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A Survey of ^{137}Cs in Sediments of the Eastern Mediterranean Marine Environment from the Pre-Chernobyl Age to the Present

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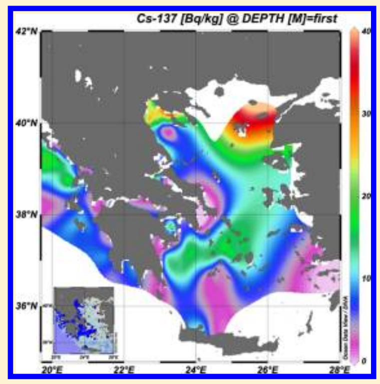
Supporting Information

ABSTRACT: A survey of ^{137}Cs in sediments from the eastern Mediterranean Sea (Aegean and Ionian Seas) during the period from 1984 to 2007 is presented. Data have been collected and analyzed in the framework of the monitoring system of Greece performed by the Environmental Radioactivity Laboratory (NCSR "Demokritos") over the past 30 years. Sediment activities reflect the impact from the Chernobyl accident one year later (1987). It is evident that sediment acts as the final receptor of ^{137}Cs, showing that fast depollution of the Mediterranean still remains a utopia. Radioactive "hot spots" were observed in the northern Aegean Sea and lower values in the southern Aegean Sea and Ionian Sea. Finally, an effort to evaluate the risk of ionizing radiation (from ^{137}Cs) to the biota inhabiting sediments was made using ERICA. The respective dose rates for two reference organisms (benthic fish and mollusks) were estimated to be far below the screening dose, at which the radiological impact on the abundance of the population begins.



INTRODUCTION

Although ^{137}Cs (half-life of 30.2 years) was first introduced into the environment by nuclear weapons testing in the 1950s and early 1960s,[1] the largest source was the Chernobyl fallout in 1986. The average density of ^{137}Cs in the Aegean Sea was estimated to be 4 kBq m^{-2}, whereas in the Ionian Sea, it was 2.5 kBq m^{-2}.[2] The total radiocesium (^{137}Cs and ^{134}Cs) input was estimated to be 2400 TBq for the Black Sea,[3−6] 820 TBq for the Aegean Sea, and 600 TBq for the Ionian Sea (60 TBq corresponding to the Greek coasts).[7−9] In addition, the northern Aegean Sea is the area where the Black Sea interacts with Mediterranean[2,10,11] through the mouth of the Dardanelles, which acts as an interregional source of ^{137}Cs. The residence times of ^{137}Cs in the water column are short enough compared to its decay half-life[12] to accumulate in sediments.

This paper comprises a baseline study for the present radiological status of the eastern Mediterranean as almost one half-life has passed from the accident of Chernobyl. Data from the period before and just after the accident (1984−1987) are reported, and the impact of fallout on surface sediments is evaluated. Additional measurements from 1999 to 2007 are presented for comparison and to evaluate trends over the past two decades. Additionally, we assess the fallout's impact on organisms inhabiting sediments using ERICA.[13,14] The results are evaluated on the basis of the conceptual model of the response of the organism to ionizing radiation suggested by Polikarpov.[15]

METHODOLOGY

During the period from 1984 to 2007, surface sediment samples (0−5 cm) were collected using several samplers employed by various oceanographic vessels and fishing boats. Some of the near-shore intertidal sediment samples were collected by divers, whereas sediment cores were collected using a Bowers and Connelly, Mark VI multicorer analyzer.[16] The samples (not corrected for decay, initially) were prepared for activity measurements according to the IAEA procedures.[17] The results refer to both enclosed marine regions and open sea environments (Figure 1), and they are therefore divided into four subregions [i.e., northern Aegean Sea (NAS 1−3), south Aegean Sea (SAS 4−11), central gulfs (CG 12−20), and Ionian Sea (IS 21−24) (Table S1, Supporting Information)].

