals a risky situation in terms of ambient conditions because this element cannot be found in the natural environment and can be found artificially as an end of radioactivity.

Keywords: ^{40}K, ^{137}Cs, ^{226}Ra, ^{54}Mn, ^{95}Z, ^{232}Th, radionuclide, Marmara Sea.

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INTRODUCTION

Humanity has lived with radiation continuously from the beginning of our existence. With the formation of the Earth, very long-lived (order of billion years) radioactive elements, inherently a part of nature, have created a natural and inevitably accepted natural radiation level in the environment (1).

Natural and anthropogenic radioactivity is related to the primary sources of radioactive composition encountered in marine, terrestrial, and atmospheric environments.

Natural airborne radiation is caused either by radioactive gases from various cracks in the earth (especially from radon gas) or by cosmic rays. The radioactivity in the soil is due to uranium, thorium, and other radioactive substances included in the

decomposition series of those radioisotopes. Radiation in water is due to the interaction of the radioactive sources in both the air and the soil with water (2).

Natural radionuclide concentrations are continually increasing in terrestrial and mainly marine environments. The reason for that is the rapid increase in the use of artificial fertilizers, fossil fuels, detergents, and pesticides and also the proliferation of phosphate processing plants (3). The most critical radioactive substances found in natural waters are uranium, thorium, radium, radon, strontium, potassium, carbon, and hydrogen (4). Radionuclides that cause artificial radioactivity are spread to the environment as a result of radiological activities, radioactive waste heaps, or nuclear accidents. Although those activities create a variety of different radionuclides, the ones that play an essential role in the radiation exposure of humans are the products of diffusion and activation (^{137}Cs and others). Those radionuclides may lead to internal and external radiation damage in the human body. Therefore, the radioactivity levels in environmental samples must be measured with an acceptable error so that the exposure dose can be accurately assessed (5).

Nuclear weapons trials that began in 1945 and continue to date, along with the Chernobyl (1986) and Fukushima (2011) nuclear reactor accidents, have resulted in high levels of artificial radionuclides spreading to the atmosphere.

Among radionuclides spreading to the environment since 1945 are the long-half-life radionuclides, including ^{14}C, ^{137}Cs, and ^{90}Sr, which are important for environmental and human health. Those radionuclides accumulate in the marine environment and the living body, damaging both living spaces and living things (6).

In Turkey, Aközcan (2012) (7) studied the distribution of natural radionuclide concentrations in sediment samples in Didim and Izmir Bay. In his study, the activity concentrations of ^{238}U, ^{226}Ra, ^{232}Th, and ^{40}K in sediments collected from two different locations in the Aegean Sea have been defined. The highest activity concentrations of ^{238}U and ^{232}Th were observed in sediments from the Gulf of Izmir. The highest activity concentrations of ^{40}K in sediments were measured from Didim (7). Aytaş et al. (2012)(8) evaluated thirty sediment samples from different distances along the Maritza and Tundja River. These samples were analyzed by gamma spectroscopy for ^{238}U, ^{232}Th, and ^{40}K. The measured average activity concentrations of ^{238}U and ^{232}Th are above the world average, but the average activity concentration of ^{40}K is within the world average value (8).

Natural and artificial radioactivity assessment of dam lakes sediments in the Çoruh River (Turkey) was studied by Kobya et al. (2015) (9). In the study, ^{238}U, ^{232}Th, ^{40}K, and ^{137}Cs activity concentrations were determined. The mean concentrations of ^{238}U, ^{232}Th, ^{40}K, and ^{137}Cs were found to be 15.8, 13.9, 551.5 and 18.1 Bq/kg respectively in Deriner Dam Lake (zone 1); 3.7, 12.5, 473.8 and 6.8 Bq/kg in Borçka Dam Lake (zone 2); 14.4, 30.0, 491.7 and 18.2 Bq/kg in Muratlı Dam Lake (zone 3) was determined. The fact that the activity concentration values of radionuclide are maximum in the Deriner Dam Lake, where the sediment deposition is at its maximum level (9).

