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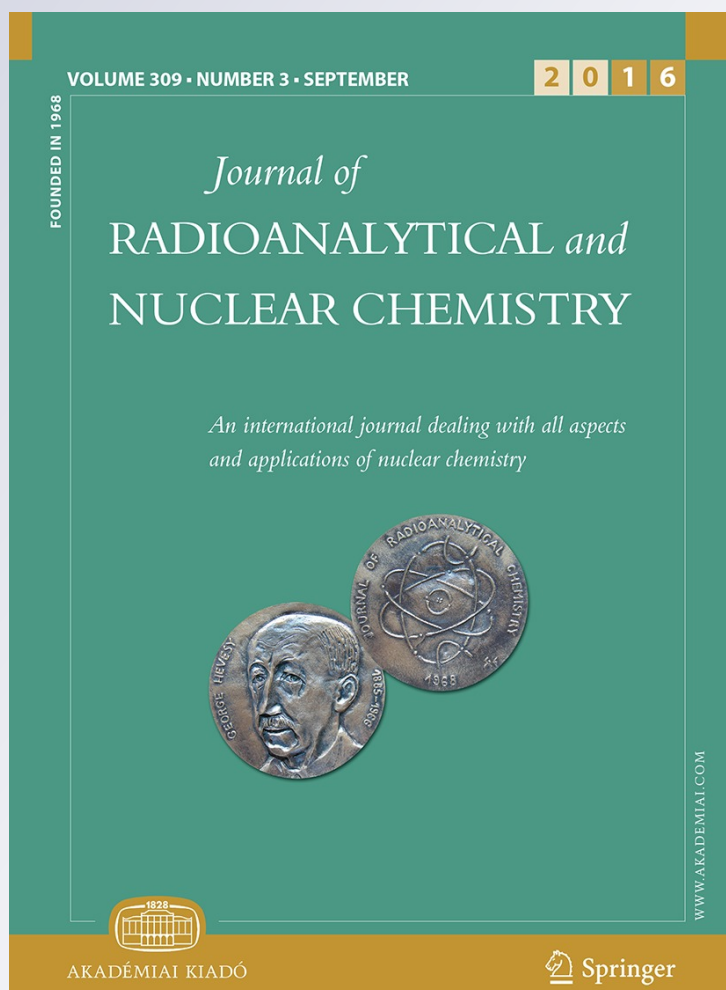
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# $^{137}\text{Cs}$ distribution in the northern Adriatic Sea

Dijana Pavičić-Hamer<sup>1</sup> · Delko Barišić<sup>2</sup> · Brigita Šimunac<sup>3</sup> · Branko Petrinec<sup>4</sup> · Marko Štrok<sup>5</sup>

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**Abstract** Anthropogenic radioactivity in the northern Adriatic Sea was assessed by analyzing  $^{137}\text{Cs}$  activity concentrations in seawater, sediment, and marine organisms in the period from 2006 to 2010. The investigated locations were the Po River delta, Lim Bay and the Rovinj coastal area. At all locations  $^{137}\text{Cs}$  activity concentrations in seawater were lower than pre-Chernobyl values ( $<4 \text{ Bq m}^{-3}$ ). The highest  $^{137}\text{Cs}$  activity concentrations in sediment were found in the Po River delta ( $8.70 \text{ Bq kg}^{-1}$ ). The data indicate that some species, such as sardine *Sardina pilchardus*, red mullet *Mullus barbatus* and the brown alga *Fucus virsoides* are better bioaccumulators of  $^{137}\text{Cs}$  than others. In addition, the ERICA Assessment Tool was used to calculate the total dose rates for reference organisms included in the software, as well as for specific sampled marine organisms.

**Keywords** Adriatic Sea ·  $^{137}\text{Cs}$  · Bioaccumulation · Bioindicators · Dose assessment

## Introduction

The northern Adriatic Sea is a unique region of the Mediterranean due to its peculiar climatic and oceanographic characteristics [1]. The northernmost part of the Adriatic Sea is shallow, with depths around 40 m, and is strongly influenced by the Po River which brings high level of nutrients and pollutants into the sea and causes high ecosystem productivity in the region. The dominant sources of anthropogenic radionuclides in the Adriatic Sea stem from the fallout of nuclear weapon testing in the 1960s [2] and the Chernobyl accident of 1986 [3]. Radionuclides released from various sources into the atmosphere are then transported and deposited to the land and sea. River discharge is secondary transfer of radionuclides in the marine environment [4, 5].

$^{137}\text{Cs}$  is considered the most important radionuclide from fallout in the assessment of marine pollution by anthropogenic radionuclides. Due to its contribution to radiation doses in humans via seafood consumption [5, 6].  $^{137}\text{Cs}$  is a long-lived radionuclide with a half-life of 30 years. It is often used as a radionuclide tracer in seawater. Conservative, soluble in seawater, caesium remains in the water column for a long time and is widely dispersed in the marine environment via water mass transportation [7–9]. The mean residence time of  $^{137}\text{Cs}$  in the Eastern Mediterranean Sea is estimated to be  $15 \pm 4$  years, lower than the average reported for the world oceans (28.6 years) [10, 11].

The presence of  $^{137}\text{Cs}$  in the Adriatic Sea is provoked by several different sources of contamination and the overall water circulation regime [4]. The general circulation controls the distribution of the soluble fraction of  $^{137}\text{Cs}$ . The Adriatic Sea is a semi-enclosed sea connected with the Mediterranean Sea through the Otranto Strait, and it is

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characterised by cyclonic circulation (inflow along the eastern and outflow along the western coast). The northern Adriatic Sea can be excluded from this circulation. This usually happens during the warm part of the year when the northern part of the Adriatic Sea is under the influence of the Po River [12] which causes pronounced vertical stratification (pycnocline) [13]. On average, based on the difference in water density stratification in the water column, north-eastern Adriatic is well mixed in December and January–February and stratified during the rest of the year [14]. However, in the vicinity of the Po River delta, influx of freshwater can induce stratification of the water column even in winter months [15].

