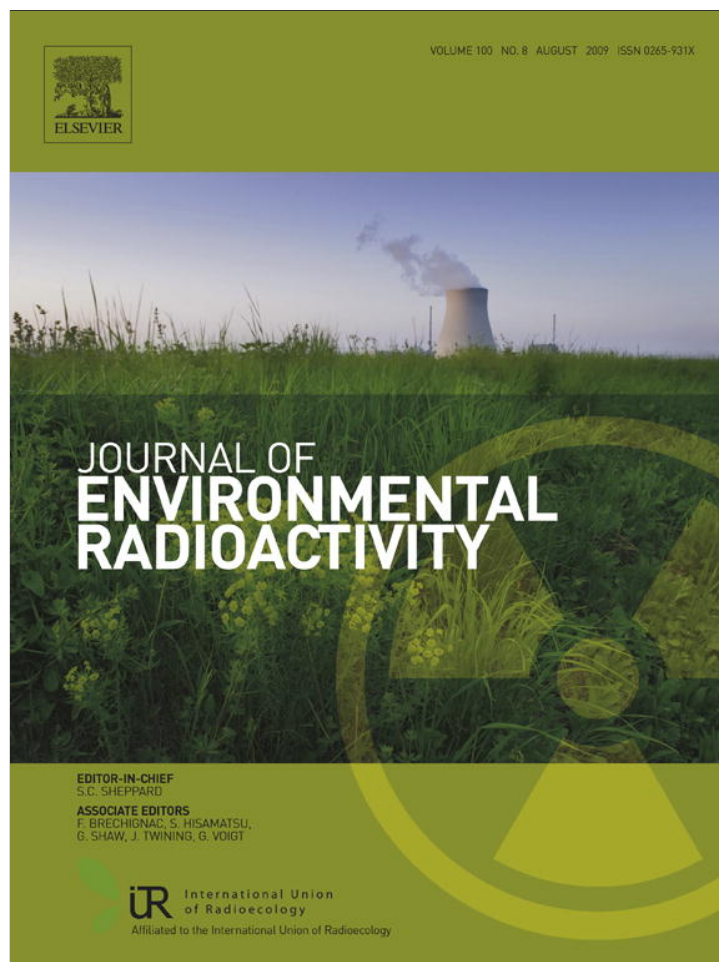


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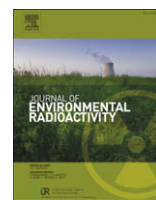
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Temporal and spatial distribution of ^{137}Cs in Eastern Mediterranean Sea. Horizontal and vertical dispersion in two regions

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ABSTRACT

Caesium-137 activity concentration in the water columns of the Gulf of Patras (Central Greece) and the North-Eastern Aegean Sea (easterward to Lemnos Island) was investigated in selected sampling stations during the period September 2004–June 2006. The methodology followed was based on the sorption of caesium (Cs) on cotton wound cartridge filters impregnated by $\text{Cu}_2[\text{Fe}(\text{CN})_6]$ via in-situ pumping. In terms of the horizontal and vertical records, the activity concentrations of ^{137}Cs in the Gulf of Patras ranged between 1.2 and 6.7 Bq m^{-3} , depending on the sampling period and the prevailing physico-chemical regime at the sampling station. The general pattern of the decreased activity concentrations of ^{137}Cs with increasing depth was reversed in the Gulf of Patras during the cold period attributed to the prevailing advective processes of the area. The activity concentrations of ^{137}Cs in the North-Eastern Aegean Sea ranged from 2.6 to 12.8 Bq m^{-3} , whereas significant stratified curves were observed during the warm period and also, in one station during the cold period. In terms of temporal variation, the discharges in the Gulf of Patras resulted in enhanced levels of ^{137}Cs , whereas in the North Aegean Sea the incoming water masses from the Black Sea had an apparent influence throughout the year by increasing the ^{137}Cs levels, hence presenting a weak seasonal variation. Comparing the two studied areas, one could say that the North Aegean Sea, as an open sea environment, presented higher concentrations due to the influence of the Black Sea water masses. The estimated inventories of ^{137}Cs in the Gulf of Patras ranged 0.25 ± 0.03 – $0.79 \pm 0.03 \text{ kBq m}^{-2}$, whereas in the North-Eastern Aegean Sea they ranged 0.33 ± 0.02 – $0.92 \pm 0.03 \text{ kBq m}^{-2}$.

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1. Introduction

Artificial radionuclides have been introduced to the marine environment from several sources, such as the fallout from atmospheric nuclear weapon tests (1945–1960), accidents such as the former USSR Chernobyl reactor accident in 1986, discharges from nuclear installations, contributions from nuclear testing sites, nuclear submarine accidents, loss of radioactive sources, applications in medicine and in industry and the burn up of satellites using radionuclides as a power source (Nakano and Povinec, 2003; Povinec et al., 2003a). As the marine environment is a dynamic system, radionuclides introduced to surface waters by liquid discharges (wet and dry) do not stay there in steady-state conditions, but due to currents and other processes in the water column, are transported both horizontally and vertically to different regions,

as well as to bottom waters and sediments. The marine environment acts as the final repository of the terrestrial radioactive load removed by weathering processes carried by rivers.

One of the main interests in studying the behavior of radionuclides in the marine environment derives from the fact that radionuclides are powerful and very useful tracers (Povinec et al., 2005). For example, the conservative ^{137}Cs (half-life 30 y) and ^{90}Sr (half-life 29 y) have been reported in the literature as useful tools for studying water movements (Delfanti et al., 2003; Florou et al., 2006; Franic et al., 1999; Franic and Bauman, 1993; Papucci and Delfanti, 1999).

One of the most important artificial radionuclide is ^{137}Cs , due to its half-life, the radiation type it emits during radioactive decay and its bioaccumulation by organisms (as an alkali metal it follows the metabolic route of potassium, which is an essential element for life) (Woodhead, 1973). Caesium-137 is distributed in water, mostly in ionic form (70%), approximately 23% as particle-reactive and 7% in a colloidal phase. As caesium (Cs) is a conservative element, most of the Chernobyl ^{137}Cs remains in the water column in the impacted

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regions. Even after 100 years from the Chernobyl Nuclear Power Plant (NPP) accident, 10% of the ^{137}Cs released is expected to be present in the environment (Buesseler, 1987). The conservative nature of ^{137}Cs in the water column is responsible for the fact that its distribution is primarily related to the mixing process in the oceans (Povinec et al., 2003b). The observed mean effective half-life, based on the environmental elimination parameters (expressed as “the ecological half life of ^{137}Cs ”), which control the transient times of Cs, is much shorter than the radioactive decay half-life, suggesting that advection and dispersion processes are important and effective in removing this radionuclide from the water column.

It is well known that ^{137}Cs as well as other radionuclides are deposited to the sea bottom sediments through a wide range of processes, including fixation on suspended particulate matter, direct precipitation of colloidal forms (coagulation, aggregation), direct fixation by adsorption, absorption on clay minerals and complexation with organic matter. As it is known, Cs has a strongly affinity with the organic matter, and consequently, the nature of the organic matter is important in terms of effective binding and fixation of ^{137}Cs to particles by active ligand groups, affecting transfer and sedimentation processes (Ligero et al., 2001).

The major sources of ^{137}Cs to the Mediterranean Sea have been identified to be the global fallout from nuclear weapon testing, mainly in the period 1945–1960 and the Chernobyl accident in 1986 (Florou et al., 1994; Lee et al., 2003). Since 1986, the radiological status of the Eastern Mediterranean Aegean Sea has changed. During 1986 the average deposition of ^{137}Cs from the fallout was estimated to be approximately 4 kBq m^{-2} , whereas the respective value for the Ionian Sea was considerably lower, 2.5 kBq m^{-2} (Florou and Kritidis, 1994). The total Cs ($^{137}\text{Cs} + ^{134}\text{Cs}$), 2/3 of this value was ^{137}Cs input from Chernobyl fallout in the Black Sea and the Eastern Mediterranean has been estimated to be 2400 TBq for the Black Sea, 820 TBq for the Aegean Sea and 600 TBq in the Ionian Sea (60 TBq in the zone of 50 km across the Greek coasts) (Florou, 1996; Kritidis et al., 1990).

