Radiation Protection Dosimetry (2012), Vol. 151, No. 1, pp. 102–111
Advance Access publication 12 December 2011
doi:10.1093/rpd/ncr449

ESTIMATION OF SEDIMENTATION RATE IN THE MIDDLE AND SOUTH ADRIATIC SEA USING ^{137}Cs

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Received July 28 2011, revised November 4 2011, accepted November 4 2011

^{137}Cs activity concentrations were studied in the sediment profiles collected at five locations in the Middle and South Adriatic. In the sediment profiles collected from the South Adriatic Pit, the deepest part of the Adriatic Sea, two ^{137}Cs peaks were identified. The peak in the deeper layer was attributed to the period of intensive atmospheric nuclear weapon tests (early 1960s), and the other to the Chernobyl nuclear accident (1986). Those peaks could be used to estimate sedimentation rates by relating them to the respective time periods. Grain-size analysis showed no changes in vertical distribution through the depth of the sediment profile, and these results indicate uniform sedimentation, as is expected in deeper marine environments. It was not possible to identify respective peaks on more shallow locations due to disturbance of the seabed either by trawlers (locations Palagruža and Jabuka) or by river sediment (location Albania). The highest sedimentation rates were found in Albania ($\sim$4 mm y^{-1}) and Jabuka (3.1 mm y^{-1}). For Palagruža, the sedimentation rate was estimated to be 1.8 mm y^{-1}, similar to the South Adriatic Pit where the sedimentation rate was estimated to be 1.8 ± 0.5 mm y^{-1}. Low sedimentation rates found for the Middle and South Adriatic Sea are consistent with previously reported results for the rest of the Mediterranean.

INTRODUCTION

In marine and coastal environments, radiotracer techniques, using either natural or anthropogenic radionuclides, have proved to be extremely useful tools to investigate various oceanographical, geochemical, biological and other processes, as well as the behaviour and fate of contaminants such as radionuclides, metals, organic and inorganic pollutants, etc.

Because of their ubiquitous nature, radioactive isotopes of caesium and strontium, particularly ^{137}Cs and ^{90}Sr, that originated in an atmospheric nuclear test conducted in the 1960s are especially important radioactive tracers in physical oceanography for water mass transport, sedimentation processes, etc. Larger quantities of radioactive isotopes of caesium have also been introduced into the environment, including the Adriatic Sea, by the Chernobyl nuclear accident[1, 2]. Its almost unlimited solubility and chemical similarity to potassium (K) means that it can be easily assimilated by terrestrial and aquatic organisms, and its bioavailability in natural systems depends on the sorption properties of the solid phases[3].

In marine sediments, ^{137}Cs can be deposited by a variety of mechanisms, including fixation on suspended matter and sedimentation, direct precipitation of colloidal forms, direct fixation by adsorption and deposition of organic waste.[4] The organic fraction present is important in terms of the binding and fixation of ^{137}Cs to sediments. Cs is also strongly adsorbed on clay particles because of their high sorption properties (large surface area and fine particle size) and abundance in natural systems[5]. It has been demonstrated that at low concentrations Cs sorbs strongly on micaceous minerals such as illite[6]. The concentration of ^{137}Cs in marine sediments is influenced by particle size, mineral composition and content of organic matter.

The use of caesium as a time marker assumes no mobility within the sediment once it is deposited. In this context, ^{137}Cs that originated from the Chernobyl nuclear accident provided a unique opportunity to trace caesium behaviour in the Adriatic coastal environment. Radioactive isotopes of caesium, particularly ^{134}Cs and ^{137}Cs, were delivered to the seabed in a relatively high amount and short time. Their depth distribution was then driven by sediment accumulation, mixing and diffusion through the water. However, due to the much shorter radiological half-life of ^{134}Cs (2.065 y), compared with that of ^{137}Cs (30.17 y), it soon decayed. Peaks of ^{137}Cs activity occurring within the Adriatic sediment were emplaced in 1963 (the year of maximum fallout from atmospheric weapon testing) and in 1986 (related to the Chernobyl event). It should be noted that ^{134}Cs, being a shielded radionuclide, is not produced in explosions of nuclear weapons in any significant amount and can, therefore, be introduced into the environment only through discharge from nuclear objects.

In this investigation, distribution of ^{137}Cs radionuclide through the middle and southern Adriatic Sea was determined in sediments collected on five locations in order to better understand some of the key processes that influence the distribution of radionuclides in the topmost part of the sediment column and to estimate sedimentation rates.