Previous studies have shown that  $^{137}\text{Cs}$  from the Po River was transported to the northern Adriatic Sea, where it accumulates in coastal marine sediment. The lowest values can be found in sandy areas, relatively high values (from 2 to 20 Bq kg $^{-1}$ ) are characteristic for fine sediments, while the prodelta areas of the river show the highest activities [16].

The biological behaviour of  $^{137}\text{Cs}$  is similar to that of potassium. After entering the organism, caesium is mainly accumulated in soft tissue in bivalves and in muscle tissue in fish [8, 9]. Significant differences in  $^{137}\text{Cs}$  uptake were found among different species living under similar environmental conditions, suggesting that uptake does not follow physical and geochemical processes [10] but is influenced by allometric parameters, mostly through physiological processes [8, 9, 17]. The removal of  $^{137}\text{Cs}$  from surface seawater can also be influenced by intense biological activity in eutrophic areas. The bioaccumulation of  $^{137}\text{Cs}$  in organisms is affected by the food chain. Plankton absorbs the radionuclide from seawater and transfers it to high trophic levels in the pelagic and benthic communities [8]. Marine fishes had different concentrations of  $^{137}\text{Cs}$  in relation to their trophic level and in accordance with its diet [18]. This suggests the possibility of using specific marine organisms for the purpose of biological monitoring. The brown alga *Fucus vesiculosus* has been used as a bioindicator for the investigation of the impact of the Chernobyl accident on the spatial and temporal distribution of radionuclides in the Baltic Sea [19]. Mussels are recognised worldwide as pollution bioindicator organisms (Mussel Watch) because they accumulate pollutants in their tissues at elevated levels in relation to pollutant biological availability in the marine environment [6, 20]. They are recognized to show an early response to radioactive contaminants added to the ecosystem [8, 21].

Historically, radiation protection from the harmful effects of radionuclides, such as  $^{137}\text{Cs}$ , has focused on the protection of human beings and it was supposed that, if humans are protected, all other biota are also sufficiently protected. However this approach has changed in recent

years, focusing more on wildlife protection since humans are only one of the actors in the complex interactions occurring in the environment [22]. This subsequently caused the need to calculate actual dose rates due to the specificity of radioactive contaminants and assess total dose rates for reference organisms that can be found in specific environments. The ERICA Assessment Tool is a versatile and widely available tool for calculating total dose rates in reference organisms. In addition, it allows the calculation of dose rates for specific organisms [23, 24].

This study aimed to accomplish a radioecological assessment of the northern Adriatic Sea through the determination of  $^{137}\text{Cs}$  activity concentrations in seawater, sediments and organisms, calculate total dose rates in organisms using the ERICA Assessment Tool, and identify possible indicator species. A comparison of radioactive contamination was made among three different parts of the marine ecosystem: the Po River delta, the Lim Bay protected area and the Rovinj coastal area in period from 2006 to 2010.

## Materials and methods

### The study area

In the northern Adriatic Sea were investigated three stations: the Po River delta (108), Lim Bay, and the coast of Rovinj (Fig. 1). Sampling stations were chosen to take into account all major parts of the marine ecosystem of the northern Adriatic Sea and possible coastal influences.

The station Po River delta is located in north-west of the Adriatic, with an average depth of 30 m. The station is directly influenced by river discharge, with a large influx of fresh water and particulate matter.

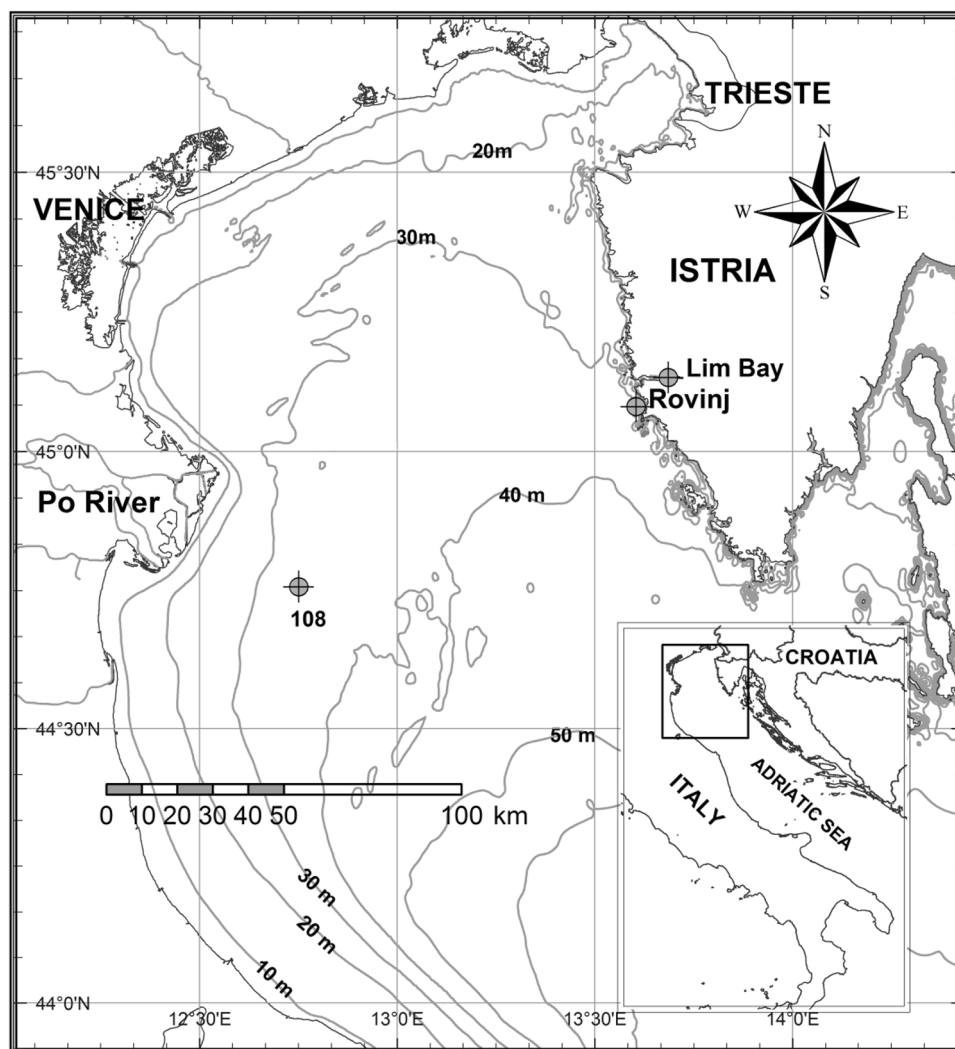
Lim Bay, located on the west coast of the Istrian peninsula, is the 11 km long estuary of the underground river Pazinčica, which plunges into the Lim channel-like bay. The sampling station is located 10 km from the mouth of the bay, with a depth of 15 m. Lim Bay is a protected landscape, biologically important as a fish spawning site, while its inner part is reserved exclusively for mariculture.