The γ spectrometry measurements were performed by two high-purity germanium (HPGe) detector systems. The first one presents an efficiency of 20% (relative to a 3 in. × 3 in. NaI) and a resolution of 2.0 keV (at the photopeak of ^{60}Co), and it is connected to a 4K multichannel analyzer. The second detector consists of an HPGe detector with a relative efficiency of 90% and a resolution of 2.1 keV, connected to an 8K multichannel analyzer. Energy calibration was performed using standard sources of ^{22}Na, ^{54}Mn, ^{57}Co, ^{60}Co, ^{109}Cd, ^{133}Ba, ^{137}Cs, and ^{241}Am covering an energy range of up to 2000 keV in 4K channels (0.5 keV/channel) at 20% HPGe and 2000 keV in 8K channels (0.25 keV/ch) in the 90% HPGe system. The efficiency was calibrated using a ^{226}Ra source (240 Bq) with the same geometry as the measurement pot. The samples were measured for at least 70000 s, reducing the relative statistical error (1σ) to <10%. The quality assurance of the measurements is checked periodically through participation in proficiency tests

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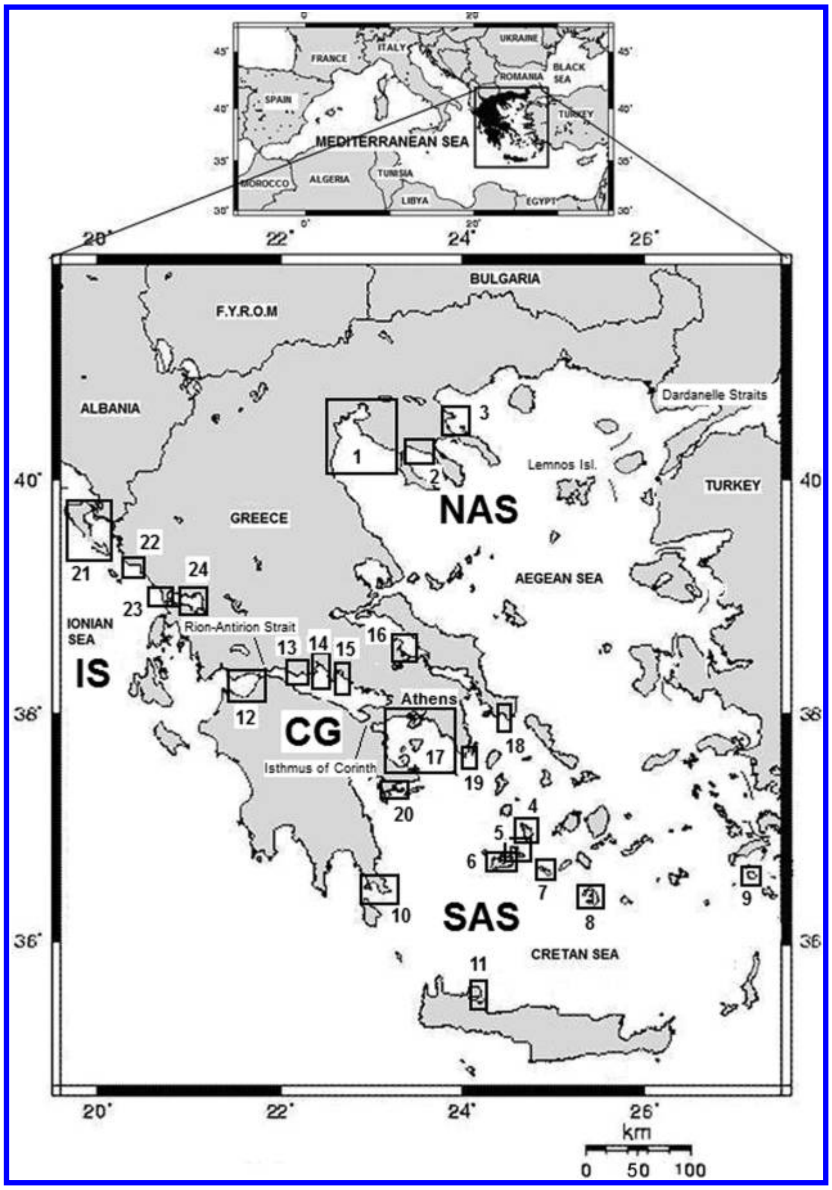