After 2000, the radiological status of the Greek marine environment was characterized by higher values in the North Aegean Sea (mean ^{137}Cs activity concentration $13.3 \pm 1.3 \text{ Bq m}^{-3}$) and decreasing ones in the Southern Aegean (with a mean value of 5 Bq m^{-3}) (Florou et al., 2006). Similarly, in the Ionian Sea higher ^{137}Cs activity concentrations were observed in its northern part and lower ones in the southern ($6.1 \pm 0.5 \text{ Bq m}^{-3}$ and $3.0 \pm 0.1 \text{ Bq m}^{-3}$, respectively) (Papucci and Delfanti, 1999).

The objectives of this work were the following: (i) to examine the horizontal and vertical distribution of ^{137}Cs in two relatively shallow but different regions, which are subject to different influences, namely the Gulf of Patras adjacent to the Ionian Sea (Western Greece), and the wide area of the mouth of Dardanelles, near Lemnos Island in the North-Eastern Aegean Sea (Fig. 1), (ii) to evaluate a contribution of the regional impact sources to the ^{137}Cs activity concentration, (iii) to compare the distribution pattern of ^{137}Cs in a semi-enclosed gulf (Gulf of Patras), with that of the open sea (North-Eastern Aegean Sea), and (iv) to define the seasonal variation of ^{137}Cs in the areas of study.

2. Background oceanography of the study areas

2.1. The Gulf of Patras

The Gulf of Patras is located in central part of Greece between the mainland and the Peloponesus peninsula (Fig. 1). It is a shallow marine embayment linking the relatively deep Gulf of Corinth with the Ionian Sea. At its east end, the Rio Straits connect it to the Gulf of Corinth. The Evinos River delta and the Messolongi lagoon lie at the north-western shore of the gulf. The streams Pirros and Glafkos

discharge at the southern shores of the gulf. The river Evinos (catchment area 635 km^2) with mean annual discharge of approximately $27.6 \text{ m}^3 \text{ s}^{-1}$ is the most important fresh water input in the system. The freshwater supplied by Pirros and Glafkos is considerably lower. Freshwater inputs are important for the system as they are directly linked with the particulate material transported from the land and as a result, with the pollutants adsorbed onto particles, which enter the sea (Lu and Matsumoto, 2005; Paller et al., 2005). Furthermore, low salinity fresh waters are mixed with the highly saline seawaters decreasing the surface salinity of the gulf near the river outlets and thus affecting the local regime (Uncles et al., 2000). The city of Patras with a population of about 200,000 is situated on the east side of the gulf and it is the biggest urban centre of the region, characterized by a busy port and an industrial area spread along the southern coast (Frigilos, 1987).

During summer, the water column of the Gulf of Patras is stratified into two layers. The upper one has a thickness of about 12 m with a mean temperature of $23.5 \text{ }^\circ\text{C}$, and the salinity varying between 38.3 and 39.1 (Hasiotis et al., 1996). The bottom layer, which extends to the seabed, is of approximately 17 m thick with a mean temperature of $16.6 \text{ }^\circ\text{C}$, and salinity ranging between 38.9 and 39.1. The two layers are separated by a sharp thermocline with a gradient of $1.3 \text{ }^\circ\text{C m}^{-1}$. The water circulation in the Gulf of Patras is characterized by cyclonic structures. Seawater masses coming from the Ionian Sea flow through the southern coastline, whereas seawater coming from the Corinthiakos Gulf, through Rion – Antirion Straits, affects mostly the northeastern part of the Gulf (Papageorgiou et al., 1987). During winter the surface temperature is considerably lower and virtually there is no stratification. The advective transport in the area is affected by the exchange of seawater with Ionian Sea at the western part of the gulf and with the Gulf of Corinth at the eastern part.

2.2. The North-Eastern Aegean Sea

One of the basic characteristics of the North-Eastern Aegean Sea is the fact that it exchanges waters with the Sea of Marmara, which acts as a conduit between the Aegean and the Black Sea. The Sea of Marmara is an enclosed depression connected with the Aegean Sea through the Dardanelles Straits and with the Black Sea through the Bosphorus Straits (Ergin et al., 1991).

Within this system two-layer stratification and diverse flow circulation is established, on the basis of density differences between the Aegean and Black Sea Waters (BSW) (Oguz and Sur, 1989). The two different water masses are separated by a pycnocline, at an average depth of 25 m. The density profile is dominated by salinity, even though there are also significant seasonal variations in water temperature. The upper layer is occupied by the less saline BSW, whereas the lower layer by the denser Aegean Sea Waters (ASW). The surface layer in the Dardanelles flows towards the Aegean Sea, at velocities varying between 50 and 200 cm s^{-1} , with an average of 85 cm s^{-1} . The current at the bottom layer moves to the opposite direction, towards the Sea of Marmara, with velocities ranging from 20 to 40 cm s^{-1} (Lykousis et al., 2002).

A volume of approximately $1.3 \times 10^3 \text{ km}^3$ of colder and less saline water flows annually into the Aegean Sea whilst, at the same time, $1.0 \times 10^3 \text{ km}^3$ of the more saline ASW enters the Sea of Marmara through the bottom currents, resulting in a net annual flow ($\text{BSW}_{\text{surface}} - \text{ASW}_{\text{bottom}}$) of $300 \text{ km}^3 \text{ yr}^{-1}$ through the Dardanelles Straits (Lykousis et al., 2002). Both surface outflow and bottom water inflow fluctuate seasonally, as they are dependent upon wind and density gradients above and below the pycnocline. The largest flow towards the Aegean occurs in late spring and summer, corresponding to periods at which the precipitation and river runoff discharged into the Black Sea increase substantially.