The station in the Rovinj coastal area is located on the west coast of the Istrian peninsula near city Rovinj. The station is under intense anthropogenic influence. Pressures on the marine ecosystem come from the fish processing factory, harbour, marina, and tourism especially in the summer months. The sea depth at the station is 12 m.

### Sampling and samples preparation

The salinity and seawater temperature were measured with pIONneer 65 apparatus (Radiometer Analytical S.A.) in situ at each station during the sampling. Measuring

**Fig. 1** Sampling stations the Po River delta (108), Lim Bay and the Rovinj coastal area in the northern Adriatic Sea. The stations are marked by *circle* with *cross*



simple ancillary data such as salinity and temperature can indicate environmental conditions at the sampling sites.

Samples of seawater and sediment from the Po River delta were collected seasonally from 2006 to 2010. Samples of seawater and mussels from Lim Bay and samples of seawater, fish and algae from the Rovinj coastal area were collected in April and October in the period 2006–2010.

Seawater samples were collected from the Po River delta, Lim Bay, and the Rovinj coastal area (Fig. 1). At each sampling station, 50 L of seawater were collected. Surface seawater samples were collected from a depth of 0.5 m which is presented in this study as 0 m. At the Po River delta seawater samples were collected at 0, 15, and 30 m of depth, whereas at the other two stations, only surface seawater was collected. The determination of  $^{137}\text{Cs}$  in seawater was done by a coprecipitation of Cs from a 50 L seawater sample using microcrystalline AMP and stable CsCl as the carrier. The AMP with adsorbed caesium was separated from the seawater and analysed for  $^{137}\text{Cs}$  content with gamma spectrometry.

Sediment samples were collected using a grab corer at the Po River delta and in the Rovinj coastal area; the upper 10 cm layer was taken for analysis. A grain-size analysis of sediment samples was carried out before [25]. The Po River delta sediment type is silty clay, and the Rovinj coastal sediment type is silty sand. Afterwards samples were transferred to plastic bags, sealed and frozen at  $-20\text{ }^{\circ}\text{C}$ . In the laboratory, they were dried at  $80\text{ }^{\circ}\text{C}$  and homogenised, and then, packed into counting containers. The geometry used for gamma-spectrometric measurement is standard cylindrical bow with a volume of 100 ml and average mass of measured samples was  $\sim 100\text{ g}$ .

Mussels *Mytilus galloprovincialis* were collected from the mussel farm located in Lim Bay. The brown alga *Fucus virioides* was collected in the Rovinj coastal area. The pelagic fish sardine *Sardina pilchardus*, and the benthic fishes grey mullet *Mugil cephalus* and red mullet *Mullus barbatus* were collected from local fishermen in the Rovinj coastal area.



The methodologies for biota samples preparations are described in the report RER/7/003 Monitoring Project IAEA [26]. Briefly, each sampling considered about 4–5 kg wet weight of mussels ranging in size from 4 to 6 cm, average length of mussels was 5.59 cm. The mussels were cleaned from external material and their byssus, opened using a mikro-wave oven, and then the soft parts were separated and pooled for analysis. Fish samples considered about 4–5 kg wet wt, average length for *Sardina pilchardus* was 14.5 cm, *Mugil cephalus* was 31 cm and *Mullus barbatus* was 18 cm. From fishes the muscle tissue were separated for analyses. Brown algae sample considered about 2 kg wet wt. At the laboratory, algae were cleaned with water and dried with filter paper. All of the biota samples were dried at 105 °C to reach constant weigh, but the results are reported on a wet weight basis. After draying, all samples were homogenised and transferred to calibrated plastic containers for gamma spectrometry.