Figure 1. Sampling sites in the eastern Mediterranean where sediment samples were collected. Numbers in rectangles represent the sampling areas covering the number of stations listed in Table 1.

(e.g., in 2010 ERL participated in two tests, an IAEA-CRP1471-01 proficiency test and an interlaboratory comparison JRC-IRMM).

■ RESULTS AND DISCUSSION

Distribution in the Aegean Sea (NAS and SAS), Ionian Sea (IS), and Central Gulfs (CG) before and after Chernobyl (1984–1985 and 1986–1987). Activity concentrations of ^{137}Cs before (1984–1985) and just after the Chernobyl accident (1986–1987) are shown in Figure 2 and Table S1 of the Supporting Information. The data indicate that the Chernobyl-derived ^{137}Cs started to arrive on the surface sediments approximately one year after the accident, as atmospheric fallout reached seawater and gradually sank to the sediment. Other significant contributions were washout and soil erosion from the land, as well as river discharges that carried and transported ^{137}Cs. For example, the ^{137}Cs concentration ranged from 0.9 to 18.6 Bq kg^{-1} in SAS (Table S1 of the Supporting Information) in the period of 1986–1987; hence, small amounts were transferred to the sediments just after the accident, as ^{137}Cs remains mostly in ionic form in seawater[18] and is distributed spatially via marine processes (e.g., advection and diffusion). Taking into account the fact that (i) the sinking velocity of ^{137}Cs is low (16–82 cm year^{-1}, reported by Cai et al.[19]) and (ii) the ecological half-life of ^{137}Cs (which controls the transit times of Cs in seawater) has been estimated to be 7.8 and 8.2 years in central Greece and the northern Aegean Sea, respectively,[13,20] we found that vertical advection and diffusion are the major drivers that govern the sinking of ^{137}Cs.

An example of the Chernobyl-derived ^{137}Cs spatial distribution can be seen in Figure S1 of the Supporting Information for SAS 4–8. According to measurements and modeling data,[21] the fallout from Chernobyl reached the Greek territory on May 2, 1986, and >99% of the ^{137}Cs was deposited before December. Here, it is clearly shown that (one year after the accident) there was trivial deposition on the surface sediment. The activity concentrations of ^{137}Cs varied between

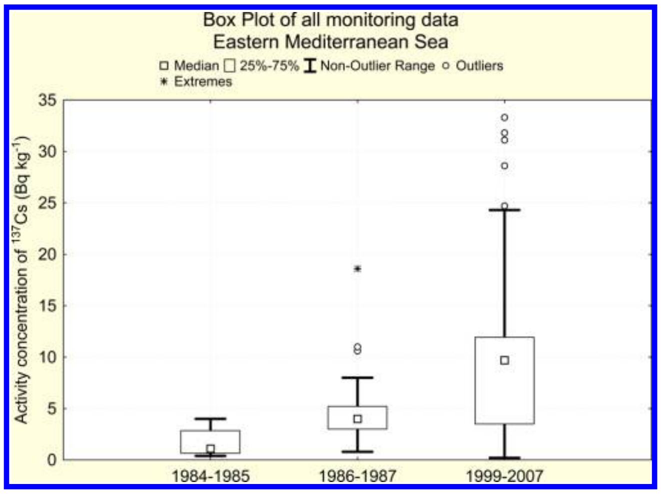

Figure 2. Box plots of the monitoring database of ^{137}Cs in sediments for the three different periods of study. Prior to the accident (1984–1985), very little ^{137}Cs originating from nuclear weapons tests had reached the sediments, whereas just after the accident (1986–1987), a rapid decrease is recorded. In the recent values (1999–2007), it is apparent that sediment acts as an accumulation medium for ^{137}Cs, even though its levels may decrease locally (e.g., Figures S3a and S4 of the Supporting Information).