The Adriatic Sea is an epicontinental semi-enclosed sea, forming a distinct sub-region within the Mediterranean Sea region. It is a deeply indented gulf, ~800 km long and 200 km wide, situated between the Apennine and Balkan peninsulas, on longitudes between 12°15′E and 19°45′E and latitudes between 39°45′N and 45°45′N (Figure 1)[7, 8].

The southern border of the Adriatic Sea is the Strait of Otranto by a line running from the mouth of the Buttrinto River (39°44′N) in Albania to Cape Karagol in Corfu, through this island to Cape Kephali (these two capes are in latitude 39°45′N) and on to Cape Santa Maria di Leuca[9]. The surface area of the Adriatic Sea is 138 595 km^2[10] while the total length of the Adriatic coastline (mainland and islands) is 8281 km[11]. The Croatian islands area (Figure 1)[8] makes the second-largest archipelago in the Mediterranean. The Croatian part of the Adriatic Sea with 79 islands, 525 islets, and 642 rocks and rocks awash (1246 total) extends on 4398 km of insular coastline length and 6278 km of coastline length[12]. The shallowest part of the Adriatic Sea is in the Gulf of Trieste, and its deepest part is in the South Adriatic Pit (1233 m)[7].

According to sediment types and their origin, two zones of the Adriatic seabed are distinguished: external and inshore. The external zone covers the deep-sea area, from the islands towards the open sea, which is divided into a northern area covered with sand and a southern area covered with mixed sediments. The northern area occupies the entire north Adriatic as far as the line connecting the island of Kornat with Pescara. The seabed is covered with sand and in some places with a mixture of sand, mud and silt (Figure 2)[8]. According to Frignani *et al.*[13], the highest mass accumulation rates occur near the Po delta, due to the high supply of sedimentary material. There the values for sedimentation rate range between 0.5 and 1.8 g cm y^{-1}.

The following locations were investigated in this study: Jabuka and Palagruža in the middle Adriatic Sea, two locations in the South Adriatic Pit (SA PIT 1 and 2) and Albania (ST.7) in the southern part of the sea. The middle part of the Adriatic Sea has interesting geological characteristics, as it has been discovered that the islands Jabuka and Brusnik consist of rocks of magmatic origin[14]. It is well known that magmatic rocks show higher levels of background radiation, while sedimentary rocks have much lower levels of radiation[15], which also proved to be true in the case of the Adriatic Sea[16].

Jabuka is an uninhabited, solitary and separated island in the middle Adriatic Sea, situated 50 km west from the island Vis. It is a small island (surface area 0.02 km^2) with a height of 97 m[12]. It has a simple conical form and the coast is steep and hardly approachable. The island is composed of magmatic rocks, dark in colour, which could be characterised as quartz-diabase. Mineral composition is dominated by plagioclase and pyroxene, biotite, quartz, chlorite and apatite.

The islands of Palagruža constitute an archipelago on the open sea, which is composed of 10 islands of various sizes. It is situated 125 km south from the city of Split. The main island, Vela Palagruža, has a height of 92 m and a surface area of 0.28 km^2[12]. It is dominantly composed of dolomite rocks.

The South Adriatic Pit is the deepest part in the Adriatic Sea, away from the coast with no influence of material supplied by the rivers.

From a geological standpoint, the last station on which the sediments were taken, Albania, belongs to the Dinarides, related to the Alpine/Mediterranean orogenesis where the coast is composed of dolomite rocks.

Radioecological monitoring in the Adriatic Sea water, especially on the Eastern coast, started in the early 1960s and still takes a significant part in an extended and ongoing monitoring programme of radioactive contamination of the human environment in Croatia. The results of this monitoring are well documented[7, 17–20]. The long-term data on ^{90}Sr activity concentrations in the Adriatic Sea water, which are efficient intrinsic tracers of seawater movement, allowed estimation of the Adriatic Sea water turnover time that was estimated to be ~3.4 years[1, 21]. In addition, the assessment of the radiological impact on a population as well as on the environment is of particular importance since it may have a significant contribution to the collective radiation dose of a population.

The sediment samples analysed in this paper were collected during the 'International Scientific Cruise to Adriatic and Ionian Seas', 17–28 September 2007, organised under the International Atomic Energy Project (IAEA) regional TC project RER/7/003 'Marine Environmental Assessment of the Mediterranean Region'.