## Measurements

The methodology used for radiochemical analyses and measurements of  $^{137}\text{Cs}$  in seawater samples, sediments and organisms is based on the IAEA TRS No. 118 [21, 27]. Mass activities of  $^{137}\text{Cs}$  were determined by the gamma-spectrometric method using an HPGe semiconductor detector system coupled to a Canberra 8196 channel analyzer. Counting time was 80,000 s. The obtained spectra were analyzed by Genie2 K Canberra software.  $^{137}\text{Cs}$  mass activities were calculated from the photo peak at 661.6 keV. The detector system was calibrated using gamma mixed standards supplied by Eckert & Ziegler (Analytics USA). Efficiency of the system was regularly checked during inter-comparison calculations. Precision and accuracy of the system were additionally checked by simultaneous measurement of IAEA Reference Materials since the laboratory is accredited by the Croatian Accreditation Agency for gamma-spectrometric measurements (HRN EN ISO/IEC 17025:2007). Efficiency was calculated as a function of energy and geometry at the base of experimental data. Measured uncertainty budget includes net peak area uncertainty, efficiency uncertainty and background fluctuation uncertainty. It was multiplied by a coverage factor  $k = 2$ , representing a  $2\sigma$  confidence interval with a confidence level of 95 %. Values are expressed in  $\text{Bq kg}^{-1}$  wet weight  $\pm 2\sigma$  propagated analytical uncertainty.

## Dose assessment

Total dose rates due to the presence of  $^{137}\text{Cs}$  in the marine environment of the northern Adriatic Sea were assessed

with the help of Tier 2 of the ERICA Assessment Tool. Total dose rates were calculated for all of the marine reference organisms available in the ERICA Assessment Tool. A comparison between the Po River delta sampling site, and the Rovinj coastal area was performed. For the calculation, average, maximum and minimum  $^{137}\text{Cs}$  activity concentrations in seawater and sediment at the respective sites and timescale were used. Other parameters were left unchanged. These include all of the concentration ratios for different reference organisms (occupancy factors), which indicate the habitat in which specific organisms reside (water surface, water, sediment surface, and sediment), and radiation weighing factors (10 for alpha, 1 for beta and gamma, and 3 for low beta). In addition, total  $^{137}\text{Cs}$  dose rates to the specific sampled organisms (*Fucus virsoides*, *Mugil cephalus*, *Mullus barbatus* and *Sardina pilchardus*) were calculated and compared with respective marine reference organisms from the ERICA Assessment Tool. For the purpose of comparing the dose rate for specific organism with reference organism from the ERICA Assessment Tool, the average  $^{137}\text{Cs}$  activity concentration in a specific organism was used for calculation the dose rate.

## Results and discussion

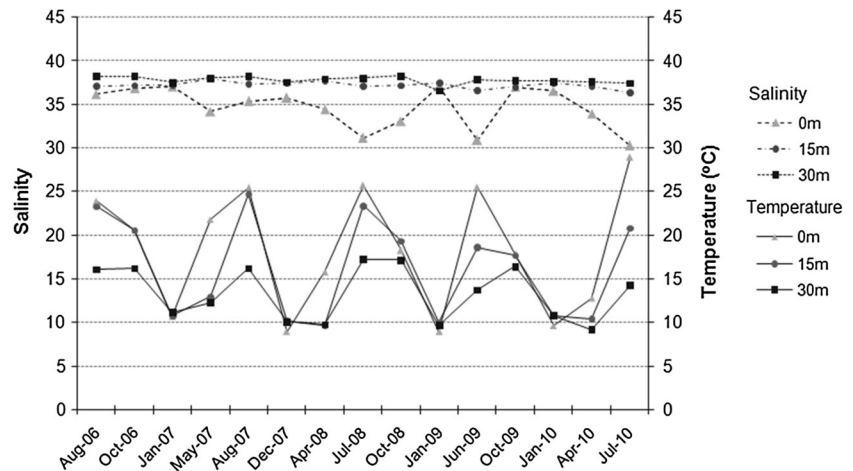
The baseline level of  $^{137}\text{Cs}$  in the Adriatic surface seawater in 1985 was around  $4 \text{ Bq m}^{-3}$ . The Chernobyl accident of 1986 increased  $^{137}\text{Cs}$  activity concentrations by 2 orders of magnitude, but by 1990, they went back to their pre-Chernobyl values [28]. At the present, the  $^{137}\text{Cs}$  activity concentration in the seawater of the northern Adriatic ranges from 1.5 to  $6.5 \text{ Bq m}^{-3}$  (Fig. 3 and 4).

### The station Po River delta

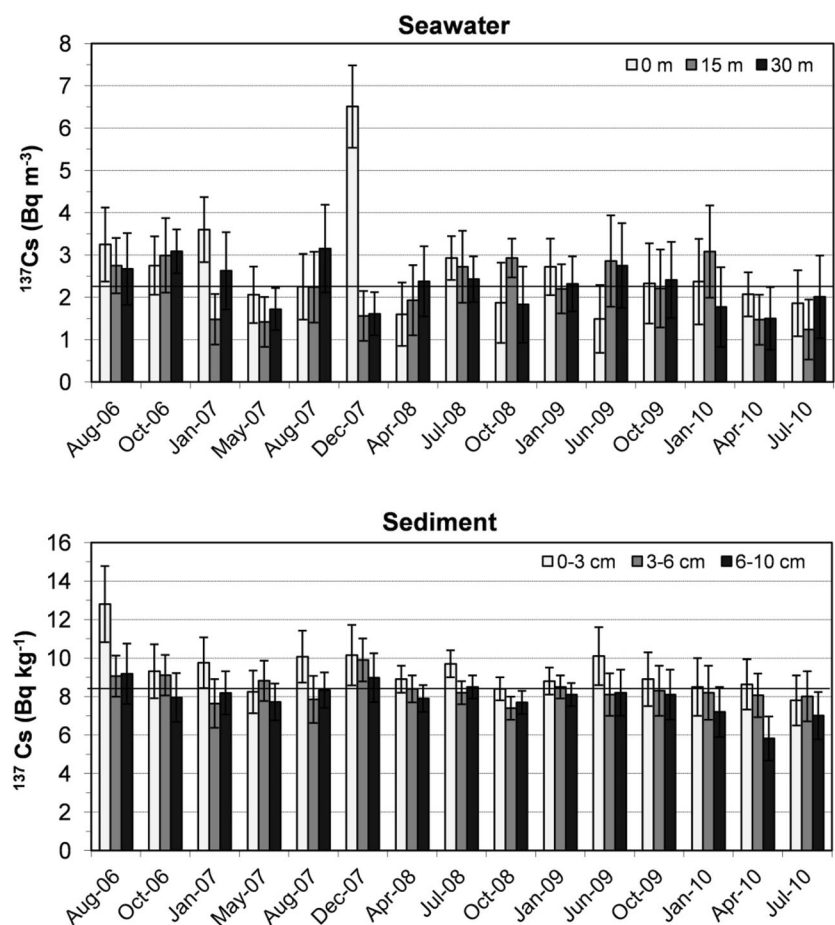
The main source of freshwater in the Adriatic is the Po River, with an annual mean discharge rate of  $1700 \text{ m}^3 \text{ s}^{-1}$ , carrying 28 % of the total runoff into the Adriatic [1]. The seawater temperature and salinity at the Po River delta are presented in Fig. 2. This station had a lower salinity in the surface seawater (0 m in Fig. 2) due to the influx of fresh water from the Po River. Decreased salinity in the surface seawater was most pronounced in the spring and in the summer particularly rainy months. However, in vicinity of the Po River delta freshwater influx can induce stratification of the water column (pycnocline), which is generally pronounced in the warm part of the year [13].