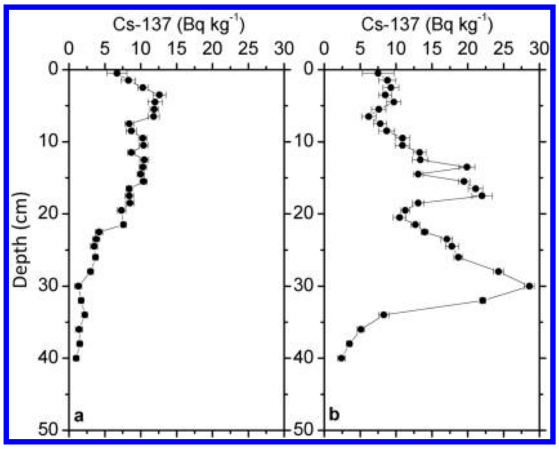

Figure 3. Vertical profiles of ^{137}Cs in the Ionian Sea (IS 24) in 2007 (IAEA RER7.003). The profiles were used to estimate rates of accumulation of ^{137}Cs using the respective dating method.[34]

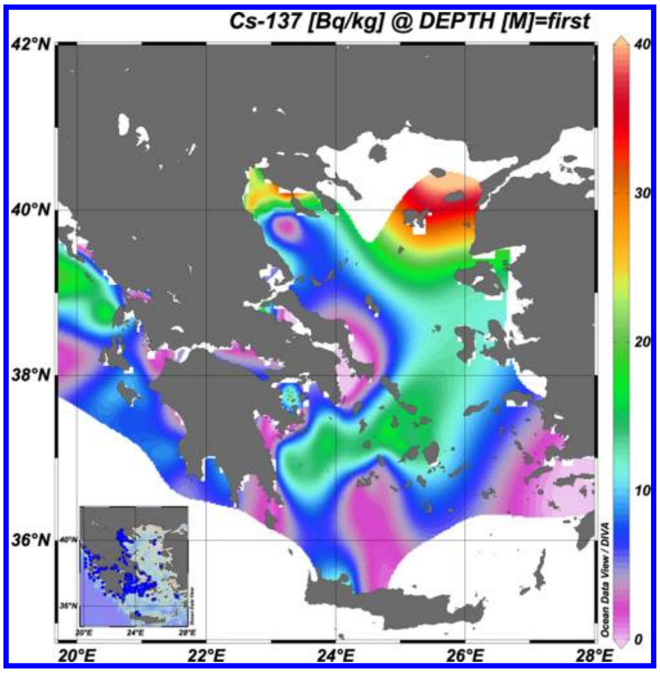

Figure 4. High-resolution map presenting the impact of ^{137}Cs in surface sediments after interpolation of all the monitoring values (since 1986) corrected for radioactive decay to May 1, 2007.

1.0 and 18.0 Bq kg^{-1}, although in >90% of the samples it was below 5.0 Bq kg^{-1}.

The weaker impact of Chernobyl on the Ionian Sea is due to the smaller direct fallout that settled on the area. However, because the Ionian Sea is a deep basin (5267 m), particle sinking is a longer process, and therefore, sediments were not influenced just after the accident (period of 1986–1987) (Table S1 of the Supporting Information).