MATERIALS AND METHODS

Sampling and sample preparation

Before the sampling of representative samples from the seabed, some preliminary investigations with 'side scan sonar' were performed. In this investigation, a box-corer with a diameter of 10 cm was used as a tool for sediment sampling and the depth of the sampling was 30 cm (Figure 3). Samples were taken

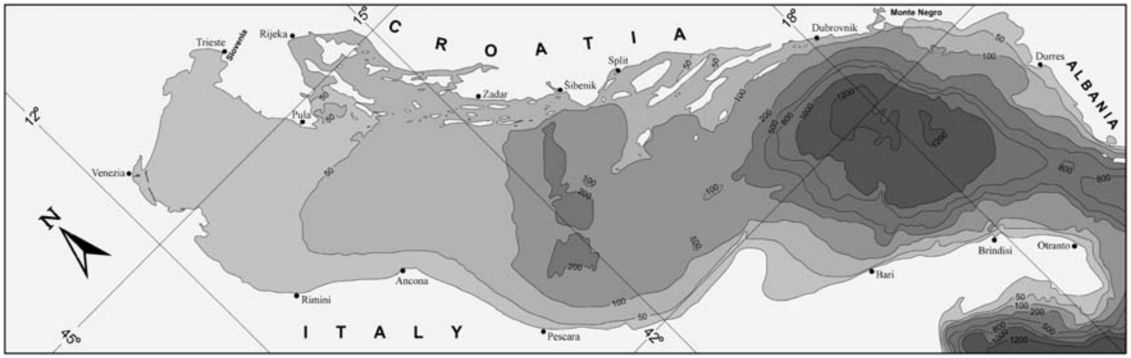

Figure 1. Bathymetric map of the Adriatic Sea[8]. The northern part is considerably shallower than the middle and southern parts.

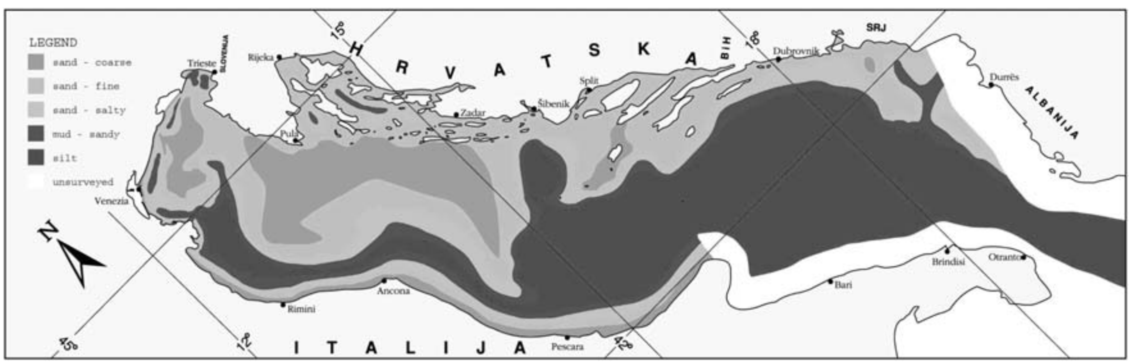

Figure 2. Map of the texture of sediments in the Adriatic Sea[8]. The northern part is sandier, while the middle and southern parts are dominantly covered by silt.

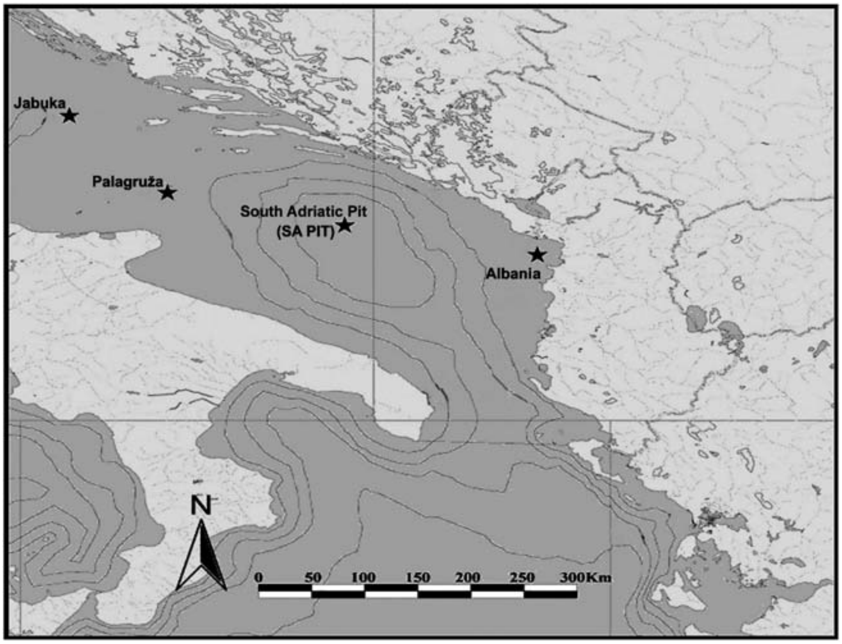

Figure 3. Locations of the sampling with the box-corer.