At the station Po River delta,  $^{137}\text{Cs}$  concentrations measured in the seawater were in the range between 1.42 and  $6.51 \text{ Bq m}^{-3}$  (Fig. 3). Seasonal differences in  $^{137}\text{Cs}$  concentrations at the three investigated depths are not

**Fig. 2** Seawater temperature and salinity at the Po River delta



**Fig. 3**  $^{137}\text{Cs}$  activity concentrations in seawater at 3 depths (0, 15, and 30 m) and in 3 sediment layers (0–3, 3–6, and 6–10 cm) at the Po River delta (station 108). Error bars indicate  $2\sigma$  propagated analytical uncertainty. Horizontal lines represent average values of all measurements



significant, with the exception of the surface seawater in December 2007 ( $6.51 \text{ Bq m}^{-3}$ ). Caesium is present in the seawater as the positive ion  $\text{Cs}^+$  and its distribution in the water column is mainly controlled by water dynamics, but hydrological barriers (e.g. halocline) obstruct free transport of caesium ions in the water column [5]. Intensity of the Po River spreading across the northern Adriatic depends not

only on discharge rates but also on meteorological factors, so in spite of lower discharge in 2007 ( $<335 \text{ m}^3 \text{ s}^{-1}$ ) [29] large amounts of fresh water can reach the investigated station Po River delta [15]. In addition, the fresh water accumulates within closed gyres [29], increasing the amount of  $^{137}\text{Cs}$  with time. When the water column is highly stratified, as seems to be the case in December 2007

[15], stratification can inhibit  $^{137}\text{Cs}$  sinking to lower layers and the surface concentration of  $^{137}\text{Cs}$  increase. This stratification in December 2007 causes the lowest  $^{137}\text{Cs}$  activity at 30 m depth (Fig. 3).

Average of all  $^{137}\text{Cs}$  activity concentrations in seawater ( $2.22 \pm 0.77 \text{ Bq m}^{-3}$ ) was lower than the pre-Chernobyl values. This is most likely due to the reduced global  $^{137}\text{Cs}$  fallout from other nuclear activities compared to the baseline levels from 1985 [4, 18]. The vertical profile of  $^{137}\text{Cs}$  in the seawater column at the station did not decrease from the surface to the bottom. The average activity concentrations of  $^{137}\text{Cs}$  at 0 m was  $2.64 \text{ Bq m}^{-3}$ , in 15 m was  $2.21 \text{ Bq m}^{-3}$  and in 30 m was  $2.28 \text{ Bq m}^{-3}$ . In the seawater column the differences in  $^{137}\text{Cs}$  concentrations are not significant as the depth increased (Fig. 3). The vertical profiles of  $^{137}\text{Cs}$  in the seawater column of the open sea, usually show decreasing activity concentrations with depth in the Eastern and Western Mediterranean [5, 6, 10]. As explained earlier, the station Po River delta has specific water mass dynamics with a pronounced seasonal stratification. At this station salinity presented at 15 and 30 m depths are nearly equal (Fig. 2), indicating uniform salinity during the research period, and this is the main reason for absence of differences in vertical distribution of  $^{137}\text{Cs}$  concentrations.

At the Po River delta, the  $^{137}\text{Cs}$  activity concentration in the surface marine sediment was measured to a depth of 10 cm in 3 sediment layers. As it is shown in Fig. 3, in the sediments the lowest  $^{137}\text{Cs}$  activity concentrations measured in the layer from 6 to 10 cm ( $5.82 \text{ Bq kg}^{-1}$ ) in April 2010, and the highest in the layer from 0 to 3 cm ( $12.80 \text{ Bq kg}^{-1}$ ) in August 2006. The average activity concentration of  $^{137}\text{Cs}$  in the 0–3 cm layer was  $9.34 \text{ Bq kg}^{-1}$ , in 3–6 cm was  $8.37 \text{ Bq kg}^{-1}$  and in 6–10 cm was  $7.93 \text{ Bq kg}^{-1}$ .  $^{137}\text{Cs}$  decline with depth of sediment is evident, but the difference between  $^{137}\text{Cs}$  activity concentrations in the measured sediment layers was not significant, even seasonally. The vertical profiles of  $^{137}\text{Cs}$  in Mediterranean sediments had a progressive decrease towards the surface, depending on the type of sediment and the sedimentation rate [5]. The Po River delta had a silty clay sediment type with sedimentation rate approximately around  $2.0 \text{ cm yr}^{-1}$  [4, 25]. The granulometric and geochemical composition of the sediment in this station reflected big river inputs and high suspended matter content in the seawater [4]. Differences in  $^{137}\text{Cs}$  activity concentrations in sediments might be related not only to the type of sediment, but also with input source. Results indicate that the discharge from the Po River could be an important supplier of  $^{137}\text{Cs}$  to the coastal environment. These results from the Po River delta seemed in agreement with previously published results by Delfanti et al. [16] and Nonnis Marzano and Triulzi [18], which were proved that