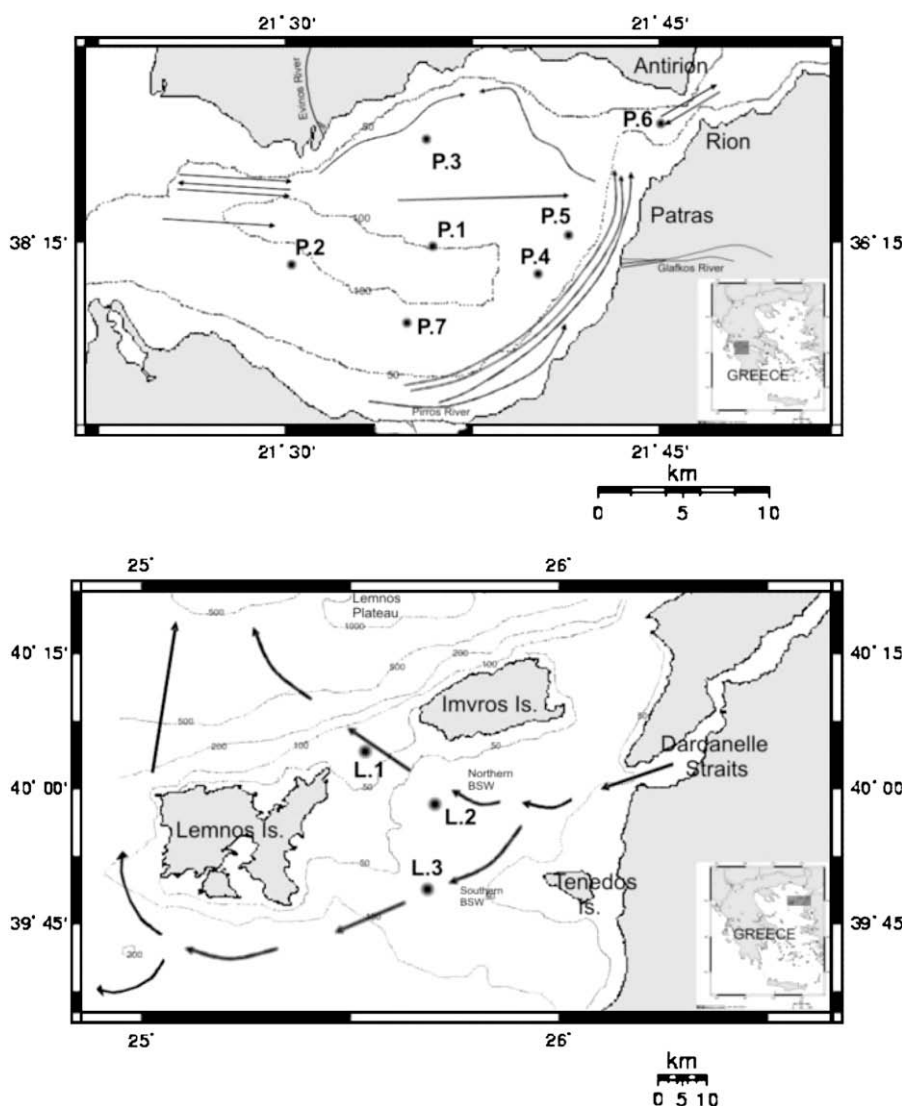


Fig. 1. Sampling stations in the areas of study (gulf of Patras and North-Eastern Aegean Sea) and main current circulation.

Zodiatis (1994), investigating the advection of BSW into the northern Aegean Sea, found that it is related to the general westward and southward water movements characterized by small-scale cyclonic and anticyclonic flow structures. The BSW waters firstly bifurcate at eastern Lemnos Island. The southern BSW current then bifurcates into northerly and southerly directions (see Fig. 1). In addition, the spreading of the BSW is characterized by an interannual variability; the latter results from changes in the dynamic regime of the area (e.g. wind stress) (Zodiatis, 1994). Another interesting feature is the formation of the frontal zone caused by the northern BSW current, over the Lemnos Plateau, between the colder, less saline BSW and the warmer, saltier Levantine Waters (LW). At this location, dense and oxygen-rich water masses are formed; sinking subsequently, contributing to the ventilation of the deep waters of the Northern Aegean Sea. During the warm periods (summer–autumn) the BSW flows, in general, in a south-westerly direction. The core of this water mass is located to the south of Lemnos Island (Theocharis and Georgopoulos, 1993).

Notably, the temperature of the BSW is lower than the surface temperature of the Northern Aegean Sea throughout the year. The annual maximum of the surface temperature exceeds 24 °C around

August, whereas the minimum one is below 13 °C and it is reached in February, although in few cases the minimum occurs in January. Sea surface salinity values vary seasonally, ranging from less than 31 to more than 39. The relatively lower salinity values are associated with the BSW input and the riverine freshwater inputs. The latter are more important during spring. More saline surface waters cover the south-eastern part of the Aegean, influenced by the adjacent Levantine Basin.

3. Materials and methods

3.1. Preparation for sampling

3.1.1. Filter preparation – impregnation

Various transition metal ferrocyanides (copper ferrocyanide, zirconium ferrocyanide and potassium hexacyanocobalt) have been used for about 30 years for collecting Cs from sea water, either through precipitation or through in-situ adsorption (Lehto and Harjula, 1987; Loewenschuss, 1982; Nilchi et al., 2003). The advantage of using transition metal ferrocyanides is that they can be easily attached to the supporting material, due to their gelatinous form. The exchange capacity of some transition metal ferrocyanides is very high and it is also easy and cost effective to be prepared in the laboratory.

Table 1
Results of limit of detection (LOD) and limit of quantification (LOQ) experiments.

Date 13/05/2004	Blank (Bq m ⁻³)	Standard solution		
		Standard solution 1 4 Bq m ⁻³ ¹³⁴ Cs	Standard solution 2 6 Bq m ⁻³ ¹³⁴ Cs	Standard solution 3 8 Bq m ⁻³ ¹³⁴ Cs
Activity concentration (Bq m ⁻³)	0.2 ± 0.1 ^a (n = 2)	3.5 ± 0.4 ^a (n = 2)	5.2 ± 0.9 ^a (n = 2)	7.6 ± 1.0 ^a (n = 2)
Recovery %		88	87	95
LOD = Blank + 3 × SD _{Blank} ^a		0.5 Bq m ⁻³ (n = 2)		
LOQ = Blank + 10 × SD _{Blank} ^a		1.2 Bq m ⁻³ (n = 2)		

^a Standard deviation.

The impregnation of the filters was carried out by circulating water suspensions of K₄[Fe(CN)₆] and Cu(NO₃)₂ in a closed loop, employing a small pump and the filter to be impregnated. The filters collected the reddish brown precipitate that formed. When the circulating solution was clear, the impregnated filters were removed and dried in an oven at 60–80 °C for 24 h and packed for the sampling (Evangeliou et al., 2005; Roos et al., 1994).

3.1.2. Validation of the filter impregnation procedure

In order to validate this work, the in-situ pumping method was simulated in the laboratory to determine the recovery of the filters used in the method, the limit of detection (LOD) and the limit of quantification (LOQ) using the guidelines of Eurachem (Eurachem, 1998) (Table 1). Thus, six standard solutions of ¹³⁴Cs (2 in each concentration) with a concentration similar to that of ¹³⁷Cs in seawater samples (4, 6, 8 Bq m⁻³) were produced. The purpose of using ¹³⁴Cs was the fact that it is widely used as a carrier and yield tracer in radiochemical analyses, so it is easily available. The impregnated (ready-to-use) filters were submerged in the standard solutions and forced flux was applied by using a Watson–Marlow Briedel peristaltic pump. The flow rate was adjusted to 0.5 m³ h⁻¹, according to the flow rate of the in-situ pumping method. Afterwards, the filters were dried, ashed and measured by gamma spectrometry. Finally, two more ready-to-use filters were ashed and measured by gamma spectrometry in order to estimate the low limit of detection and the eventual background contamination rate due to the laboratory environment and the reagents used (mean activity in the blank filter was found approximately 0.2 ± 0.1 Bq m⁻³).

3.2. Sampling

Seawater samples were collected during two cruises carried out in the Gulf of Patras (Central Greece) in September 2004 and February 2005 and two more carried out in the North-Eastern Aegean Sea, near Lemnos Island, in December 2005 and June 2006. The grids of the stations sampled are given in Fig. 1, whereas the station co-ordinates in Tables 3–6.

Samples were collected through in-situ pumping performed in several depths using a battery powered large volume pump manufactured by Challenger Oceanic Services and Systems (COSS, United Kingdom). Sample volumes of 100–1500 L were processed at flow rates of 0.3–1.0 m³ h⁻¹. For the in-situ pre-concentration of ¹³⁷Cs, 2 impregnated filters were used in series with a 25 cm polypropylene wound cartridge blank pre-filter of 1 μm pore size for particulate matter scavenging following the procedure described by Roos et al. (1994). The accuracy of the flowmeter is ± 2%, operating in a range 22.7–2730 L h⁻¹ (Simpson, 1997). Temperature, salinity and pH were determined in-situ using CTD, model YSI 53.

Table 2
Surface and vertical activity concentrations of ¹³⁷Cs in the gulf of Patras (September 2004).