Table 1. Sampling locations.

Location	Date	Geographic coordinates		Depth	Sampling
SA PIT 1 (South Adriatic Pit 1)	29. 9. 2007.	N 42°20′20.77″	E 17°47′18.21″	1041.4	Box corer 1
SA PIT 2 (South Adriatic Pit 2)	29. 9. 2007.	N 42°20′44.70″	E 17°44′49.09″	1030.0	Box corer 2
Jabuka	25. 9. 2007.	N 43°03′10.25″	E 15°16′30.94″	230.8	Box corer
Palagruža	3. 10. 2007.	N 42°28′34.86″	E 16°11′27.09″	169.8	Box corer
Albania (ST.7)	2. 10. 2007.	N 41°43′24.33″	E 19°19′54.29″	59.0	Box corer

on the following locations (Table 1): South Adriatic Pit 1 and 2 (SA PIT 1 and 2), Jabuka, Palagruza and Albania (ST.7). Locations, except for Albania, were generally chosen to be in the open sea area to minimise the influence of suspended matter from the rivers and other land sediments. Samples from SA PIT 1 and SA PIT 2 were selected as the deepest in the Adriatic Sea. Samples were cut into slices of 2 cm, freeze-dried and transported to the laboratory. Before further analysis, samples were dried at a temperature of 60–80°C[(22)].

Gamma-ray spectrometry

A gamma-ray spectrometry system, based on a High-Purity Germanium Coaxial Photon Detector System ORTEC HPGe detector (FWHM 2.24 keV at 1.33 MeV ^{60}Co and relative efficiency 74.2 % at 1.33 MeV), coupled to a computerised data acquisition system was used to analyse collected samples. The detector was shielded by a 10-cm-thick lead well internally lined with 2 mm copper and 2 mm cadmium foils. Energy and efficiency calibration of the gamma spectrometer was carried out using calibration sources supplied by Czech Metrological Institute covering the energy range between 80 and 2500 keV[(22)]. Quality assurance and intercalibration measurements were conducted through participation in international intercalibration programmes organised by IAEA, World Health Organization (WHO) and Joint Research Center (JRC). The testing method is accredited by the Croatian Accreditation Agency[(23)] according to ISO Norm 17025.

Laser diffractometry

Grain-size distribution was analysed by Beckman-Coulter LS 13320, an instrument based on the laser

Table 2. ^{137}Cs activity concentrations in the analysed locations.

Layer in cm	Activity concentration ^{137}Cs in Bq kg^{-1}				
	SA PIT 1	SA PIT 2	Palagruža	Jabuka	Albania
0–2	8.79 ± 0.83	10.42 ± 0.72	4.05 ± 0.58	5.07 ± 0.62	11.80 ± 0.71
2–4	3.70 ± 0.51	5.53 ± 0.28	3.98 ± 0.49	4.27 ± 0.42	12.00 ± 0.65
4–6	2.97 ± 0.46	3.66 ± 0.41	3.84 ± 0.46	4.20 ± 0.41	12.25 ± 0.51
6–8	1.98 ± 0.49	3.22 ± 0.45	2.43 ± 0.44	4.23 ± 0.37	11.48 ± 0.28
8–10	1.27 ± 0.39	1.37 ± 0.21	1.84 ± 0.44	3.93 ± 0.52	9.11 ± 0.57
10–12	0.58 ± 0.39	0.91 ± 0.37	0.93 ± 0.28	3.47 ± 0.47	10.85 ± 0.29
12–14	1.37 ± 0.36	0.86 ± 0.38	0.90 ± 0.39	2.51 ± 0.46	11.81 ± 0.45
14–16	0.71 ± 0.36	0.82 ± 0.38	0.59 ± 0.38	1.37 ± 0.41	9.75 ± 0.56
16–18	1.13 ± 0.37	0.54 ± 0.38	0.54 ± 0.36	1.07 ± 0.22	5.34 ± 0.48
18–20	0.70 ± 0.38	0.61 ± 0.39	0.44 ± 0.37	1.14 ± 0.40	2.01 ± 0.42
20–22	0.80 ± 0.39	1.16 ± 0.36	0.62 ± 0.37	0.58 ± 0.39	2.03 ± 0.39
22–24	0.49 ± 0.38	0.53 ± 0.37	0.56 ± 0.36	0.52 ± 0.39	0.71 ± 0.35
24–26	0.48 ± 0.37	0.80 ± 0.37	0.72 ± 0.41	0.40 ± 0.38	0.83 ± 0.38
26–28	0.40 ± 0.36	0.67 ± 0.40	0.83 ± 0.38	0.69 ± 0.40	1.17 ± 0.36
28–30	0.35 ± 0.37	0.63 ± 0.41	0.50 ± 0.38	0.22 ± 0.07	0.61 ± 0.38

