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Original Article

Sediment Radioactivity Levels of Deep-Water Fishery Grounds in Antalya Bay

Süleyman Fatih Özmen[1], Olgaç Güven[2]

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ABSTRACT

To evaluate the radiological load of the fisheries ground sediments of deep-water areas in the Antalya Bay, ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs activity concentration levels were measured with the Gamma Spectroscopy technique using a HighResolution Germanium Detector (HPGe). Sediment samples were collected from the seabed surface of five different depth ranges (between 400 – 800 m). Detected mean radionuclide activities of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs were 16.53±2.41, 17.9±2.54, 371.44±18.44 and 3.91±1.27 Bq kg^{-1}, respectively. The effect of the Chernobyl Nuclear Power Plant disaster in deep water sediments of Antalya Bay was observed. However, the detected radionuclide concentrations are at acceptable levels according to the International Atomic Energy Agency (IAEA).

Keywords: Marine sediment, HPGe, Natural radioactivity, ^{137}Cs

ORCID IDs of the author:
S.F.Ö. 0000-0003-2168-124X;
O.G. 0000-0002-0920-673X

[1]Akdeniz University, Vocational School of Technical Sciences, Antalya, Turkey

[2]Akdeniz University, Faculty of Fisheries, Antalya, Turkey

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Correspondence:
Süleyman Fatih Özmen
E-mail:
fatihozmen@akdeniz.edu.tr

INTRODUCTION

Antalya, with a 2.4 million population, is the fifth most populated city in Turkey. Increasing population size also increases urbanization and industrialization pressures. Moreover, as a tourism hub, the area hosts an increased population density through the tourism season (KTB, 2019). In addition to regional pollutant factors, additional pressure arising from current systems and atmospheric flows were reported for the coastal areas of the region (Özhan et al., 2016). Domestic, industrial, and agricultural wastes and river discharges were known to be the main source (80-90%) of the marine pollution in the Mediterranean Sea coastline of Turkey (Yemenicioğlu, 2016). Moreover, offshore activities were considered to be the secondary sources of pollution (Yemenicioğlu, 2016).

Following the Chernobyl Nuclear Power Plant (Chernobyl NPP) disaster on 25 April 1986, a major radioactive (^{137}Cs) fallout was released into the environment. ^{137}Cs, as a common fission product, is one of the most problematic radioisotopes due to its high-water solubility, meaning it easily spreads in the environment. Radionuclides adversely affect biota in the impact area (Yilmaz and Özmen 2019; Özmen and Yilmaz 2020). As a response to the Chernobyl NPP disaster and increasing radioactivity levels in the Marmara region, a radiation monitoring program was launched by the Turkey Atomic Energy Agency (TAEK, 2004). Within the framework of this program, the detected ^{137}Cs activity was 15 Bq kg^{-1} from 0 – 5 cm layer of soil in the Antalya region in 1995.

Deep sea grounds of Antalya are important areas for commercial deep-water fishing activities. Several different fish (Deval et al., 2018) and invertebrate species (Deval and Kapiris, 2016; Deval et al., 2017) are caught from these fishing grounds and marketed in the local fisheries markets. Despite several studies having been carried out in the region to assess the state of the pollution in the marine environment (Türkmen et al., 2014; Özhan, 2015; Yilmaz, 2020), to the best of our knowledge, no literature information is available on the radionuclide activity levels of deep-sea sediments. The present study aims *i)*

to evaluate the activity levels of natural (^{226}Ra, ^{232}Th, ^{40}K) and anthropogenic (^{137}Cs) radionuclide activities in the deep sea fishing grounds of Antalya Bay and *ii)* to create a reference database of radionuclide background activity for the region.