Co-ordinates	Depth (m)	Salinity (psu)	Temperature (°C)	pH	Density (sigma-t)	¹³⁷ Cs (Bq m ⁻³ ± SE ^a)
P.1 38° 14.498 N 21° 35.511 E	5	38.9	23.4	8.09	26.79	3.6 ± 0.1
	47	38.7	19.2	7.96	27.81	3.5 ± 0.1
	100	37.8	16.5	7.46	27.79	1.2 ± 0.1
P.2 38° 14.518 N 21° 29.158 E	5	36.8	24.4	7.80	24.91	2.8 ± 0.4
P.3 38° 17.340 N 21° 35.286 E	5	37.1	23.6	8.02	25.37	2.8 ± 0.3
P.4 38° 13.304 N 21° 39.250 E	5	38.7	21.7	8.96	27.14	2.2 ± 0.3
P.5 38° 14.131 N 21° 41.631 E	5	37.8	23.0	8.14	26.08	2.7 ± 0.6

^a The uncertainties quoted for ¹³⁷Cs activities are 1σ values derived from counting statistics.

3.3. Laboratory treatment and measurements

The filters were transferred to the laboratory, where they were dried and ashed in ovens at 350 °C. The resulting ash was then transferred into special measurement pots (radius 3.4 cm and height 2.0 cm) for gamma spectrometry measurements.

The measurements were carried out in a Camberra gamma spectrometry system comprising an HPGe Detector System with a relative efficiency of 90% and resolution of 2.1 keV (at the 1.33 MeV (photopeak of ⁶⁰Co)). The HPGe detector is connected to 8-k multi-channel analyzer operated with the Genie 2000 software. The energy calibration was performed using standard active sources of ²²Na, ⁵⁴Mn, ⁵⁷Co, ⁶⁰Co, ¹⁰⁹Cd, ¹³³Ba, ¹³⁷Cs and ²⁴¹Am covering an energy range up to 2000 keV (0.25 keV/ch). The duration of each measurement was about 7 × 10⁴ s. The detector's efficiency was calculated (in connection with energy) using a standard active source of ²²⁶Ra (240 Bq) under the same geometry as the measurement pot.

3.4. Sample analysis

Assuming the two impregnated filters have the same collection efficiency (*E*) and thus adsorb the same fraction of Cs, one may calculate the amount of dissolved Cs *A* (¹³⁷Cs) in the water volume sampled by considering the two filters to be the two first terms in a geometrical series. The total activity of ¹³⁷Cs can be calculated from the following equation:

$$A(^{137}\text{Cs}) = \frac{A_1}{E} = \frac{A_1}{1 - \frac{A_2}{A_1}}$$

where *A*₁ and *A*₂ are the ¹³⁷Cs activities of the first and second Cu₂[Fe(CN)₆] cartridge in series, respectively and *E* is the collection efficiency (Roos et al., 1994). The uncertainties quoted for ¹³⁷Cs activities are 1σ values derived from counting statistics. The final uncertainties were calculated by uncertainty propagation.

The statistical analyses were performed using the software Statistica. Specifically *t*-test for independent samples (groups) was performed in order to assess the seasonal variation of data obtained both from the Gulf of Patras and the North-Eastern Aegean Sea during winter and summer. Origin Pro 70 software was also used for plotting ¹³⁷Cs activity concentrations, salinity, temperature and density.

The activity concentrations of ¹³⁷Cs adsorbed onto particles of size up to 1 μm (deposited on pre-filters) were found below the LOD of 0.011 Bq m⁻³ indicating that almost all the injected Cs was in the soluble phase. The LOD and LOQ were estimated using the guidelines of Eurachem (Eurachem, 1998).

4. Results and discussion

4.1. The Gulf of Patras

4.1.1. September 2004

During the warm period (September 2004) the stations P.1–P.5 (Fig. 1) were used for surface sampling. Moreover, in P.1 (Central Gulf of Patras) vertical samples down to 120 m were collected, too. The results concerning the sampling campaign (activity concentrations of ¹³⁷Cs and associated environmental parameters) are given in Table 2. For P.1 the vertical distribution of ¹³⁷Cs is illustrated in Fig. 2.

The activity concentrations of ¹³⁷Cs in the 5 stations of the Gulf of Patras ranged from 2.2 ± 0.3 to 3.6 ± 0.1 Bq m⁻³ (Table 2). These values correspond to the lowest level found in the marine environment of the region, probably due to the small land impact and restricted mixing processes of the warm period. The maximum surface concentration was observed in the centre of Gulf of Patras, which is attributed to the cyclonic water mass movements that are

Table 3
Surface and vertical activity concentrations of ^{137}Cs in the gulf of Patras (February 2005).

Co-ordinates	Depth (m)	Salinity (psu)	Temperature ($^{\circ}\text{C}$)	pH	Density (sigma-t)	^{137}Cs ($\text{Bq m}^{-3} \pm \text{SE}^{\text{a}}$)	Ratio $\frac{\text{Cs}_{\text{surface}}}{\text{Cs}_{\text{bottom}}}$
P.1 38° 14.498 N 21° 35.511 E	5	37.5	15.8	6.40	27.75	4.2 ± 0.5	1.05
	47	38.4	14.0	8.47	28.83	3.5 ± 0.2	
	80	38.2	14.5	8.43	28.52	5.9 ± 0.1	
	100	38.7	14.1	8.41	29.07	5.0 ± 0.1	
P.2 38° 14.518 N 21° 29.158 E	5	38.1	17.7	8.41	27.72	4.0 ± 0.3	0.89
	54	38.5	13.8	8.43	28.94	4.5 ± 0.2	
P.3 38° 17.340 N 21° 35.286 E	5	38.3	16.6	8.41	28.17	4.4 ± 0.4	1.63
	54	37.8	15.3	8.40	28.30	2.7 ± 0.4	
P.4 38° 13.304 N 21° 39.250 E	5	38.5	17.3	8.41	28.15	6.7 ± 1.1	2.68
	54	38.1	14.7	8.44	28.67	2.5 ± 0.1	
P.5 38° 14.131 N 21° 41.631 E	5	38.1	16.3	8.43	28.08	4.4 ± 0.6	1.63
	54	38.2	14.3	8.46	28.84	2.7 ± 0.1	
P.6 38° 18.572 N 21° 44.807 E	5	36.2	15.7	6.33	26.75	6.1 ± 0.7	1.45
	50	37.6	14.6	8.35	28.05	4.2 ± 0.3	
	68	37.9	14.8	8.40	28.09	4.3 ± 0.2	
P.7 38° 11.854 N 21° 34.990 E	5	38.0	16.6	8.42	27.94	4.0 ± 0.4	1.11
	54	38.4	14.1	8.43	29.04	3.6 ± 0.3	

^a The uncertainties quoted for ^{137}Cs activities are 1σ values derived from counting statistics.

structures of convergence gathering the pollutants, consequently overlapping their sinking rate.

In terms of the associated environmental parameters recorded (salinity, temperature, density), two layers characterize the water column in the central station of the Gulf of Patras (Fig. 2). The surface layer extends to approximately 50-m depth followed by a denser layer characterized by low temperatures. In the upper part ^{137}Cs remains constant (surface 3.6 ± 0.1 to 50-m depth $3.5 \pm 0.1 \text{ Bq m}^{-3}$) and decreases in the deeper layer reaching $1.2 \pm 0.1 \text{ Bq m}^{-3}$ in 100-m depth. However, one could note that the separation of the column at 50-m depth is not so effective, due to the low resolution of the vertical profiles.