diffractometer method. Samples were immersed in de-ionised water, left overnight and dispersed in an ultrasound bath for 3 min. The instrument measures particle size in the range 0.4–2000 μm. This range is achieved by combining the results from laser light dispersion with the results from polarised intensity of differential scattering. Measurements were performed in a module for measurement in de-ionised water (*Aqueous Liquid Module*). Grain-size composition was determined on the sea-bottom sediment samples from Jabuka, Palagruža, SA PIT 1, SA PIT 2 and Albania (ST.7). Samples from the upper part, middle part and the bottom of the profiles were taken for analysis. Sediments were then classified according to the internationally accepted classification of sediments[24].

RESULTS AND DISCUSSION

Grain-size analysis

The results of grain-size analysis of sediments from all the five locations showed that samples from SA PIT 1 and SA PIT 2 contain 1–2 % of sand, 58–62 % of silt and 36–40 % of clay. Similar composition was also identified in the sample from location Jabuka, which is much shallower (231 m) compared with the previous two locations. Samples from Palagruža location at a depth of 170 m had a slightly lower content of silt (53–59 %) and clay (30–34 %), but a higher content of sand (almost 16 %). Sample ST.7 (Albania) taken from a depth of 59 m showed a composition of <1 % of sand, around 72 % of silt and around 27 % of clay, which is consistent with the fact that the Bojana River (Buna in Albanian) and the Drim River deliver large contents of sediment material from the mainland. Despite being short, the river has quite a large watershed, covering 5187 km^2, because the whole drainage area of Lake Scutari, the largest lake in south-eastern Europe, is also part of it. Also, due to the waters from the Great Drin, the Bojana/Buna ranks second place among all tributaries to the Adriatic, measured by the annual discharge, after the Po in Italy with 352 m^3 s^{-1} [25].

Generally, from the grain-size analysis, as it did not show significant differences in their distribution through the depth profile, it can be concluded that on all locations, except for Albania (which is shallow and influenced by the river), sedimentation is quite uniform, as should be expected in deeper marine environments.

Gamma-spectroscopic analysis

The results of the gamma-spectrocopic analysis of the samples are shown in Table 2. ^{137}Cs concentrations are similar in samples SA PIT 1 (Figure 4) and SA PIT 2 (Figure 5). In the upper part of the profiles, exponential fall of the activity concentration of ^{137}Cs from cca 10 Bq kg^{-1} to around 0.5–0.6 Bq kg^{-1} in deeper layers is obvious. In samples from Palagruža, almost all ^{137}Cs is retained in the few upper layers and after that it falls to a value of 0.6 Bq kg^{-1}. Values for location Jabuka are consistent with earlier investigations[26]. Results for activity concentrations of ^{137}Cs for the samples from Albania differ from the others. Namely, samples from Albania were taken on the location where the sea was quite shallow while the location itself was closest to the coast and highly influenced by the Bojana River and the Drim River. It could be argued that most of the ^{137}Cs is transported by the