MATERIALS AND METHODS

Sampling and sample preparation for radionuclide activity detection

Surface sediment samples were collected to investigate the availability of natural (^{226}Ra, ^{232}Th, ^{40}K) and artificial (^{137}Cs) radionuclides at deep sea fishing areas (400 - 800 m) in Antalya. Sediments accumulated in the trawl net during the deep-sea trawl hauls were transferred to the laboratory for the subsequent radionuclide analysis.

Prior to spectrometric measurement, samples were dried (72 hours) at room temperature and homogenized by grinding. The samples were weighed and filled into cylindrical containers (r=3cm, h= 6 cm) by passing through a 2 mm sieve. Samples were stored for 30 days in an airtight manner in order to stabilize the Compton region and establish a radioactive equilibrium between ^{226}Ra and ^{222}Rn (Yaprak and Aslani, 2010).

Radionuclide analysis

Following the formation of radioactive equilibrium, the gamma spectroscopic measurements of the samples were performed by using an electrically cooled high purity Germanium detector (HPGe) of p-type coaxial, with a relative efficiency of 40% and 768 eV Full Width Half Maximum (FWHM) values at 122 keV for ^{60}Co and 1.85 keV FWHM at 1332 keV for ^{60}Co. The energy calibration of the gamma spectrometer system was carried out with point sources, and IAEA RGU-1, RGTh-1, and RGK-1 radioactive standards of the same geometry as the samples were used for activity measurement. A detailed description of the measurement system has been given by Özmen et al. (2013, 2014).

All samples were placed into the detector chamber and counted for 86400s. The ^{226}Ra activity concentrations of the samples were calculated by the 352 keV (^{214}Pb) and 609 keV (^{214}Bi) energy peaks released from the ^{238}U decay series. ^{232}Th activity concentrations were calculated by the 911 keV (^{228}Ac), 583 and 2615 keV (^{208}Tl) energy peaks. Both ^{40}K and ^{137}Cs activity concentrations were evaluated by the 1461 keV and 662 keV energy peaks, respectively. Background measurements were also performed with an empty sample container before and after measurements. To calculate the radionuclide activity concentrations, the following equation has been used;

$$A = \frac{N/t}{\varepsilon \, . \, I_{\gamma} . m}$$

A: activity of radionuclide (Bq kg^{-1}), *N*: net count of energy in total (background removed), ^{t}: live time (second), ε: efficiency of HPGe detector, $I\gamma$: abundance of gamma ray and *m*; mass of sample in kg.

RESULTS AND DISCUSSION

The detected radioactivity levels of deep-sea sediment samples were ranged from 10.65 to 23.76 Bq kg^{-1} for ^{226}Ra, 11.63 to 24.15 Bq kg^{-1} for ^{232}Th, from 316.35 to 414.83 Bq kg^{-1} for ^{40}K, and from 1.48 to 8.58 Bq kg^{-1} for ^{137}Cs. Activity concentrations of radioisotopes (^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs) in sediments samples from different depths were given in Table 1.

Table 1. Radioactivity levels of the deep-sea sediments (Bq kg^{-1} dry weight).

Depth (m)	^{226}Ra	^{232}Th	^{40}K	^{137}Cs
400	10.65	11.63	414.83	8.58
500	13.88	12.43	372.51	1.48
600	13.95	18.92	346.54	4.56
700	20.43	24.15	406.99	2.32
800	23.76	22.35	316.35	2.59

The correlations matrix of radioisotopes and depth were given in Table 2. A significant positive correlation between depth and concentrations of ^{226}Ra and ^{232}Th radioisotopes was observed ($p<0.05$). However, no clear correlation was observed for ^{40}K. For ^{137}C, with the exception of 500 m samples, a negative decreasing trend was observed with increasing depth. The correlation analysis between radioisotopes also revealed a positive strong relationship between ^{226}Ra and ^{232}Th isotopes.

Table 2. Correlations matrix of radioisotopes (^{226}Ra, ^{232}Th, ^{40}K, ^{137}Cs) and depth.