4.1.2. February 2005

During the cold period (February 2005) the stations P.1–P.7 (Fig. 1) were sampled at the surface and near the bottom. Vertical profile sampling was carried out in P.1 and P.6 (Fig. 3). The ^{137}Cs activity concentrations, obtained during this survey, are given in Table 3 together with the recorded physicochemical parameters. The surface concentrations varied from 4.0 ± 0.3 to $6.7 \pm 1.1 \text{ Bq m}^{-3}$. These values are comparable to those reported by several workers in other marine areas, namely $3.9\text{--}4.2 \text{ Bq m}^{-3}$ in the Gulf of Corinth (Evangelidou, 2005); $1.96 \pm 0.26 \text{ Bq m}^{-3}$ in South

Ionian Sea (Gulin and Stokozov, 2005); 3.5 Bq m^{-3} in the Levantine Basin and 3.3 Bq m^{-3} in the Lybian Sea (Papuacci and Delfanti, 1999), and lower than those reported for the North Aegean and the Black Sea, which were $13.3 \pm 1.3 \text{ Bq m}^{-3}$ (Florou et al., 2006) and $18.29 \pm 2.14 \text{ Bq m}^{-3}$, respectively (Gulin and Stokozov, 2005). On the other hand, they are higher than those measured in September 2004 (Tables 2 and 3), since during winter stronger mixing processes and higher impact from runoff usually occur. The maximum surface ^{137}Cs activity concentrations were observed at the coastal stations P.3, P.5 and P.7 and especially at the P.4 ($6.7 \pm 1.1 \text{ Bq m}^{-3}$) which is located next to the mouth of Glafkos River (Fig. 1). This may be partly attributed to desorption of ^{137}Cs from riverine particles entering the mixing system (Hayes and Sackett, 1987; Olsen et al., 1981).

The determined ^{137}Cs profiles (Table 3) can be classified in two groups according to the ratio between the surface and bottom activity concentrations. In the first group (stations P.1, P.2 and P.7) the bottom concentrations are similar to the surface ones. It is noteworthy that this group is located to the central-western part of the gulf (Fig. 1), where mass exchanges with the Ionian water are observed superficially. Moreover, the deep currents from the neighboring eastern Ionian Sea are transported to the Gulf of Patras (Papatheodorou and Kontopoulos, 1998). Thus, the high values

Table 4
Surface and vertical activity concentrations of ^{137}Cs in the North-Eastern Aegean Sea (December 2005).

Co-ordinates	Depth (m)	Salinity (psu)	Temperature ($^{\circ}\text{C}$)	pH	Density (sigma-t)	^{137}Cs ($\text{Bq m}^{-3} \pm \text{SE}^{\text{a}}$)
L.1 40° 04.22 N 25° 30.08 E	4	33.4	14.6	6.76	25.60	7.1 ± 0.2
	22	37.3	15.5	8.36	27.63	4.4 ± 0.1
	45	39.0	16.2	8.32	28.78	3.0 ± 0.3
	63	39.0	16.2	8.30	28.78	4.3 ± 0.3
L.2 39° 52.06 N 25° 39.16 E	4	35.2	15.1	5.89	21.35	5.1 ± 0.5
	22	38.3	15.7	8.03	28.32	7.3 ± 0.5
	45	38.7	16.0	8.04	28.52	3.7 ± 0.5
	65	38.9	15.9	8.08	28.92	2.8 ± 0.4
L.3 39° 48.18 N 25° 39.21 E	4	37.9	15.8	6.25	28.03	5.1 ± 0.6
	22	38.1	15.9	7.84	28.16	12.8 ± 0.3
	45	38.7	15.9	7.93	28.62	6.3 ± 0.4
	67	38.9	16.2	8.06	28.70	3.9 ± 0.3

^a The uncertainties quoted for ^{137}Cs activities are 1σ values derived from counting statistics.

Table 5
Surface and vertical activity concentrations of ^{137}Cs in the North-Eastern Aegean Sea (June 2006).

	Co-ordinates	Depth (m)	Salinity (psu)	Temperature ($^{\circ}\text{C}$)	pH	Density ($\sigma\text{-t}$)	^{137}Cs ($\text{Bq m}^{-3} \pm \text{SE}^a$)
L.1	40° 04.22 N 25° 30.08 E	4	29.3	23.0	8.86	21.00	12.6 \pm 0.2
		22	38.9	20.7	8.73	27.64	3.9 \pm 0.1
		45	38.4	17.2	8.75	28.28	2.6 \pm 0.3
		63	38.0	16.8	8.74	28.15	3.6 \pm 0.3
L.2	39° 52.06 N 25° 39.16 E	4	30.5	20.0	8.82	21.35	9.6 \pm 0.3
		22	38.5	16.9	8.81	28.32	4.2 \pm 0.3
		45	38.5	16.5	8.79	28.52	4.0 \pm 0.4
		65	38.6	15.5	8.77	28.92	3.7 \pm 0.4
L.3	39° 48.18 N 25° 39.21 E	4	32.9	20.0	8.86	23.18	8.7 \pm 0.2
		22	38.3	17.8	8.82	27.95	4.0 \pm 0.2
		45	39.0	16.5	8.81	28.91	3.4 \pm 0.3
		67	38.8	16.2	8.84	28.92	3.4 \pm 0.3

^a The uncertainties quoted for ^{137}Cs activities are 1σ values derived from counting statistics.

observed in the deep layer of stations P.1, P.2 and P.7 may be attributed to the joint action of the less charged surface water and the resuspension of ^{137}Cs from sediments (Fukai et al., 1979; Hetherington and Harvey, 1978).

Concerning the second group (P.3, P.4, P.5, P.6 – eastern part of the gulf), the trend was the opposite with surface samples having higher ^{137}Cs activity concentrations. This group corresponds to the coastal stations more directly affected by runoff. As mentioned already, P.4 and P.5 are near the Glafkos River outlet, P.3 in the northern coastline of the gulf, near the Evinos River mouth and station P.6 in the mouth of the channel to Gulf of Corinth, affected by its surface currents entering the Gulf of Patras (Papatheodorou and Kontopoulos, 1998). However, salinity, density, temperature and pH data did not show any significant variation, indicating the high mixing grade of the water caused mostly by the weather processes dominating the cold period.

Fig. 3 shows the vertical distribution of ^{137}Cs in P.1 and P.6 in February 2005. These profiles show some gradient of density and ^{137}Cs activity concentrations between the surface layer and the middle ones. More specifically, the temperature exhibits a moderate decrease paralleled by a notable increase of salinity and density with depth. Although Cs concentrations are somehow the

mirror images of salinity and density, no correlation was found between activity concentration of Cs and salinity, density or temperature. This is likely attributed to the impact from the adjacent land and/or the fresh water (decreased density and salinity in the surface) (Fig. 3).

The seasonal variation of Cs activity concentrations at three depth intervals of the water column (station P.1, central area of the gulf) can be seen in Tables 2 and 3. The measurements show a weak seasonal variation of ^{137}Cs activity concentrations at the surface and medium layers (0–47 m depth), with only slightly higher values at the surface during winter. Of more interest are the different profile patterns obtained during the two seasons (t -test applied among the seasonal values shows significant variation: t -value = -1.24 , P -value = 0.34).

One would expect higher concentrations during the summer, due to the strong evaporation, but the opposite was observed in the surface layer (Figs. 2 and 3). Besides, higher concentrations were observed at the deeper layers during the winter compared to the respective ones of summer (5.9 ± 0.1 in 80 m and 5.0 ± 0.1 Bq m^{-3} in 100 m depth during winter and 1.2 ± 0.1 Bq m^{-3} in 100 m depth during summer). This is probably attributed to the intensive runoff from the extended catchment area (Kuwara et al., 1999; Sutula et al., 2003) and the consequent impact of the nearby land to the gulf. Moreover, the fact that elevated concentrations of ^{137}Cs are observed at the deeper layers can be explained by the rapid sinking of the colder and denser water masses to the deeper layers as the thermocline is breaking down during the winter. Furthermore, the sinking water masses reaching the interface water–sediment layer, cause, as a side effect, some resuspension of the fine sea-bottom sediments, which is usually formed near the sea bottom in the semi-enclosed deep embayments (e.g. the phenomenon recorded at the adjoined Ionian Sea) (Belias et al., 2003, 2007; Scoullou et al., 1996).