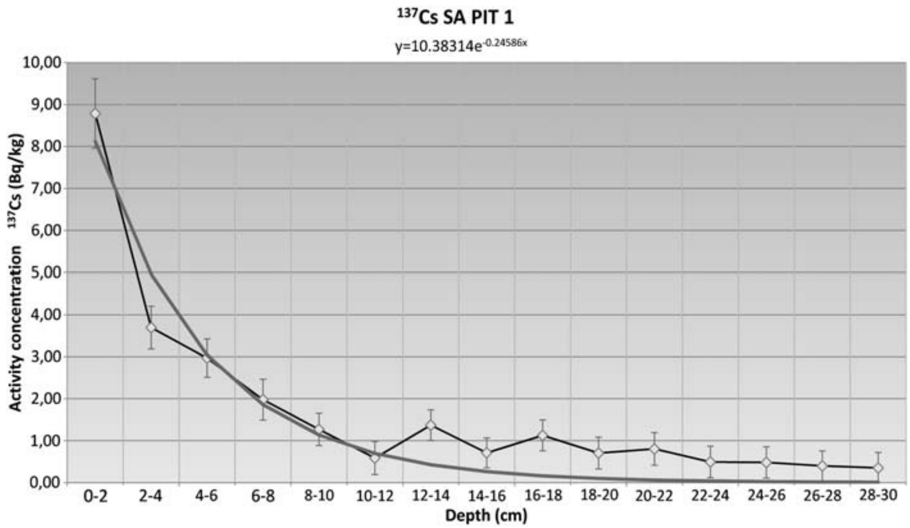

Figure 4. Activity concentration ^{137}Cs of the samples from SA PIT 1.

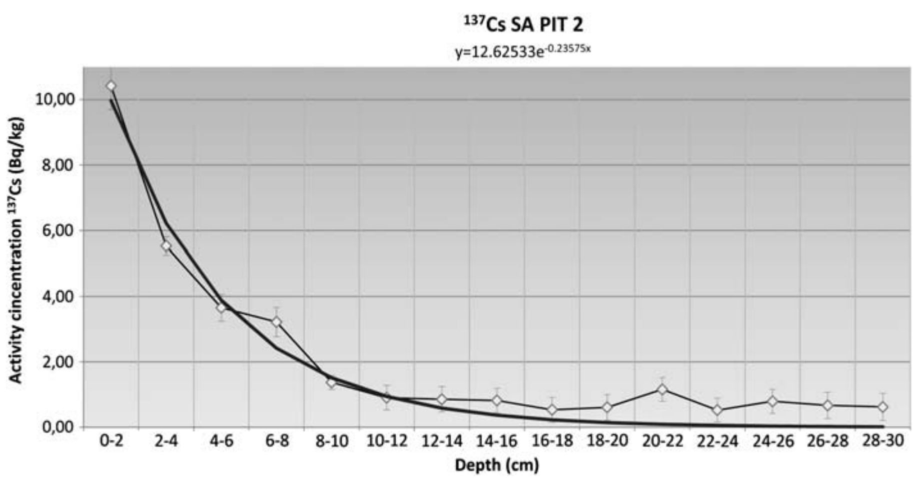

Figure 5. Activity concentration ^{137}Cs of the samples from SA PIT 2.

river runoff from the mainland and deposited on the sea bottom. This is the reason for higher values for sedimentation rates and retaining ^{137}Cs, even to 20 cm of the depth.

These results show that almost all ^{137}Cs is retained in the upper parts of the sediments. In the lower parts, the activity concentration of ^{137}Cs is pretty much constant. Barisic *et al*. (1996) obtained similar results in previous investigations. The existence of ^{137}Cs in the lower parts of the sediments can be explained by cationic exchange with ions of potassium in clay minerals in sediments[27, 28].

In order to estimate the effective depth for retention of ^{137}Cs, for samples from all locations, fitting of the results to the following exponential function was performed (Figures 4–8):

$$A_s(t) = A_s(0)\ e^{-kt} \qquad (1)$$

The physical meaning of the terms in equation (1) is as follows: $A_s(t)$ is the time-dependent activity concentration of ^{137}Cs in sediment (Bq kg^{-1}); $A_s(0)$ is the initial activity concentration of ^{137}Cs in sediment (Bq kg^{-1}); and $1/k = d_{1/2,\mathrm{eff}}$ is the effective (observed) depth for retention of ^{137}Cs in sediments (cm).