This study aims to determine natural and artificial radionuclide distribution in both intense industrialization and non-industrialized areas of the West of Marmara Sea by using gamma spectrometry and bring out the risky areas for human life.

MATERIALS AND METHOD

The study area covers the area between Silivri (Istanbul) and Tekirdağ-Gelibolu (Çanakkale) in the north and between Lapseki (Çanakkale) and Bandırma (Balıkesir) in the south in the Western Marmara Sea (Figure 1).

The bathymetry of the sea from which the samples were taken ranges from 20 m to 35 m. In the Marmara Sea, the Anatolian (Southern Marmara) and Thrace (Northern Marmara) shores show different characteristics. Thrace shores, which meet the sea with large plateaus, feature gulfs (Tekirdağ and Silivri Gulfs) with wide arcs and beaches with small coves that have turned into lagoons. On the other hand, the Anatolian coast features the gulfs (Izmit, Gemlik, Bandirma, and Erdek) extending in the east-west and south-north directions and the islands (Kızıladalar, İmralı, and Marmara Islands) located in front of the rugged coasts (10). Core samples taken from 29 locations (sediment from the seafloor) were used in the study (Figure 2). The samples were taken between 15.10.2016 and 20.10.2016, and the coordinates and depth information of the samples are given in Table 1.

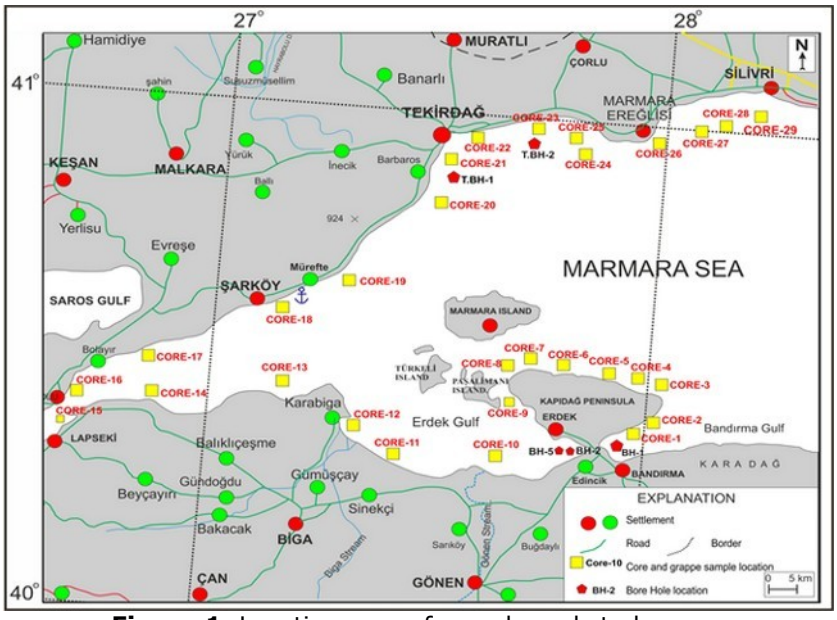

Figure 1. Location map of sample and study area.

Radionuclide analyses of the samples were made at the Çekmece Nuclear Research and Training Center. Samples were first dried at room temperature and prepared for analysis by sieving with 100-micron mechanical sieve. The samples were kept in the dry state for forty days were provided balance in radium, thorium, and other decomposition products. Concentration values of the radionuclides were analyzed using a Canberra GX5020 device.

The gamma activity peaks were: ^{238}U series, 609.3 keV; ^{232}Th series, 583 keV; ^{228}Ac, 911.2 keV; ^{226}Ra, 185.7 keV; ^{137}Cs, 661.7 keV; and ^{40}K, 1460.8 keV . In the gamma spectrometry analysis method, gamma ray-emitting radionuclides that irradiate at the energy range of 40 keV to 2000 keV are usually measured directly, without being separated from the sample matrix (such as air, water, soil, sediment, plant material, vegetable, and animal food.). Therefore, it is one of the most widely-used methods in the environmental analysis (5).