Central gulfs of the Greek marine environment (CG 12–20) are areas adjacent to the continental margin of Greece (Figure 1). Activity concentrations of ^{137}Cs in CG 12–20 prior to the accident (1984–1985) ranged between 0.6 and 4.6 Bq kg^{-1} (average of 2.6 ± 0.6 Bq kg^{-1}), whereas they doubled one year afterward (1986–1987) to 5.2 ± 3.5 Bq kg^{-1} (range of 1.0–18.6 Bq kg^{-1}). This increase is attributed to washout and erosional processes occurring in coastal regions[12] and the transfer of terrestrial ^{137}Cs. An example can be seen in the Saronikos Gulf (Figure S2 of the Supporting Information), which is adjacent to Athens (capital of Greece). The gulf is fed by streams and rivers; thus, it is affected by washout inputs. This explains the rapid increase of approximately 35% seen from 1986 to 1987 (Table S1 and Figure S2 of the Supporting Information).

Recent Distribution (1999–2007) in the Aegean Sea (NAS and SAS), Ionian Sea (IS), and Central Gulfs (CG). In general, ^{137}Cs concentrations have increased over the past 20 years (Figure 2) because of direct (sinking of particulate ^{137}Cs) or lateral (vertical advection and diffusion) processes that transferred ^{137}Cs onto sediment. Unfortunately, no site has a consistent set of samples from all three time periods (see Table S1 of the Supporting Information), and the most robust interpretation of increasing ^{137}Cs concentrations can be made by examining the combined data set from the Eastern Mediterranean as a whole. Recent measurements in sediments showed increased levels of ^{137}Cs in the northern Aegean Sea (Figure S3 of the Supporting Information). Thermaikos (NAS 1) is one of the most important gulfs in the eastern Mediterranean.[22] The anthropogenic charge of the gulf includes urban effluents from Thessaloniki (second largest city in Greece) and the discharges from four rivers. Data show that ^{137}Cs levels tend to decrease with time, despite some occasional extreme values (e.g., 32.9 Bq kg^{-1} in 2007). ^{137}Cs was found in higher concentrations here compared to other Greek marine regions (see Table S1 of the Supporting Information) likely because of terrestrial runoff from the rivers. Kritidis and Florou[23] reported that Chernobyl fallout was heavier in northern Greece, in both continental and open sea environments. Another source of ^{137}Cs for the area is the Black Sea.[2,12] When water from the Black Sea outflows to eastern Lemnos Island, it bifurcates into two currents: one that passes through northeastern Lemnos moving toward the northern Aegean Sea and one that passes through the southern part of the island. The latter again bifurcates into northerly and southerly directions. Therefore, accumulation of Black Sea ^{137}Cs is

Table 1. Estimated Dose Rates Based on ^{137}Cs Activity Concentrations Using ERICA[a]

			dose rate ($\times 10^{-6}$ $\mu Gy\ h^{-1}$)		annual dose rate ($\mu Gy\ year^{-1}$)		dose rate ($\times 10^{-6}$ $\mu Gy\ h^{-1}$)		annual dose rate ($\mu Gy\ year^{-1}$)	
			benthic fish		benthic fish		benthic mollusks		benthic mollusks	
year	area	average ^{137}Cs ($Bq\ kg^{-1}$)	internal	external	internal	external	internal	external	internal	external
2007	NAS 1	14.6 ± 2.1	53	2913	0.47	25.52	36	2803	0.32	24.56
2007	IS 24	10.9 ± 6.0	40	2174	0.35	19.05	27	2093	0.24	18.34
2006	CG 13−15	1.1 ± 0.5	4	220	0.04	1.92	3	211	0.02	1.85
2006	SAS 9	0.5 ± 0.2	2	100	0.02	0.87	1	96	0.01	0.84
2004	CG 12	4.6 ± 1.9	17	918	0.15	8.04	11	883	0.10	7.74

[a]Two reference organisms that live, develop, and reproduce near the surface sediment were selected (benthic fish and benthic mollusks).

more likely in the northern Aegean Sea.[24] As a result, ^{137}Cs concentrations in the northern Aegean Sea are enhanced, ranging from 4.8 to 33.3 $Bq\ kg^{-1}$, whereas in the southern Aegean Sea, they are <0.8 $Bq\ kg^{-1}$ (Figure S3 and Table S1 of the Supporting Information).