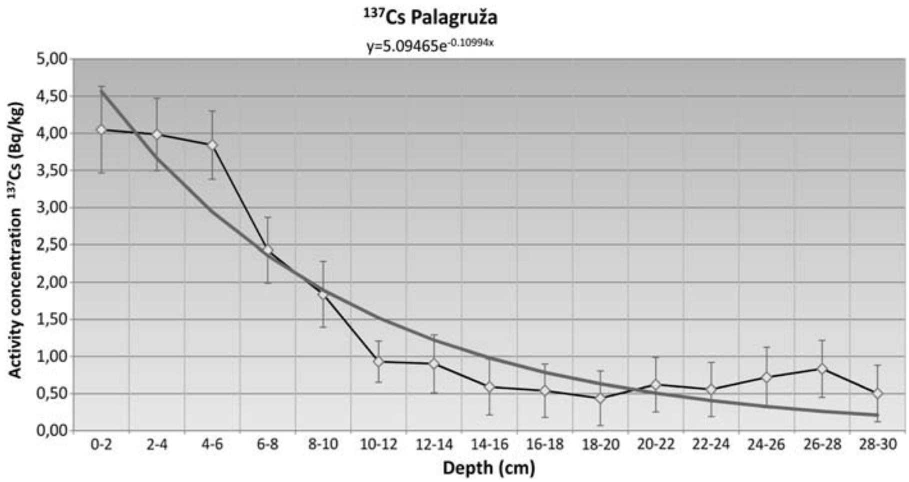

Figure 6. Activity concentration ^{137}Cs of the samples from Palagruža.

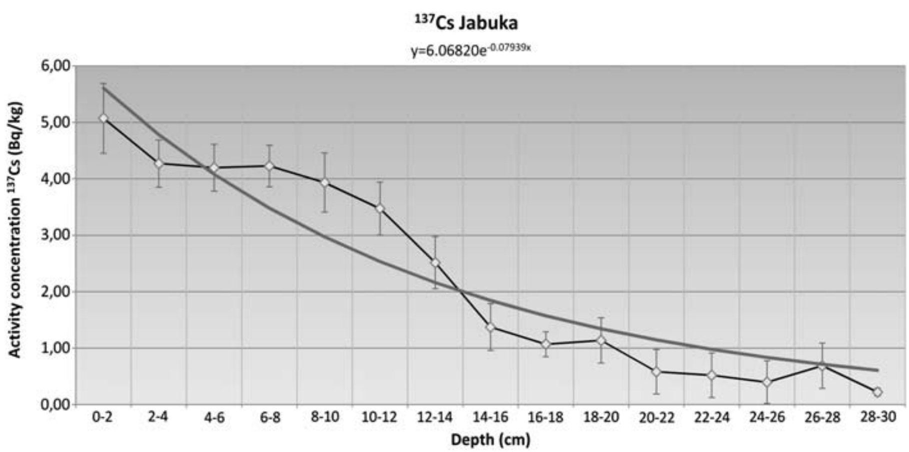

Figure 7. Activity concentration ^{137}Cs of the samples from Jabuka.

From the resulting coefficients, it is possible to estimate the mean depth for retention of ^{137}Cs. For the South Adriatic Pit (SA PIT), this value has been found to be 4 cm, for Palagruža 8.5 cm, for Jabuka 12.5 cm and for Albania 15.5 cm. It should be noted, however, that in the entire area around the islands of Jabuka and Palagruža is a major fishing site with quite intensive fishing activities. Therefore, trawlers are constantly disturbing the seabed, causing the mixing of sediments and therefore allowing ^{137}Cs to penetrate into deeper layers. Consequently, for the estimation of sedimentation rate using ^{137}Cs only locations in the South Adriatic pit are representative.

As ^{137}Cs is fission product, i.e. anthropogenic radionuclide, it is present in the environment since the first atmospheric explosions of nuclear weapons. The most intensive period of nuclear weapon tests was around the 1960s, resulting in the introduction of large quantities of ^{137}Cs in the marine environment. The second peak of ^{137}Cs in the environment was caused by the Chernobyl nuclear accident in 1986.

Relating to the time of emission of ^{137}Cs in the environment, we can use it as an indicator of sedimentation rate[(29)]. To determine sedimentation rates, it is possible to identify two peaks on the graphs, the deeper of which could be related to the period of 1962–64, when most of the nuclear probes happened, and the other one to the Chernobyl accident in 1986[(30)]. These two peaks were also identified in investigations of the fallout and air in Croatia[(31)].

In the Adriatic Sea, sedimentation rates have been mainly studied in the North Adriatic in which sedimentation is highly influenced by the Po River.