RESULTS

The range of radionuclide activity concentrations in the sediment samples, in Bq/kg, was 0.9–9.4 for ^{137}Cs; 18.9–86 for ^{232}Th; 10–50 for ^{226}Ra, and 24.4–670 for ^{40}K. Tables 2 and 3 present core sample radionuclide activities. The values were interpreted in comparison with the international average values as given in Table 4 (11).

In all locations, the analyses for ^{40}K were obtained in the core samples, the ^{40}K levels were above the world limits, whereas the ^{232}Th levels were below the limit in eleven locations and above world standard in eighteen locations. The values of ^{226}Ra were above the limit value at only one location (K-10, 50 Bq/kg). It is noteworthy that the ^{232}Th values (86–186 Bq/kg) reach the highest level in the Core-10 sample. One of the radioactive isotopes of cesium, ^{137}Cs, which is an alkali metal with similar chemical properties to potassium, reaches the highest value (9.4 Bq/kg) at the Core-20 location. That isotope has a considerably long physical half-life of 30.17 years; therefore, it is considered to be biologically hazardous for the ecosystem (12). The distributions of artificial and natural radionuclides at sea may differ from region to region, depending on various factors. The number of radionuclides deposited in marine environments vary with the physical and chemical properties of the radionuclide, dry or wet accumulation pattern, and topographic and meteorological environmental conditions (13).

Therefore, numerical results obtained from sediment samples show large variations. Figures 2, 3, 4, and 5 show the distribution of radionuclides for all sample locations in the west of the Marmara Sea. Figure 2 shows the map of ^{40}K, and radionuclides have been produced with the kriging method. Similarly, Figure 3 shows ^{226}Ra and Figure 4 shows ^{232}Th and Figure 5 shows ^{137}Cs radionuclides**.**

Table 1. Geographic coordinates of core samples.

CORE SAMPLE NUMBER	GPS-NO	DEPTH (M)	GEOGRAPHIC POSITION (WGS-84)		CORE SAMPLE NUMBER	GPS-NO	DEPTH (M)	GEOGRAPHIC POSITION (WGS-84)	
			East	North				East	North
CORE 1	116	30	058 14 59	447 46 77	**Core 14a/b**	142	29	049 02 82	447 33 47
CORE 2	117	29	058 49 85	447 66 92	**Core 15**	143	36	047 39 14	446 99 71
CORE 3	119	30	058 81 05	448 16 39	**Core 16**	144	43	047 29 08	447 24 87
CORE 3	120	35	058 62 29	448 27 80	**Core 17**	145	26	048 80 99	448 10 25
CORE 4A	121	30	058 23 93	448 35 51	**Core 18**	146	41	051 10 08	449 48 09
CORE 4B	123	32	058 22 60	448 39 37	**Core 19**	147	30	540197	4522435
CORE 4C	124	35	057 98 35	448 54 62	**Core 20**	148	33	542591	4530386
CORE 5	125	38	057 74 01	448 53 35	**Core 21**	149	32	546815	4533986
CORE 6A	126	40	056 68 40	448 63 34	**Core 22**	150	30	560139	4536953
CORE 7	127	48	056 21 01	448 68 10	**Core 23**	151	32	569767	4535792
CORE 8	128	39.5	055 81 97	448 48 62	**Core 24**	152	33	580368	4533824
CORE 9A	129	28	055 82 82	448 01 29	**Core 25**	153	33	588549	4541739
CORE 9B	137	32	055 83 61	447 97 91	**Core 26**	154	32	596782	4544493
CORE 10	138	19	055 62 81	446 36 87	**Core 27**	155	30	603222	4545684
CORE 11A/B	139	26	054 24 36	446 46 34	**Core 28**	156	31	540197	4522435
CORE 12	140	18	052 87 96	447 06 48	**Core 29**	157	35	542591	4530386
CORE 13	141	46	051 44 52	447 87 84					

Table 2. Results of gamma spectrometric analyses of core samples.