^{137}Cs activity concentrations were found to vary widely between 1.1 and 22.5 $Bq\ kg^{-1}$ in the Ionian Sea (IS 24); however, these samples were collected from only one sampling area (Figure 1), which is a semiclosed ecosystem accepting effluents from two main rivers.[25] Average activity concentrations of ^{137}Cs were 14.4 ± 3.2 $Bq\ kg^{-1}$ in 1999 (Figure S4a of the Supporting Information) and 11.1 ± 2.2 $Bq\ kg^{-1}$ in 2005 (Figure S4b of the Supporting Information). The expected wide vertical variability of ^{137}Cs is attributed to the intense sedimentation of the gulf,[26,27] which is caused by the rapid formation of riverine particles carrying terrestrial ^{137}Cs. Moreover, the absence of wide spatial variability of the surface sediment during 2007 reinforces this notion. Surface ^{137}Cs has migrated to deeper layers as newly introduced ^{137}Cs particles are submerged in the seabed.

The reported sedimentation rate (0.55 ± 0.02 cm $year^{-1}$)[28] in Amvrakikos is intense enough to identify the Chernobyl and nuclear weapons testing peaks by examining depth profiles of sediments in the area. For this reason, two core samples were collected at the entrance of the gulf (a), as well as at the center (b); the profiles are shown in Figure 3. No clear variation in core a can be seen, whereas two peaks are clearly defined in core b, one at 15 cm and another at 30 cm. Considering the sedimentation rate reported above and the 21 years since the accident (2007), a depth of ∼14 cm is derived. This is reasonably close to the 15 cm peak in our findings, which confirms the Chernobyl signal. In addition, the second peak detected at a depth of 30 cm could be attributed to global fallout from atmospheric nuclear weapons tests in the 1960s. The significant differences between the two profiles are due to the fact that profile a was collected very close to the straits of Amvrakikos (IS24), where a developed front current has been observed due to the outflow of brackish waters of the gulf and the inflowing saline open seawaters. The saline water inflow transports sediments from the open sea, resulting in variation of the seafloor morphology close to the straits of the gulf.[30]

^{137}Cs concentrations in CG (Table S1 of the Supporting Information) were found to be below 7.3 $Bq\ kg^{-1}$, which is the lowest level compared to what was observed in NAS and IS. More specifically, in the Corinthiakos Gulf (CG 13−15), a semi-enclosed basin located in central Greece,[29] the concentration of ^{137}Cs decreased by more than 1 order of magnitude in comparison to that in the northern Aegean Sea (Figure S5a of the Supporting Information). On the other hand, in the Gulf of Patras (CG 12), a shallow marine embayment (120 m deep) that connects the Corinthiakos Gulf with the Ionian Sea, ^{137}Cs concentrations showed a wider variation [0.3−7.3 $Bq\ kg^{-1}$ (Figure S5b of the Supporting Information)], due to freshwater discharges from three major rivers.

To verify the impact of the accident on the eastern Mediterranean, all the measurements of ^{137}Cs since 1986 were corrected for decay to May 1, 2007, and plotted on a map to record the present radiological status. Considering that sediment is a more stable environment than seawater, it is expected that ^{137}Cs levels would not be affected by processes other than radioactive decay (additional accumulation and bioturbation are slow processes). The results are depicted in Figure 4. The highest values in the surface sediments are observed in the northern Aegean Sea as a result of the major impact of the Chernobyl fallout on northern Greece in conjunction with ^{137}Cs inflow from the Black Sea. ^{137}Cs levels in the southern Aegean Sea tend to be those of the pre-Chernobyl age, whereas higher values can be observed in the Ionian Sea. Finally, in the central gulfs of the Greek territory, a weak impact on surface sediments was found (Figure 4). These results indicate that ^{137}Cs levels are far from the pre-Chernobyl levels (especially in the northern Aegean Sea) as sediment acts as a final receptor of ^{137}Cs (Figure 2) originating from the Black Sea and/or coming from the land.