4.2. The North-Eastern Aegean Sea

4.2.1. December 2005

The activity concentrations of ^{137}Cs for the surface and bottom layer of the North-Eastern Aegean Sea varied from 5.1 ± 0.5 to 7.1 ± 0.2 Bq m^{-3} and from 2.8 ± 0.4 to 4.3 ± 0.3 Bq m^{-3} , respectively. These values are of the same level with those reported by Florou et al., 2006 for the same area and significantly lower than those measured in the Black Sea (18.29 ± 2.14 Bq m^{-3}) (Gasco et al., 2002). The fact that the North-Eastern Aegean levels are between those of the Gulf of Patras and those of the Black Sea, coupled with the fact that the surface layers (which are influenced directly by the

Table 6
Estimated inventories of ^{137}Cs in the gulf of Patras and North-Eastern Aegean Sea.

Gulf of Patras			
	Date	Total Depth (m)	Cs-137 Inventory ($\text{kBq m}^{-2} \pm \text{SE}^a$)
P.1	28/9/2004	120	0.36 \pm 0.01
P.1	31/03/2005	120	0.79 \pm 0.03
P.2	30/03/2005	70	0.38 \pm 0.02
P.3	30/03/2005	63	0.25 \pm 0.03
P.4	31/03/2005	80	0.40 \pm 0.04
P.5	31/03/2005	73	0.31 \pm 0.02
P.6	30/03/2005	68	0.38 \pm 0.03
P.7	31/03/2005	50	0.34 \pm 0.03
North-Eastern Aegean Sea			
	Date	Total Depth (m)	Cs-137 Inventory (Bq m^{-2})
L.1	30/11/2005	73	0.55 \pm 0.02
L.2	29/11/2005	71	0.92 \pm 0.03
L.3	1/12/2005	67	0.38 \pm 0.03
L.1	22/06/2006	73	0.36 \pm 0.04
L.2	21/06/2006	71	0.36 \pm 0.02
L.3	23/06/2006	67	0.33 \pm 0.02

^a The uncertainties quoted for ^{137}Cs activities are 1σ values derived from counting statistics.

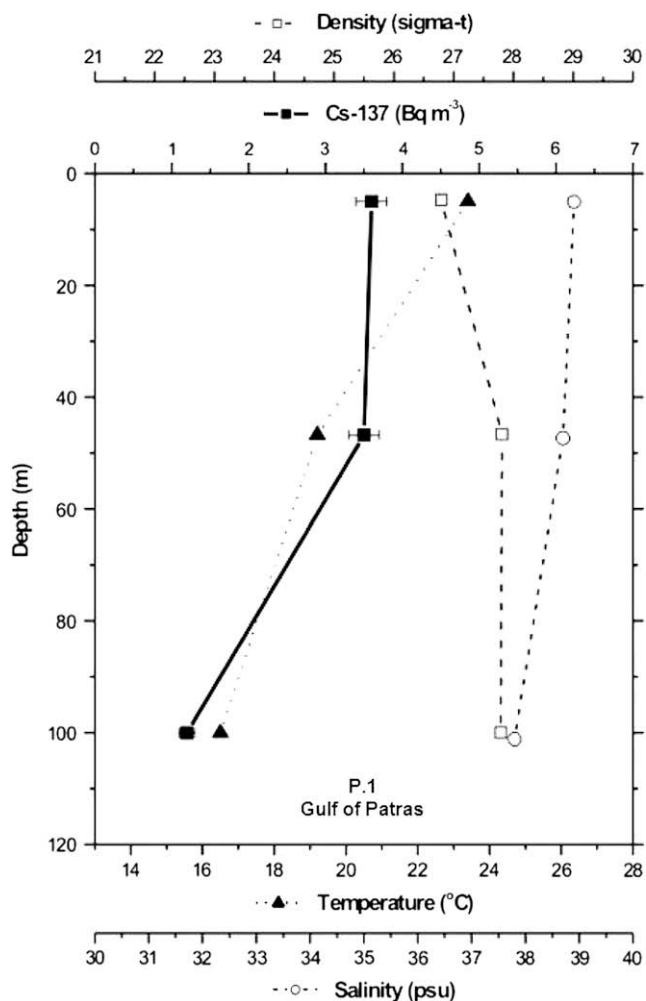


Fig. 2. Vertical profiles of salinity, density, temperature and ^{137}Cs activity concentration in P.1 of the gulf of Patras on September 2004.

Black Sea outflow to the Aegean) have higher concentrations, indicates clearly that part of ^{137}Cs found in the North-Eastern Aegean originates from the Black Sea. Nevertheless, the North-Eastern Aegean Sea is the area affected by the BSW masses (Florou and Kritidis, 1994; Florou et al., 1994) and as it has been shown, they are purified through the surface current circulation (Aarkrog et al., 1994). Thus, all the study stations are affected by BSW as the maximum activity concentrations of ^{137}Cs are found in the surface layer (down to 25 m) (Table 4). The maximum activity concentration is found at station L.1 (Fig. 1). Zodiatis (1994) has reported that “during winter periods the main volume of BSW was directed from the entrance of the Dardanelles towards the northwest of Lemnos island” (Fig. 1). As station L.1 is located in the main route of the prevailing currents, it is considered as the more affected during winter. The values observed may be considered as enhanced in relation to the ones for open seas such as $0.82\text{--}1.46\text{ Bq m}^{-3}$ in the North Atlantic Sea, $0.71\text{--}0.99\text{ Bq m}^{-3}$ in the South Atlantic Sea and $1.96 \pm 0.26\text{ Bq m}^{-3}$ in the South Ionian Sea (Gulin and Stokozov, 2005), 3.5 Bq m^{-3} in the Levantine Basin and 3.3 Bq m^{-3} in the Lybian Sea (Papucci and Delfanti, 1999).

In terms of the ratio between the surface and bottom layers activity concentrations of the 3 stations, it varies from 1.3 to 1.8, which further supports the former statement of the Black Sea surface currents influence. As one could observe, further down in the present paper, the difference between surface and bottom

layers became even more visible during the beginning of summer, when more fresh waters enter the Black Sea from the runoff of Danube, Dnieper, Dniester and other rivers discharging in the Black Sea. Consequently, the low salinity – less dense waters formed in the Black Sea outflows into the Aegean Sea through the Bosphorus and the Dardanelles Straits.

Fig. 4 shows the vertical distribution of ^{137}Cs and the physico-chemical parameters in the three sampling stations. In L.2 and L.3 the maximum values were found in 20–25 m depth, whereas in L.1 the maximum value was observed at the surface. This is again attributed to the influence of the Black Sea waters. The stratification observed in the water column was not demonstrated by the small decrease of temperature. The profile of ^{137}Cs together with that of density indicate clearly that the low salinity – low density waters of the Black Sea were the ^{137}Cs rich ones that occupied the surface and gradually mixed with the underlying waters.

In L.2, the stratification between the surface and the bottom layer was very significant. The surface layer (1–25 m depth) had considerably higher concentrations ($5.1 \pm 0.5\text{--}7.3 \pm 0.5\text{ Bq m}^{-3}$) than the bottom one ($3.7 \pm 0.5\text{--}2.8 \pm 0.4$). In cases of such stable pycnocline, as observed here, it is not uncommon that around it, higher concentrations of trace elements are observed in comparison to the surface layer, due to accumulation on the density interface of colloidal and very fine particles.

The same vertical distribution of ^{137}Cs with even higher concentrations at the 25 m depth was observed at station L.3. However, surprisingly, the vertical distribution of density did not demonstrate any considerable stratification there. The maxima in stations L.2 and L.3 were observed at 22-m depth, whereas in station L.1 it was at the surface. Moreover, the variation curve for the stations L.2 and L.3 followed similar patterns, which differentiate from that observed in the station L.1.

4.2.2. June 2006

During the summer period the situation in North-Eastern Aegean Sea changed. The surface and bottom layer activity concentrations differed considerably and ranged, respectively, between 8.7 ± 0.2 and $12.6 \pm 0.2\text{ Bq m}^{-3}$ and 3.4 ± 0.3 and $3.7 \pm 0.4\text{ Bq m}^{-3}$ (Table 5). These values compared to those found during December 2005 indicate higher activity concentrations in surface waters in the summer, following the increase of temperature, as expected, whereas the ones found at the bottom layer were similar or lower than those found in winter (Table 5). The highest ^{137}Cs concentration was found again at L.1, most probably associated to the current coming from the Black Sea mentioned previously.