	^{226}Ra	^{232}Th	^{40}K	^{137}Cs	Depth
^{226}Ra	1				
^{232}Th	0.900*	1			
^{40}K	-0.700	-0.400	1		
^{137}Cs	-0.300	-0.400	0.200	1	
Depth	1.000**	0.900*	-0.700	-0.300	1

(*) significance level 0.05, (**) significance level 0.01

The detected activity levels of the naturally occurring radionuclide (^{226}Ra, ^{232}Th, ^{40}K) were consistent with the activity levels reported from different regions of Turkey's surrounding seas. Furthermore, the levels of these radionuclide were below the worldwide mean activity levels (^{226}Ra: 35 Bq kg^{-1}, ^{232}Th: 30 Bq kg^{-1}, ^{40}K: 400 Bq kg^{-1}). The regional activity levels of naturally occurring radionuclide were directly linked with the geochemical structure of the region and anthropogenic activities such as mining, oil, and gas exploration. Our results indicated that (for naturally occurring radionuclides) the study area could be classified as a normal area in a radiological point of view.

The main pathways of the naturally occurring radionuclide entrance into the marine environment were river transport, rain water, fallout, etc. Moreover, the activity concentration of these radionuclides was reported to be dependent on physicochemical parameters such as organic matter content and pH levels of sediments (Tripathi et al., 2013; Özmen, 2020). A slight decreasing trend of ^{226}Ra and ^{232}Th activity levels was reported with the increasing distance from shoreline and depth (coastal zone, max.

Table 3. Literature information on radioactivity of the deep-sea marine sediments (Bq kg^{-1} dry weight).

Region	Study Area		^{226}Ra	^{232}Th	^{40}K	^{137}Cs	Sampling Year	Literature
Mediterranean Sea	Antalya Bay	Mean±SEM	16.53±2.41	17.90±2.54	371.44±18.44	3.91±1.27	2019	Present Study
		Range	10.65-23.76	11.63-24.15	316.35-414.83	1.48-8.58		
	Marmara Sea	SPS <63 µm	10.97-20.16	13.97-27.25	341.4-683.0	8.58-67.92	2008	Kılıç & Cotuk, 2011
		SPS >63 µm	7.18-19.18	6.41-18.30	281.9-662.2	1.12-26.40		
	Izmit Bay		18±6	24±8	568±16	21±2	2008-2009	Ergül et al., 2013
	Greece		9.7±5.4	7.8±3.0	132±54	3.3±2.0	*NA*	Papaefthymiou et al., 2017
			18-86	20-31	368-610	0.7-3.8	*NA*	Pappa et al., 2016
	Egypt		10.3-21.8	11.9-34.4	268-401	2.7-15.9	2002	El-Reefy et al., 2010
	Spain		12.1	15.0	188	*NA*	2006	González-Fernández et al., 2012
Arabian Peninsula Coastline	Oman Sea		11.83-22.68	11.83-22.68	222.89-535.07	0.14-2.8	2011	Zare et al., 2012
	Egypt (Red Sea)		*NA*	7.68 -22.70	160.0-356.8	1.11-7.92	2008	Dar & El-Saharty, 2012
	Kuwait		18.3-23.1	18.8-23.0	386-489	1.5-2.9	2016	Uddin & Behbehani., 2018
	Saudi Arabia		11.68±1.22	6.21±0.58	169.40±6.29	0.76±0.120	*NA*	El-Taher et al., 2018
Asia	Thailand		5-50	4-108	3-714	*BDL*	*NA*	Kritsananuwat et al., 2015
	India		34±15	75±38	782±233	*NA*	*NA*	Tripathi et al., 2013
South America	Venezuela		2.6-28.9	4.2-41.8	15-421.2	*BDL*	*NA*	Alfonso et al., 2014

BDL: Below the detection limits; *NA*: Not available; SPS: Sediment particle size

15 km from shore line) from South East India due to the weathering and denudation activity of land (Tripathi et al., 2013). However, in our case, a distinct positive correlation between these radionuclides and depth was observed. Both elements resistance to weathering effects and/or the elements contents of the crustal rock as the source of release could be the main drivers of the ^{226}Ra and ^{232}Th distribution pattern in the study area.