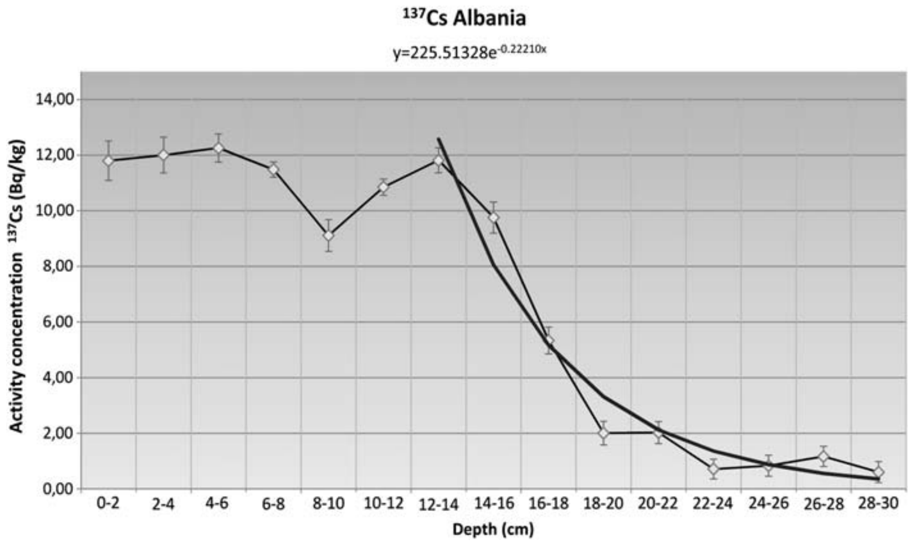

Figure 8. Activity concentration ^{137}Cs of the samples from location near the coast of Albania.

Therefore, sedimentation in that area is quite high, and sedimentation rate was estimated to be between 1.6 and 4.8 cm y^{-1} (13). As explained earlier, in profiles of sediment samples collected in Jabuka and Palagruža locations, two ^{137}Cs peaks could not be identified. However, as it is visible that ^{137}Cs is mainly retained in the upper sediment layers, in spite of constant disturbance of sediment profiles by trawlers, it can indicate a small sedimentation rate. Similarly, due to the quite strong influence of the Bojana River and the Drim River on Albania, two peaks also merged. However, assuming that the lowest value for ^{137}Cs activity concentration corresponds to the period of atmospheric nuclear weapon tests, sedimentation rates for these locations as well were estimated.

The highest sedimentation rates were found in Albania ($\sim$4 mm y^{-1}) and Jabuka (3.1 mm y^{-1}), and in Palagruža (1.8 mm y^{-1}).

In the South Adriatic Pit, where two peaks are more easily identified, sedimentation rate could be estimated to be $\sim 1.8 \pm 0.5$ mm y^{-1}, which is quite similar to the value estimated for Palagruža.

However, it should be noted that these results are just approximate due to method limitations and large uncertainties in the estimation of the exact location of ^{137}Cs activity concentration peaks in the sediment profiles.

Nevertheless, sedimentation rates estimated for Middle and South Adriatic using ^{137}Cs as a radiotracer are consistent with sedimentation rates estimated for the rest of the Mediterranean sea, i.e. 1.1–8.7 mm y^{-1} (32) using other radiotracer methods, i.e. the ^{210}Pb dating method.

CONCLUSIONS

^{137}Cs activity concentrations were studied in the sediment profiles collected on five locations in the Middle and South Adriatic.

The grain-size analysis on all investigated locations did not show significant differences in their distribution through depth profile, and it can be concluded that on all locations, except for Albania (which is highly influenced by the river), sedimentation is quite uniform.

As in most of the other environmental samples in sediment profiles, ^{137}Cs activity concentrations decreased exponentially, allowing for the estimation of its effective penetration, i.e. retention depth, which was estimated to be $\sim$4 cm in an undisturbed location of the South Adriatic Pit and 10–15 cm on other locations that are generally disturbed by the heavy fishing activities of trawlers.

Radiotracer techniques using ^{137}Cs originating from atmospheric nuclear weapon tests and the Chernobyl accident were used to estimate the sedimentation rate on five locations on the Adriatic Sea. Both peaks were visible only in sediments collected on the South Adriatic Pit, and equating elapsed time to peak depth it was possible to estimate the sedimentation rate as 1.8 ± 0.5 mm y^{-1}.

On other locations ^{137}Cs peaks merged, allowing one to estimate only the upper value for the sedimentation rate, which varied between 1.8 mm y^{-1} in Palagruža and 3.1 mm y^{-1} in Jabuka.

However, all obtained values are consistent with the sedimentation rates estimated by other techniques and previously reported in the literature.

FUNDING

This study is a part of the research projects, ‘Radioecology of the Adriatic Sea and Coastal Areas’ and ‘Environmental Radioactivity and Radiation Protection’ supported by the Croatian Ministry of Science, Education and Sports of the Republic of Croatia and IAEA TC project RER/7/003 ‘Marine Environmental Assessment of the Mediterranean Region’. The Slovenian Research Agency (contract no. P2–0075) is gratefully acknowledged.

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