Fig. 5 presents the vertical distribution of ^{137}Cs in June 2006 in the 3 stations. The three profiles followed the same pattern, with higher concentrations at the surface layer and almost constant ones in the layer between 25 and 65 m. As shown by the pycnocline curves (Fig. 5), the opposite trend in relation to ^{137}Cs activity concentration variance was observed, due to the washing out of the less saline water masses from the Black Sea. Water stratification in summer occurs due not only to salinity but also to temperature differences. Similar trends in the profile patterns of temperature and ^{137}Cs activity concentrations were observed ($R = 0.83$, $p\text{-value} = 8.3 \times 10^{-4}$). This is based to a large extent to the gradient in the surface layer since all the waters below 25-m depth are practically homogenous.

Comparing the seasonal variation of ^{137}Cs in the three sampling stations of the North-Eastern Aegean Sea, one can note that the major impact of the Black Sea water masses through the advective processes to the North Aegean Sea was recorded during summer, when evaporation along with snow melting and weathering processes (runoff of Danube, Dnieper and Dniester, snow melting and consequently higher land washout) result in higher activity concentrations of ^{137}Cs in the adjoining Black Sea (Tables 4 and 5).

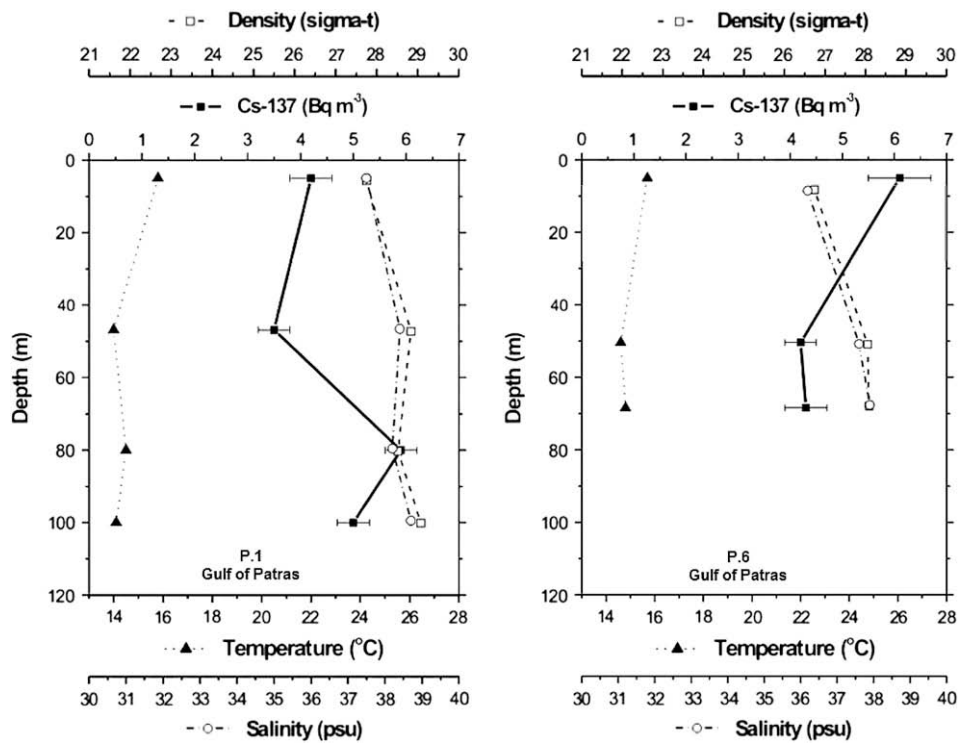


Fig. 3. Vertical distribution of salinity, density, temperature and ^{137}Cs activity concentration in P.1 and P.6 of the gulf of Patras on February 2005.

4.3. Inventories of ^{137}Cs

In order to assess the total ^{137}Cs entering the studied marine areas, the inventory was calculated on the basis that ^{137}Cs enters through the surface layer and disperses via current movements. Inventory is an important parameter for evaluating the

accumulation of anthropogenic radionuclides in the marine environment in terms of the impact sources and pathways. The inventory of radionuclides in the water column per unit area is calculated by integrating the interpolated radionuclide concentrations vertically from the sea surface to the bottom. Since the concentration data are actually discrete, the inventory is estimated

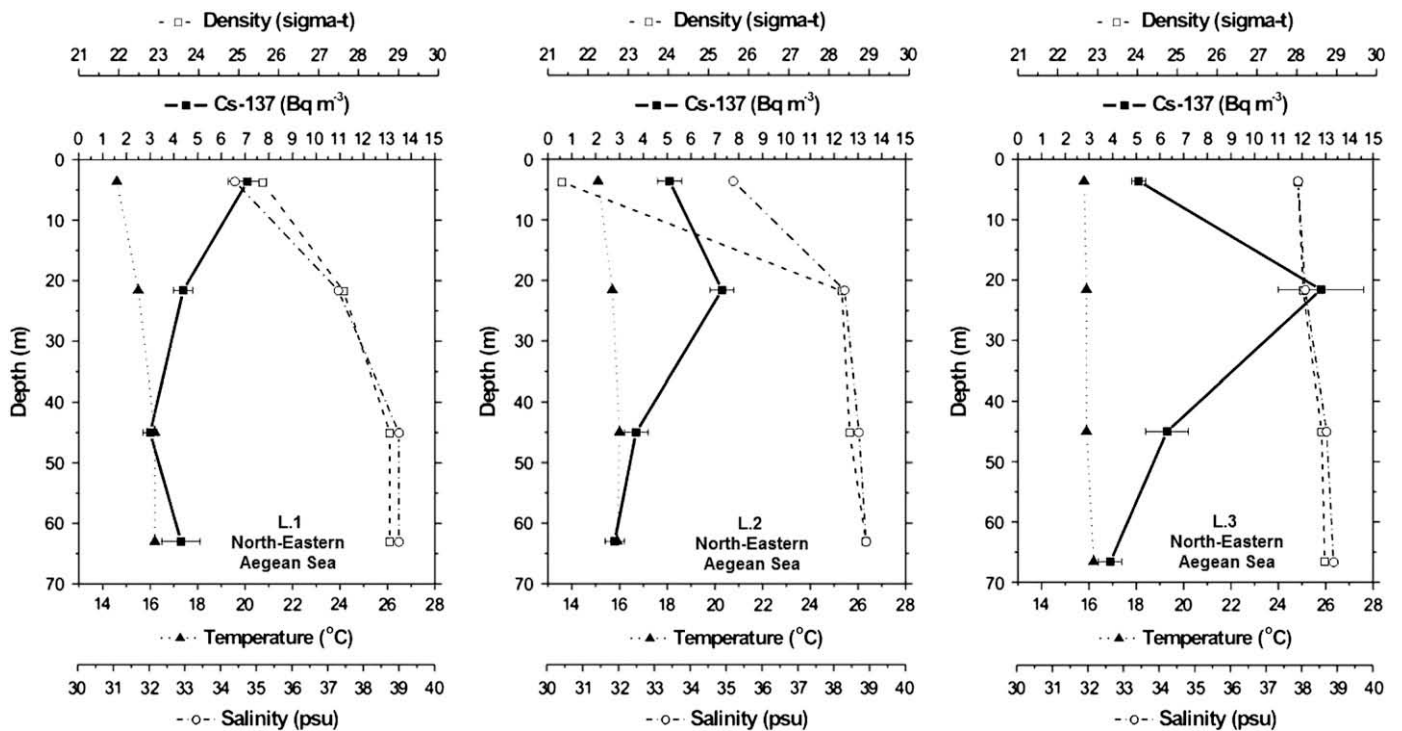


Fig. 4. Vertical distribution of salinity, density, temperature and ^{137}Cs activity concentration in the North-Eastern Aegean Sea on December 2005.