distribution of radiocesium in the Adriatic coastal sediments is mainly controlled by rivers. The results of this study are comparable to the activity concentration levels of  $^{137}\text{Cs}$  measured in the surface sediments of the Mediterranean Sea ( $2\text{--}10 \text{ Bq kg}^{-1}$ ) [10].

### The station Lim Bay

The water in Lim Bay is brackish, because underwater sources of freshwater cause the salinity to vary seasonally [30]. In the station Lim Bay, the average temperature and salinity of seawater in April is  $15.7^\circ\text{C}$  and 31.0, and in October  $18^\circ\text{C}$  and 35.6, respectively.

Inside the Lim Bay, the lowest  $^{137}\text{Cs}$  activity concentrations measured in surface seawater was  $0.91 \text{ Bq m}^{-3}$  in April 2010, and the highest was  $2.50 \text{ Bq m}^{-3}$  in October 2008 (Fig. 4). An average activity concentration of  $^{137}\text{Cs}$  in seawater was  $1.72 \pm 0.77 \text{ Bq m}^{-3}$ . The main reason for the low activity of  $^{137}\text{Cs}$  concentration in the seawater is the lack of local sources of radionuclides in the Lim Bay.

In the mussel *Mytilus galloprovincialis*  $^{137}\text{Cs}$  activity concentrations were under detection limit in this period ( $<0.3 \text{ Bq kg}^{-1}$ ) (Fig. 4). *Mytilus galloprovincialis* displayed presence of caesium only once during the monitored period,  $^{137}\text{Cs}$  activity was  $0.34 \text{ Bq kg}^{-1}$  in October 2006, which was slightly more than the detection limit. Lim bay has specific environmental conditions e.g. underwater freshwater springs and a protected area used for mariculture [31]. Therefore, according to data from previous biomonitoring research, such a chemical sediment analysis [32], toxicity and genotoxicity [33, 34], biological effects of pollution [20], the Lim Bay is selected as a reference location.

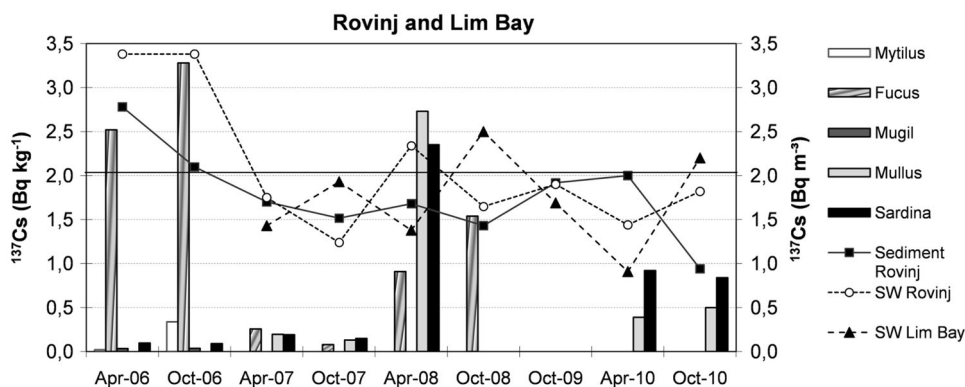
Mediterranean Mussel Watch (MMW) is a project for radioactivity monitoring of Mediterranean coastal waters using mussels *Mytilus galloprovincialis* as a bioindicator [21]. In the western Mediterranean Basin measured  $^{137}\text{Cs}$  activity in mussels were found to be very low (usually  $<1 \text{ Bq kg}^{-1}$ ) from 2004 to 2006 [35]. The concentrations at Croatian stations were  $0.08 \text{ Bq kg}^{-1}$  [35], which are comparable with the results of this study.

### The station Rovinj coastal area

In the Rovinj coastal area, average  $^{137}\text{Cs}$  activity concentration in surface seawater was  $2.10 \pm 0.65 \text{ Bq m}^{-3}$  (Fig. 4). The station lies close to the touristic city Rovinj and is under anthropogenic influence, but without influence of local radionuclide sources. The highest  $^{137}\text{Cs}$  activity concentrations measured in surface seawater was  $3.38 \text{ Bq m}^{-3}$  in April and October 2006, and lowest was  $1.24 \text{ Bq m}^{-3}$  in October 2007. The results indicate a decrease in  $^{137}\text{Cs}$  activity in surface seawater in this area



**Fig. 4**  $^{137}\text{Cs}$  activity concentrations in sediment and biota samples ( $\text{Bq kg}^{-1}$ ), seawater ( $\text{Bq m}^{-3}$ ) from Lim Bay and the Rovinj coastal area. Horizontal line represents average value of all measurements  $^{137}\text{Cs}$  activity concentrations in seawater



over time, the results are comparable to the literature data for the same trend in the Mediterranean Sea [10].