It is known that Uranium (U) (mother of Radium) and Th deposition on the seabed mainly originated from land sources. The amount adsorbed on particulates that transferred through river runoff to marine environment desorbs in the high salinity medium of the sea. Uranium (U) solubility was reported to be one of the Th sources in the marine environment (Valkovic, 2000). Mean U concentration in ocean water was given as 2.4 dpm L^{-1} (1.08 pCi L^{-1} or 3.25 µg L^{-1}). The produced Th by hydrolysis and adsorption processes is bound to the sinking particulate matter and ends up at the seabed. Our results revealed a significant correlation between depth and ^{232}Th - ^{226}Ra deposition. To understand the deposition dynamics of these radionuclides in the deep-water sediments, further investigation is needed.

The detected ^{137}Cs activity (mean: 3.91±1.27 Bq kg^{-1}) in the sediments of the study area indicate the effect of the Chernobyl NPP disaster in deep water sediments of Antalya Bay. Our results were relatively lower than reported activity levels from northern parts of Turkey. Both the distance of the study area from ground zero and the sampling time were possible factors that lead to the low activity levels in the sediment. Another aspect that needs to be emphasized is the annual sinking rate of ^{137}Cs in marine sediments. Results of radionuclide monitoring activities between 1986–2002 from the Finnish coastline indicated an annual 1 cm sink of ^{137}Cs (Ilus et al., 2008). Moreover, several other studies also reported up to 1 cm sedimentation rates in the Mediterranean Sea (Othman et al., 2000; Zuo et al., 1997; Petrinec et al., 2012; Evangeliou et al., 2013). Due to this phenomenon a relative de-

crease of ^{137}Cs activity in the marine surface sediment is expected through time.

While the main route of atmospheric radioactivity transfer to the sea is fall out, groundwater and rivers are the secondary contributor factor of the deposition to the sea (Zielinski, 2018). The main load of global ^{137}Cs fall out reported to be deposited in the sea was approximately 603 PBq (63.6%). On the contrary, the total fall (85 PBq ^{137}Cs) out from the Chernobyl NPP disaster was reported to be mainly deposited over land (69 PBq) (UNSCEAR, 2000). About 2% of ^{137}Cs is known to be removed by runoff from land deposition to the sea (Yamagata et al. 1963). Due to this phenomenon it is expected to detect higher ^{137}Cs activity levels in the coastal areas. Consistent with this, our results indicated that ^{137}Cs activity in marine sediments decreased with the depth. This outcome could be the result of deposition to the relatively shallow areas due to the impact of runoff in the study area. Moreover, evaluation of water column ^{137}Cs profiles, even with the impact of the Chernobyl NPP disaster, exhibits a steady decrease of activity up to 1000 m (Aarkrog, 2005).

CONCLUSION

Available literature on radionuclide activities in marine sediments mostly covers coastal area sediments from relatively shallow zones. The main focus of previous works was to understand the vertical distribution of the anthropogenic radionuclides (^{137}Cs and ^{239,240} Pu) in the sediments to detect the state of pollution in the environment. The present study represents natural and artificial radionuclide activities of deep-sea surface sediments (400 – 800 m). Our results provide the background activity level of deep-sea sediments for the selected radionuclides. The detected activity levels for both naturally occurring and anthropogenic radionuclides in the deep-sea fishery areas of Antalya Bay were below the world average activity levels. It could be concluded that the consumption of seafood caught from these areas will not pose any radiological health risks. Further studies are needed to understand the active factors on the distribution dynamics of the radionuclides in deep sea areas.

Conflict of interests: There are no conflicts of interest to declare.

Ethics committee approval: Ethics committee approval is not required.

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