Sample	Radionuclide	Activity ± Bq/kg	Sample	Radionuclide	Activity ± Bq/kg
CORE-1	^{40}K	490	**CORE-10**	^{137}Cs	3.5
	^{137}Cs	2.5		^{226}Ra	50
	^{226}Ra	30		^{232}Th	86
	^{232}Th	42		^{155}Eu	3.2
CORE-2	^{137}Cs	1.9	**CORE-11**	^{40}K	670
	^{226}Ra	19.8		^{232}Th	57
	^{232}Th	27.1		^{226}Ra	34
CORE-3	^{226}Ra	25		^{137}Cs	3.8
	^{232}Th	28		^{54}Mn	1.1
CORE-4	^{40}K	24.4	**CORE-12**	^{232}Th	49
	^{137}Cs	1.4		^{226}Ra	28.2
	^{232}Th	21.6		^{137}Cs	4.9
	^{54}Mn	0.71		^{232}Th	45
CORE-5	^{40}K	470	**CORE-13**	^{226}Ra	25.7
	^{137}Cs	1.8		^{137}Cs	2.2
	^{226}Ra	21.5		$^{95+}$Zr	0.27
	^{232}Th	30		^{40}K	470
CORE-6	^{40}K	450	**CORE-14**	^{232}Th	36
	^{137}Cs	1.3		^{226}Ra	25.2
	^{226}Ra	27		^{137}Cs	0.90
	^{232}Th	26		^{137}Cs	3.2
CORE-7	^{137}Cs	1.1	**CORE-15**	^{226}Ra	19.5
	^{226}Ra	24		^{232}Th	36
	^{232}Th	39		^{40}K	600
CORE-8	$^{95+}$Zr	0.2	**CORE-16**	^{137}Cs	4.7
	^{137}Cs	1.1		^{226}Ra	24.9
	^{226}Ra	18.6		^{232}Th	37
	^{232}Th	30		^{137}Cs	1.6
CORE-9	^{40}K	470	**CORE-17**	^{226}Ra	18.9
	^{137}Cs	0.9		^{232}Th	26
	^{226}Ra	19			
	^{232}Th	33			

Table 3. Results of gamma spectrometric analyses of core samples (continuation of Table 2)

Sample	Radionuclide	Activity ± Bq/kg	Sample	Radionuclide	Activity ± Bq/kg
CORE-18	^{137}Cs	4.8	**CORE-24**	^{226}Ra	10
	^{226}Ra	29		^{232}Th	21
	^{232}Th	36		^{40}K	550
CORE-19	^{40}K	650	**CORE-25**	^{137}Cs	1
	^{137}Cs	2.3		^{226}Ra	22
	^{226}Ra	27.9		^{232}Th	38
	^{232}Th	39		$^{95+}$Zr	0.19
CORE-20	^{40}K	630	**CORE-26**	^{40}K	480
	^{137}Cs	9.4		^{232}Th	24
	^{226}Ra	20.5		^{226}Ra	19.2
	^{232}Th	35		^{137}Cs	0.8
CORE-21	^{137}Cs	3.7	**CORE-27**	^{137}Cs	1.3
	^{226}Ra	20.6		^{226}Ra	16.1
	^{232}Th	30		^{232}Th	21.7
CORE-22	^{137}Cs	5.1	**CORE-28**	^{40}K	580
	^{226}Ra	27		^{137}Cs	1
	^{232}Th	20.7		^{226}Ra	18.1
CORE-23	^{137}Cs	1.7		^{232}Th	24.8
	226Ra	13.7	**CORE-29**	^{226}Ra	20.9
	^{232}Th	18.9		^{232}Th	31

Table 4. International average values of radionuclides (11)

Radionuclides	***International Average Limit Values***
^{232}Th	30 Bq/kg
^{226}Ra	35 Bq/kg
^{40}K	400 Bq/kg