Average activity concentration of  $^{137}\text{Cs}$  in surface sediment layer was  $1.88 \pm 0.63 \text{ Bq kg}^{-1}$  (Fig. 4). As in the seawater, in sediment  $^{137}\text{Cs}$  activity have declined from  $2.78 \text{ Bq kg}^{-1}$  in April 2006 to  $0.94$  in October 2010. The Rovinj coastal sediment type is silty sand [25]. Fine grained clay sediments generally contain more  $^{137}\text{Cs}$  than sandy sediments [16]. This is in accordance with higher average  $^{137}\text{Cs}$  activity concentrations in the sediments of the Po River delta ( $8.70 \text{ Bq kg}^{-1}$ ), and is lower in the sediments of the Rovinj coastal area ( $1.88 \text{ Bq kg}^{-1}$ ) (Figs. 3, 4).

The removal of  $^{137}\text{Cs}$  from seawater by biological activity has transferred the radionuclide to the pelagic and benthic organisms within the trophic web [8]. The transfer path from algae, filter feeders mollusc, planktophagous to omnivorous fish has therefore been followed. The results of  $^{137}\text{Cs}$  activity concentration measurements in selected organisms from the Rovinj coastal area indicate that some species accumulate radionuclides more intensely than others in relation to their level in the trophic chain.

During the monitored period, in the brown alga *Fucus virsoides* average activity concentration of  $^{137}\text{Cs}$  was  $1.43 \pm 0.39 \text{ Bq kg}^{-1}$  w.w. Activity was detected throughout the measurement period in the range from  $0.08$  (October 2007) to  $3.28 \text{ Bq kg}^{-1}$  (October 2006). It should be noted that since October 2009, the brown alga population has been destroyed at the station due to construction of a new hotel. The brown alga *Fucus vesiculosus* has been used in the past in the Baltic Sea as a bioindicator organism for marine radiocontamination [19]. In this case, the endemic Adriatic species *Fucus virsoides* was selected and its ability for  $^{137}\text{Cs}$  accumulation was investigated.

In sardine *Sardina pilchardus* average  $^{137}\text{Cs}$  activity concentration was detected  $0.58 \pm 0.30 \text{ Bq kg}^{-1}$  w.w., ranged from  $0.10$  (April 2006) to  $2.35 \text{ Bq kg}^{-1}$  (April 2008). But in October 2008 and 2009  $^{137}\text{Cs}$  activity were below detection limits ( $<0.3 \text{ Bq kg}^{-1}$ ) (Fig. 4). Sardines are commercially fished for a variety of uses. A coastal

pelagic fish, feeds mainly on plankton. Because they are low in the food chain, sardines are very low in contaminants such as mercury, relative to other fish commonly eaten by humans [18].

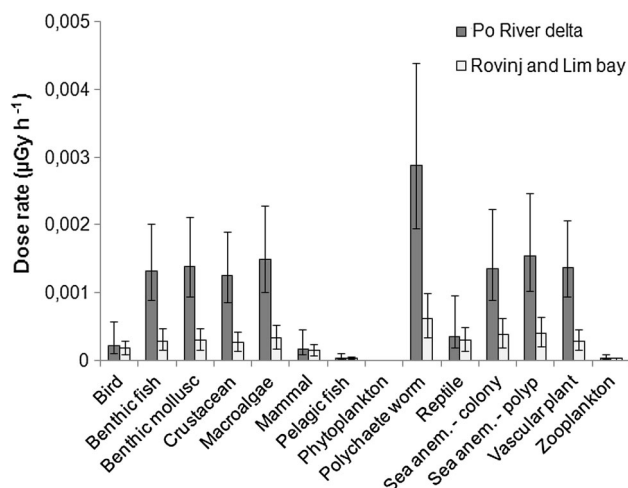
In fish the lowest  $^{137}\text{Cs}$  activity concentration was found in grey mullet *Mugil cephalus*  $<0.3 \text{ Bq kg}^{-1}$  w.w. Grey mullet is an important food fish species in the Rovinj coastal area. This is a benthic fish and usually schools over sand or mud bottoms, feeding on zooplankton and algae. After the first year of research (2006) grey mullet was replaced due to the low accumulation of  $^{137}\text{Cs}$  from seawater with another benthic fish, red mullet *Mullus barbatus*. In red mullet *Mullus barbatus*  $^{137}\text{Cs}$  activity concentration ranged from  $0.13$  (October 2007) to  $2.73 \text{ Bq kg}^{-1}$  (April 2008), with average values was  $1.02 \pm 0.69 \text{ Bq kg}^{-1}$  w.w. (Fig. 4). In the same time period as with the sardine, in October 2008 and 2009,  $^{137}\text{Cs}$  activity were below detection limits ( $<0.3 \text{ Bq kg}^{-1}$ ). This is a commercially important benthic species, which feeds on benthic invertebrates (crustaceans, worms, molluscs) along sandy and gravel bottoms.

From this study it is evident that investigated marine organisms had different values of  $^{137}\text{Cs}$  concentration and have not followed a bioaccumulation through the trophic chain. Uptake of  $^{137}\text{Cs}$  from seawater was expressed, respectively, as concentration factors (CF), defined as the ratio of radionuclide concentration in the organism and that in seawater [6]. It is simply a value that relates the concentration in the organism which may have been derived by uptake from the seawater, particulate matter, and from food, to that of the environment in which it lives [36]. In this study the highest  $\text{CF} = 641$  was found in *Fucus virsoides*, followed by *Mullus barbatus* with  $\text{CF} = 457$  and *Sardina pilchardus* with  $\text{CF} = 260$ . IAEA recommended CF value for macro-algae is 50, for mollusc is 30, and for fish is 100. Higher CF values compared to the ones recommended by IAEA indicated a higher caesium bioaccumulation of these organisms [36]. The results of the investigation indicate that *Fucus virsoides*, *Mullus barbatus* and *Sardina pilchardus* can be considered as good

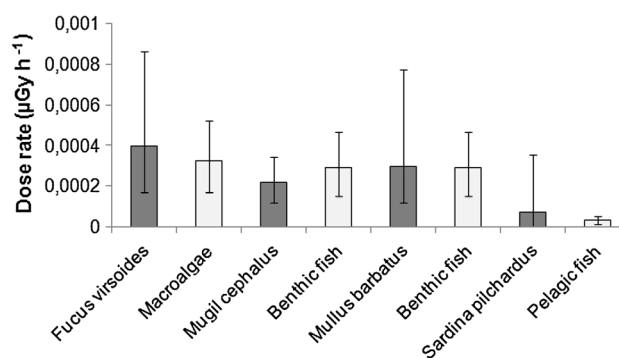
$^{137}\text{Cs}$  accumulating organisms for monitoring radiocontamination in the Adriatic Sea. Successfully determining the organisms that concentrate higher  $^{137}\text{Cs}$  activities from the marine environment could be useful in the estimation of the state of the environment, as well as in future plans for the protection of the Adriatic Sea.

