

# RADIOACTIVITY IN THE ROMANIAN BLACK SEA SECTOR ONE DECADE AFTER CHERNOBYL



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## 1. INTRODUCTION

Radioactivity monitoring in the marine environment was imposed by the increasing development of nuclear energetics and its world-wide use in many different activities [1].

Both natural and artificial radioactivity play an important role in marine ecology and human health; in this respect three major facts continue to prevail in Romania: the fallout, the presence of the Danube river, and the future nuclear energy production by using of five CANDU reactors under construction at Cernavoda.

Spatial and temporal monitoring of marine radioactivity along the Romanian Black Sea shore has been systematically performed by the Romanian Marine Research Institute (RMRI), in close co-operation with the Research Laboratory of Environmental Radioactivity (RLER) belonging to the Institute of Meteorology and Hydrology (until 1990) and to the Research Institute for Environmental Engineering (REIE) afterwards, since 1991.

## 2. MATERIAL AND METHODS

Marine emerged and submerged sediments, coastal and offshore sea water, macroalgae, invertebrates and fish off the Danube mouths and/or along the coast have been monitored for natural and artificial radioactivity by means of alpha and beta gross measurements and gamma spectrometry.

Materials and methods used for these continuous investigations have been described in previous publications [1, 2, 3, 4, 5].

Concentrations of various radionuclides in abiotic and biotic samples, environmental distribution coefficients (Kds) and concentration factors (CFs), as well as experimentally-derived CFs in marine biota as radioecological bioindicators have been assessed and stored for a national data base and for international uses.

## 3. RESULTS

The alpha gross radioactivity level was always below the lower detection limit (LDL) for all samples excepting the green seaweed *Bryopsis plumosa*.

The beta level was below LDL for sea water (salt), beach sediment (sand) and molluscs (shell). The averages for other samples were as follows: 883 Bq.kg<sup>-1</sup> sediment (dry) off the Danube mouths, 152 Bq.kg<sup>-1</sup> fresh weight (f.w.) algae, 84 Bq.kg<sup>-1</sup> f.w. molluscs (soft part), 118 Bq.kg<sup>-1</sup> f.w. fish (comparable to 113 Bq.kg<sup>-1</sup> f.w. for fish originating in the FAO 34 (Mauritania) and FAO 47 (Namibia) zones of the Atlantic Ocean. Again, *B. plumosa* is an exception with values of 10 Bq.kg<sup>-1</sup> f.w. [6].

The gamma spectrometry analyses have been performed at RLER until 1992 and at RMRI ever since. Low background, high resolution equipment has been used [7]. Data quality control was ensured by taking part in international intercomparison runs organized by the IAEA (SD-A-1, IAEA-300, IAEA-306, IAEA-307, IAEA-315).

Since the last atmospheric nuclear test in the northern hemisphere (1980), during 1982-1984 fission radionuclide concentrations continuously decreased in the environment, including the Black Sea, until they were below detection limits. This trend continued till 1985, when the lowest levels of radioactive pollution were registered in Romania. No radionuclides originating from the releases of nuclear facilities were found during this time [3].

Before the major nuclear accident at Chernobyl, Ukraine, on April 26, 1986, the gamma activity of the samples was given mainly by the natural gamma emitters (K-40 and those in the U-Ra and Th series). Among the artificial gamma emitting radionuclides originating in the previous nuclear atmospheric tests, Cs-137 was the most significant. The evolution of its concentration, showing a decrease towards detection limits in 1985 - early 1986, was marked by the mentioned accident.

Gamma radioactivity monitoring along the Romanian marine coast in 1986 indicated an evident increase of radionuclide concentrations, as compared with the period 1983-1985, directly related to the Chernobyl accident.

After the Chernobyl accident, interest in radioecological research of the Black Sea increased in Romania, as it did in a number of other countries [8]; also other fission as well as activation products appeared in the marine environmental samples, such as: Zr-95/Nb-95, Ru-103, Ru-106, Ag-110m, Sb-125, I-131, Cs-134, Ba-140/La-140, Ce-141, Ce-144.

In the marine environment K-40 continued to prevail permanently in all components. Ac-228 and Ra-226 were also determined as indicators for the uranium-radium and thorium series. Co-60, Ru-106, Cs-137 and Ce-144 were constantly analyzed since 1983, and Ag-110m and Cs-134 starting with May, 1986.

In the predanubian area the isotopic ratios Ru-106/Ru-103 and Cs-137/Cs-134, as well as distribution coefficients (Kds) for submerged sediment and sea water from 10 and 20 m depth were estimated (Table I).

The isotopic ratio values of Cs-137/Cs-134 in sediments and sea water demonstrated that the Chernobyl accident was a source of radioactive contamination along the Romanian shore.