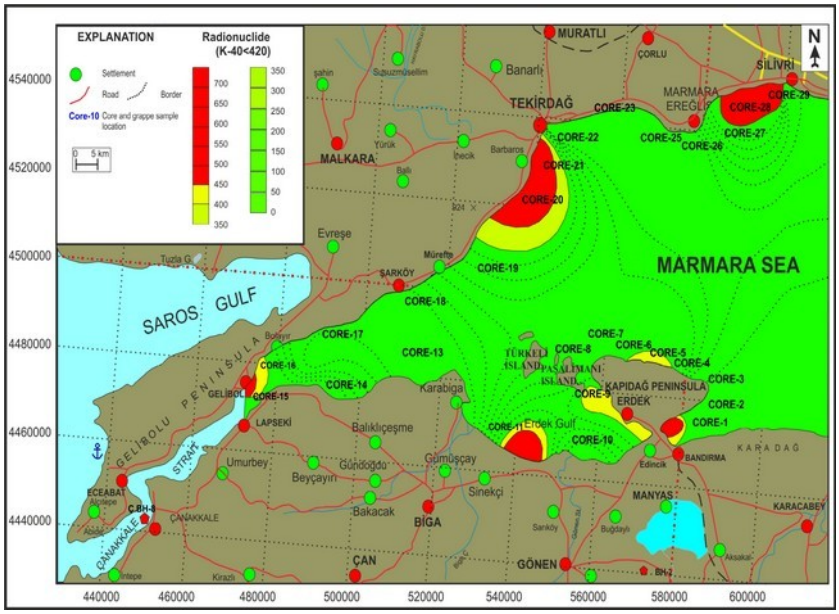

Figure 2. Distribution of the activity concentrations of ^{40}K (Bq/kg)

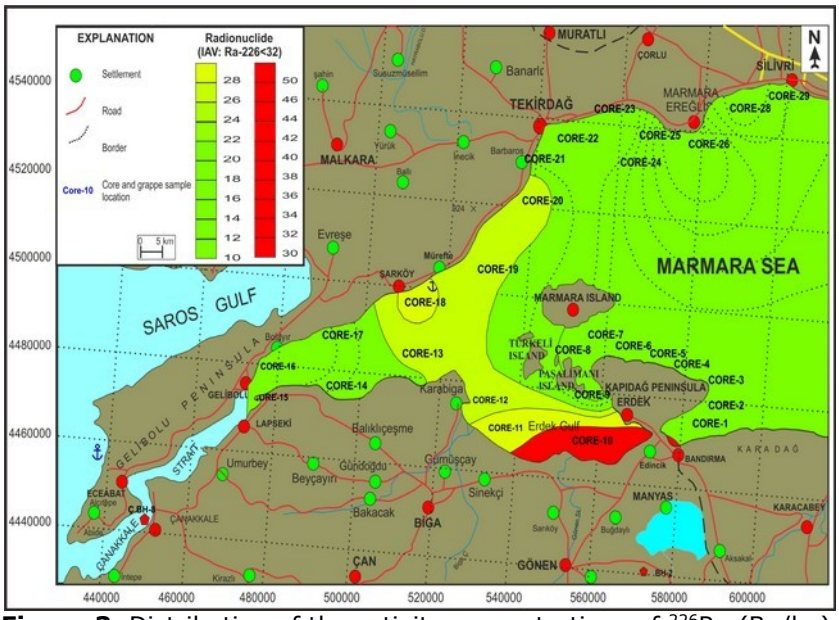

Figure 3. Distribution of the activity concentrations of ^{226}Ra (Bq/kg).