### Dose assessment

Total  $^{137}\text{Cs}$  dose rate assessment for the reference organisms from the ERICA Assessment Tool are presented in Fig. 5, where the error bars represent dose rates for minimum and maximum  $^{137}\text{Cs}$  activity concentrations measured in seawater and sediments. It is evident that the average total dose rates at the Po River delta are substantially higher compared to those in Rovinj and Lim Bay for reference organisms, whose residence time is mostly in the sediment or on the sediment surface (benthic fish, benthic molluscs, crustacean, macro-algae, polychaete worms, sea anemones and vascular plants). This is due to the substantially higher average  $^{137}\text{Cs}$  sediment activity concentration at the Po River delta as an influence from the discharge from the Po River. Higher concentrations of caesium in sediments close to the river mouth are caused by abundant organic matter in this area which facilitates radionuclide accumulation [4, 16]. Since average  $^{137}\text{Cs}$  activity concentrations in seawater were almost identical for the Po River delta, the Rovinj coastal area and Lim Bay, this difference is not evident for reference organisms residing in seawater or on the seawater surface (birds, mammals, pelagic fish, phytoplankton, reptiles, and zooplankton). Nevertheless, total dose rates for all of these organisms were significantly below the IAEA and UNSCEAR dose rate screening values ( $400\ \mu\text{Gy h}^{-1}$ ),



**Fig. 5** Total dose rates for reference organisms due to  $^{137}\text{Cs}$  calculated with ERICA Assessment Tool at the Po River delta, Lim Bay and Rovinj coastal area



**Fig. 6** Total dose rates for specific sampled organisms and reference organisms due to  $^{137}\text{Cs}$  calculated with ERICA Assessment Tool at the Rovinj coastal area and Lim Bay

below which one cannot expect to find any measurable population effects due to exposure to  $^{137}\text{Cs}$  [37, 38]. Therefore, it can be concluded that additional doses due to  $^{137}\text{Cs}$  in the environment of the northern Adriatic Sea should not cause any harm to the marine biota.

Figure 6 contains total dose rate calculations obtained using the ERICA Assessment Tool for specific organisms, which were sampled during the investigated time period in the Rovinj coastal area and Lim Bay. Similarly as in Fig. 5, the error bars represent minimum and maximum  $^{137}\text{Cs}$  activity concentration in specific organism. For comparison, the total dose rates for the reference organisms which are biologically similar to specific organism are also shown. As is evident from Fig. 6, most of the average dose rates for specific organisms are similar to the reference organism, showing that reference organisms describe average dose rates for specific organisms reasonably well. Nevertheless, the maximum values for *Fucus virsoides*, *Mullus barbatus* and *Sardina pilchardus* are much higher compared to those from reference organisms, which were calculated from seawater and sediment  $^{137}\text{Cs}$  activity concentrations. This is further evidence that these organisms could be used for monitoring radioactive contamination in the Adriatic Sea, because they could be considered as good  $^{137}\text{Cs}$  accumulating organisms. Similarly to the reference organisms, also for specific organisms, the maximum total dose rates were much lower than the threshold values below which one cannot expect to find any significant side effects of  $^{137}\text{Cs}$  on those organisms ( $400\ \mu\text{Gy h}^{-1}$ ) [37, 38].

### Conclusions

Antropogenic radioactivity in the northern Adriatic Sea was assessed by analysing  $^{137}\text{Cs}$  activity concentrations in seawater, sediment, and marine organisms at stations the Po River delta, Lim Bay and the Rovinj coastal area, from 2006 to 2010.

$^{137}\text{Cs}$  activity concentrations in the seawater were range from  $0.91 \text{ Bq m}^{-3}$  in Lim Bay to  $6.51 \text{ Bq m}^{-3}$  in the Po River delta, average  $^{137}\text{Cs}$  in the northern Adriatic Sea was  $2.01 \text{ Bq m}^{-3}$ .

The highest average activity concentration of  $^{137}\text{Cs}$  in sediments was found in the Po River delta ( $8.70 \text{ Bq kg}^{-1}$ ) the lower was found in the sediments of the Rovinj coastal area ( $1.88 \text{ Bq kg}^{-1}$ ).

In investigated marine organisms from the Rovinj coastal area results of  $^{137}\text{Cs}$  activity concentrations and concentration factors (CF) were showed that the brown algae *Fucus virsoides*, fishes *Mullus barbatus* and *Sardina pilchardus* can be considered as good  $^{137}\text{Cs}$  accumulating organisms. These organisms should be used as indicators in future monitoring schemes and possible contamination in the Adriatic Sea.

Radioecological condition of the northern Adriatic Sea is satisfactory and there is no significant input of radionuclides from fallout and rivers discharge. This is also substantiated by the results of total dose rates calculated using the ERICA Assessment Tool, which are all much lower than the IAEA and UNSCEAR proposed threshold value of  $400 \mu\text{Gy h}^{-1}$ .

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