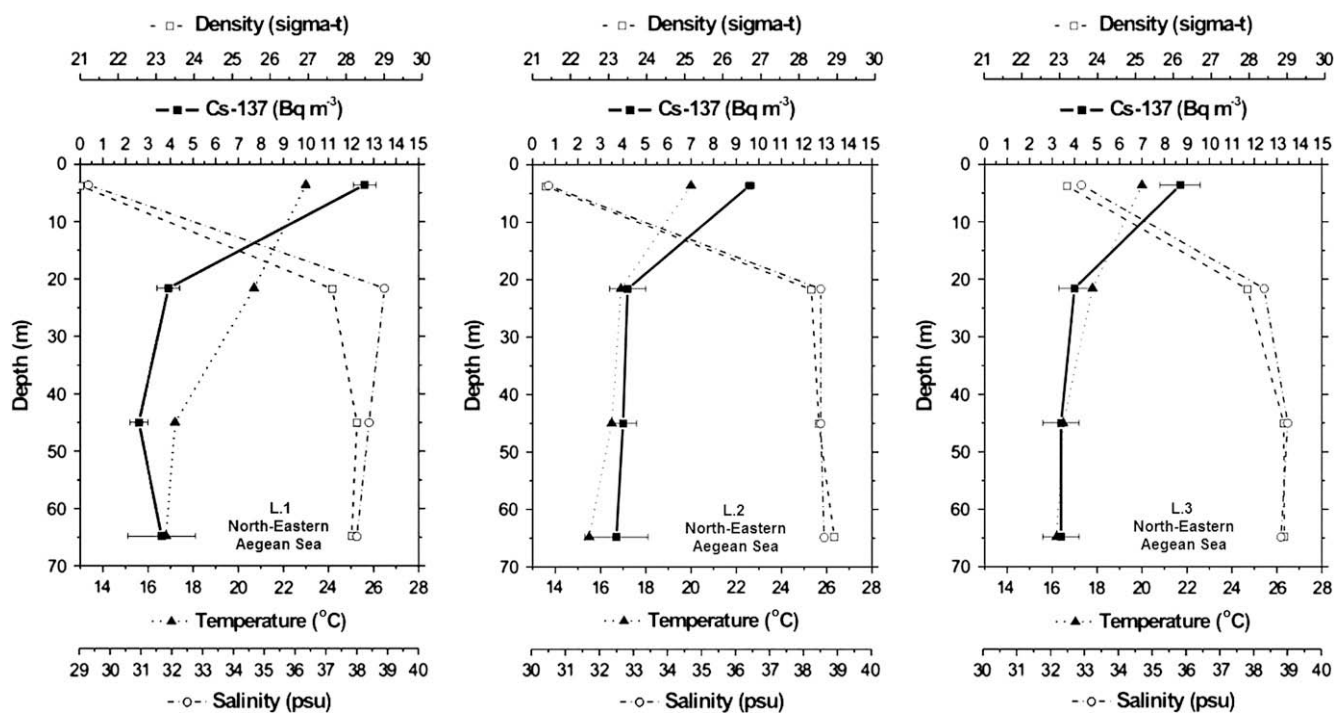


Fig. 5. Vertical distribution of salinity, density, temperature and ^{137}Cs activity concentration in the North-Eastern Aegean Sea on June 2006.

using linear interpolation (Ito et al., 2005). The results are given in Table 6.

The levels of ^{137}Cs inventory to the Gulf of Patras varied from 0.25 ± 0.03 to 0.79 ± 0.03 kBq m^{-2} . These values are considered as low if one compares them with $2\text{--}5$ kBq m^{-2} $^{137}\text{Cs} + ^{134}\text{Cs}$ (2/3 of them corresponding to ^{137}Cs deposition) which resulted after the Chernobyl NPP accident (Kritidis and Florou, 1990).

In the North-Eastern Aegean Sea the respective values varied from 0.33 ± 0.02 to 0.92 ± 0.03 kBq m^{-2} . This area was the most affected from the Chernobyl fallout in the Eastern Mediterranean (Aarkrog et al., 1994; Florou et al., 2003), with a mean value of total deposition of $^{137}\text{Cs} + ^{134}\text{Cs}$ to be $10\text{--}20$ kBq m^{-2} and the 2/3 of them corresponding to ^{137}Cs deposition (Kritidis and Florou, 1990).

Nevertheless, the dispersion and behavior of ^{137}Cs in the marine environment, especially because it is a conservative radionuclide, is strongly connected with the transfer and transit times in relation to the sources and pathways expressed parametrically by the "ecological half-life". As the estimated values of the inventory are low (if one estimates the initial Chernobyl deposition of $10\text{--}20$ ($\times 2/3$) kBq m^{-2} corrected to the dates of the recent values based on the physical half-life only), one may conclude that the ecological half-life of ^{137}Cs in the studied area is considerably low. Florou et al. (2001) reported an ecological half-life of 8.2 y for the North-Eastern Aegean Sea, whereas the respective one for the Black Sea was 6 y (Egorov et al., 1999).

Using the ecological half life of 8.2 y to calculate the present inventory of ^{137}Cs , the resulting deposition varied between 1 and 2 kBq m^{-2} . The estimated value in this study was 0.92 kBq m^{-2} , which is considerably lower than the above range. Taking into account the 20 year elapsed from Chernobyl accident in relation to the sinking time of ^{137}Cs in the open sea ($16\text{--}82$ cm y^{-1} , Cai et al., 1997), one can assume that the ^{137}Cs estimated inventory is composed mainly by the water-borne ^{137}Cs from the Black Sea, whereas the Chernobyl inventory has been removed more or less to the bottom.

5. Conclusions

The surface concentrations of ^{137}Cs at the end of summer (September 2004) in the Gulf of Patras ranged between 2.2 ± 0.3 and 3.6 ± 0.1 Bq m^{-3} , whereas at the end of winter (February 2005) it ranged from 4.0 ± 0.4 to 6.7 ± 1.1 Bq m^{-3} . These values are comparable to those measured in other areas of the Mediterranean Sea and to those measured previously in the Greek marine environment.

Regarding the vertical profile of ^{137}Cs in the water column of the Gulf of Patras, high concentrations are observed at the 0–50 m layer, decreasing with the depth. In the center of the gulf, higher ^{137}Cs concentrations are found in the bottom during late winter attributed to the surface water sinking to the bottom, as a result of the pycnocline breakdown enhanced most probably by resuspension of fine sea-bottom sediments. Considering the seasonal variation of ^{137}Cs in the Gulf of Patras for all the selected sampling points, higher values were found in winter, indicating ^{137}Cs inputs through runoff from the adjacent catchment areas and some enrichment from resuspended sediments.

The surface concentrations of Cs in the North-Eastern Aegean Sea during winter (December 2005) ranged between 5.1 ± 0.6 and 7.1 ± 0.2 Bq m^{-3} , whereas it was 8.7 ± 0.2 to 12.6 ± 0.2 Bq m^{-3} during the warm period. The values reported in this paper are comparable to those reported earlier, although a general tendency for decrease is noticed.

Vertical profiles in the North-Eastern Aegean Sea followed the same pattern, with higher concentrations in the upper layer (0–20 m depth) and lower ones in greater depths. This phenomenon can be explained by the Black Sea water outflows that mainly affect the surface layer of the study area. Considering the spatial distribution of ^{137}Cs in the Greek marine environment (integrated ^{137}Cs levels on the inventory basis), higher values were found in the North-Eastern Aegean Sea than in the central part of the Gulf of Patras confirming that the Black Sea waters are the main source of ^{137}Cs in the Greek marine environment.

In general, ^{137}Cs concentrations obtained from this study are comparable to those recorded in the literature for other parts of the Mediterranean. In comparison to those reported previously for the Greek marine environment, the present values are considerably lower, similar to those of the pre-Chernobyl period.

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