Dosimetry of the Ecosystem. A bulk of the recent measurements (1999−2007) was selected for an assessment of the radiological impact on natural populations inhabiting sediments. The selected areas provide sufficient geographical coverage of the eastern Mediterranean, whereas the corresponding time period shows the most significant levels, taking into account that a respective study has already been published for the early period after Chernobyl.[7,30] Thus, average concentrations of ^{137}Cs in sediments were applied for dose calculations using ERICA (Tier 2). ERICA is software that aims to assess the radiological risk to biota. It guides the user to record information and decisions and allow the necessary calculations to be performed to estimate risks on selected animals and plants.[14,15,31] The benthic organisms selected as references were (i) Pleuronectidae (benthic fish) and (ii) Mytilidae (benthic mollusks) that live, develop, and reproduce in and on the sediments. For these organisms, the occupancy factor was assumed to be 67% in the sediment−water interface and 33% inside the sediment for benthic fish and 80 and 20%, respectively, for the benthic mollusks. The respective dose rates, as well as the total annual doses (external and internal), are listed in Table 1.

The internal dose rates ranged from 2×10^{-6} to 53×10^{-6} $\mu Gy\ h^{-1}$ and external ones from 100×10^{-6} to 2913×10^{-6} $\mu Gy\ h^{-1}$ for benthic fish, whereas the estimated annual dose rates from internal and external exposure ranged from 0.02 to

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0.47 μGy year^{-1} and from 0.87 to 25.52 μGy year^{-1}, respectively. For the benthic mollusks, the dose rates for internal exposure ranged from 1×10^{-6} to 36×10^{-6} μGy h^{-1}, whereas the respective external dose rates ranged from 96×10^{-6} to 2803×10^{-6} μGy h^{-1}. The estimated annual dose rates ranged from 0.01 to 0.32 μGy year^{-1}and from 0.84 to 24.56 μGy year^{-1}. These values introduce a minor contribution to the total dose rate of natural radionuclides present in sediments. Florou[32] reported 0.8% of the total external dose of γ emmitters in sediments from the Greek marine environment is due to ^{137}Cs. Nevertheless, the range of the total dose rates (from 102×10^{-6} to 2966×10^{-6} μGy h^{-1} for benthic fish and from 97×10^{-6} to 2839×10^{-6} μGy h^{-1} for benthic mollusks) is far below 10 μGy h^{-1}, which is the maximal screening dose rate for organisms in terms of measurable effects (below this value, no measurable effects would occur).[32,33] Hence, we can conclude that marine populations in the eastern Mediterranean are not at risk from sedimental ^{137}Cs.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

Spatial distribution of ^{137}Cs in surface sediments of southern Aegean Sea marine environment (SAS 4–8) in 1987 (Figure S1); spatial distribution of ^{137}Cs in surface sediments of CG 17 just after Chernobyl (1986–1987), where levels in 10 of 13 samples increased within 1 year (Figure S2); spatial distribution of ^{137}Cs in surface sediments of NAS 1 and SAS 9 during the period of 1999–2007, where BDL denotes values below the detection limit of 0.1 Bq kg^{-1} (Figure S3); spatial distribution of ^{137}Cs in surface sediments of the Ionian Sea (IS 24) in 1999 and 2007 (IAEA RER.7.003) (Figure S4); spatial distribution of ^{137}Cs in surface sediments of central Greece (CG 12–15) during the period of 1999–2007 (Figure S5); variability of ^{137}Cs activity concentrations in sediments collected from the eastern Mediterranean Sea (Greece) before (1984–1985) and just after Chernobyl accident (1986–1987) together with later measurements (1999–2007) (Table S1). This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes

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