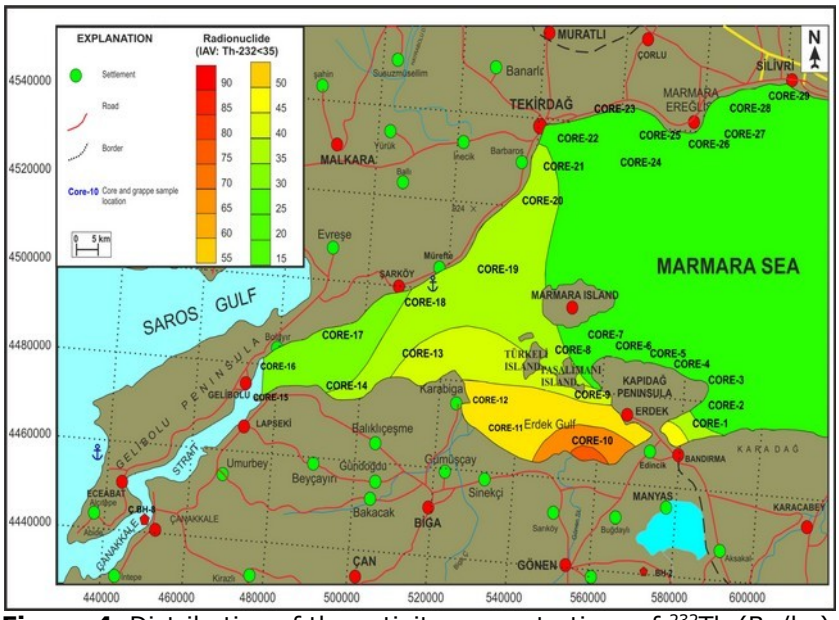

Figure 4. Distribution of the activity concentrations of ^{232}Th (Bq/kg).

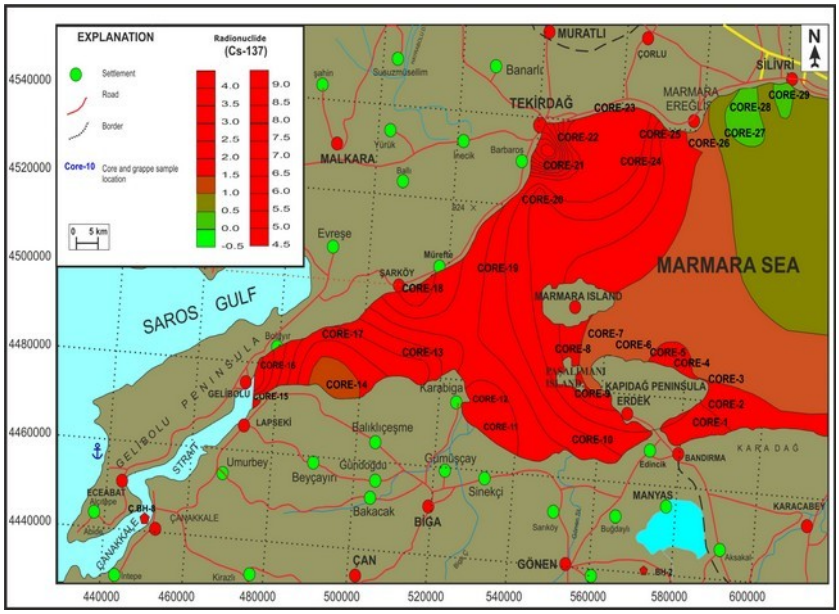

Figure 5. Distribution of the activity concentrations of ^{137}Cs (Bq/kg).

DISCUSSION AND CONCLUSION

In this study, core samples were collected from 29 different locations in the Marmara Sea for radioactivity analyses for ^{40}K, ^{232}Th, ^{137}Cs, ^{226}Ra, ^{54}Mn, and $^{95+}$Zr on the western coast of the Sea of Marmara. The Gamma Spectrometric Analysis method was used to determine the radioactivity properties of the sediments. The radionuclide concentration activity values in the sediment samples obtained from the locations, in Bq/kg, were ^{137}Cs, 0.9–9.4; ^{232}Th, 18.9–86; ^{226}Ra, 10–50; ^{40}K, 24.4–670; ^{54}Mn, 0.71–0.9; and ^{95}Zr, 0.18–0.19. These values were compared with data from the Turkish Atomic Energy Agency (TAEK) and the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), and environmental analysis was also carried out. As a result of the analyses ^{40}K values were found to be zero in Core-2, Core-3, Core-4, Core-7, Core-8, Core-10, Core-12, Core-13, Core-15, Core-17, Core-18, Core-21, Core-23, Core-24, Core-27, and Core-29. The ^{137}Cs value was zero in Core-3 and Core-24, whereas it was determined to be 9.4 (Bq/kg) in Core-20. In the Core-11 sample taken from the Sea of Marmara (between Gönen Creek and Karabiga), the ^{40}K value was higher than those of other locations and higher than the average world value. On the contrary, ^{226}Ra values measured in all the core samples were below the limit values.

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