Given their importance, special attention was paid to Cs-134 and Cs-137, for which international organizations established maximum permissible limits for food products following the Chernobyl accident. Romanian studies thus particularly focused on computing the concentration of Cs-137 for sediment and sea water in the predanubian sector as well as along the western Black Sea coast; the evolution of sediment and sea water contamination off the Danube mouths is illustrated by concentrations of Cs-137 immediately after the accident (Fig. 1).

In the Romanian Black Sea sector, the maximum values of Cs-137 in sea water and fish were found in 1986, macrophytes and molluscs in 1988, and in sediment in 1987 (Table II). In all samples Cs-137, persisted from 1987 until 1995.

The concentration abilities of various marine biota for artificial post - Chernobyl radionuclides was previously commented [2, 3, 4, 5, 7, 9, 10, 11, 12]. Thus, for example, pelagic fish species showed the following distribution of Cs-137 concentrations: *Sprattus sprattus phalericus* > *Merlangius merlangus euxinus* > *Trachurus mediterraneus ponticus* > *Engraulis encrasicolus ponticus*, due to distinctive feeding manners. These species are the most important in the present Romanian marine fishing and directly or indirectly involved in human consumption.

Concentration factors were also computed for Cs-137, as radionuclide of major interest following the Chernobyl accident [10, 11, 12] (Fig. 2). The maximum values have been obtained for the sediment at the Danube mouths. The additional radioactivity from the river contributions has increased the local impact of the accident [11, 13].

With time the content of artificial gamma radionuclides continuously decreased in all components (sediment, sea water, biota) compared to 1986. E.g., Cs-137, decreased comparatively with its maximum concentration in 1986 up to 63% during 1987-1988. This decrease was more gradual during 1990-1991 than it was before. The relatively slow decrease of Cs-137 concentrations in sediment compared to sea water confirmed the ability of sediments to concentrate radionuclides.

Although decreasing considerably between 1986-1988 Cs-137 levels in the NW Black Sea have not yet reached the pre-Chernobyl values [5]. A Cs-137 residence time of about 15 years has been estimated from a simple model. The evolution of Cs-137 concentrations in sea food following the

Table I: Isotopic ratios and distribution coefficients ( $K_d$ s) of some gamma radionuclides in sediment and sea water of the predanubian sector of the Black Sea in 1986 (May, 1)

Transect	Sampling depth (m)	Sediment		Sea water	$K_d$ ( $l.Kg^{-1}$ )	
		$\frac{106Ru}{103Ru}$	$\frac{137Cs}{134Cs}$	$\frac{137Cs}{134Cs}$	40K	137Cs
Sulina	10 <sup>*</sup>	0.3±0.1	2.1±0.1	2.5±1.1	100±11	1826±375
	20 <sup>*</sup>	0.2±0.1	2.4±0.1	1.8±1.8	135±16	2136±875
Sf.Gheorghe	10 <sup>*</sup>	0.3±0.1	2.1±0.1	1.9±0.5	107±9	985±91
	20 <sup>*</sup>	0.2±0.1	3.2±0.3	1.7±0.5	133±11	879±184
Portita	10 <sup>**</sup>	0.1±0.1	2.3±0.1	2.2±1.0	50±6	299±46
	20 <sup>**</sup>	=	=	1.0±0.3	144±26	259±8
Buhaz	10 <sup>***</sup>	0.4±0.3	2.0±0.2	1.1±0.8	27±3	675±338
	20 <sup>***</sup>	=	2.6±0.3	1.5±0.3	47±4	70±4

Sediment: \* muddy, \*\* muddy with mollusc shells, \*\*\* sandy.

Table II: Maximum Cs-137 concentrations in environmental samples from the Romanian sector of the Black Sea (1983-1995)

	Emerged sediment Bq/kg dry	Submerged sediment Bq/kg dry	Sea water Bq/l	Macro-phytes Bq/kg fw	Molluscs Bq/kg fw	Fish Bq/kg fw
1983	-	-	0.04	2.2	-	2.6
1984	4.8	-	0.03	3	-	3.3
1985	3.7	-	0.03	-	-	-
1986	29.6	-	0.85	-	4.4	44.4
1987	18.9	247.0	0.13	4.6	3.2	11.0
1988	11.5	25.2	0.10	7.1	3.3	4.3
1989	15.5	-	0.09	5.2	2.8	5.1
1990	13.3	55.0	0.07	3.4	1.3	4.0
1991	21.5	24.2	0.08	1.9	1.5	3.9
1992	10.7	-	0.06	1.4	1.2	3.5
1993	8.2	189	0.12	2.8	2.3	4.2
1994	-	150	0.07	-	-	-

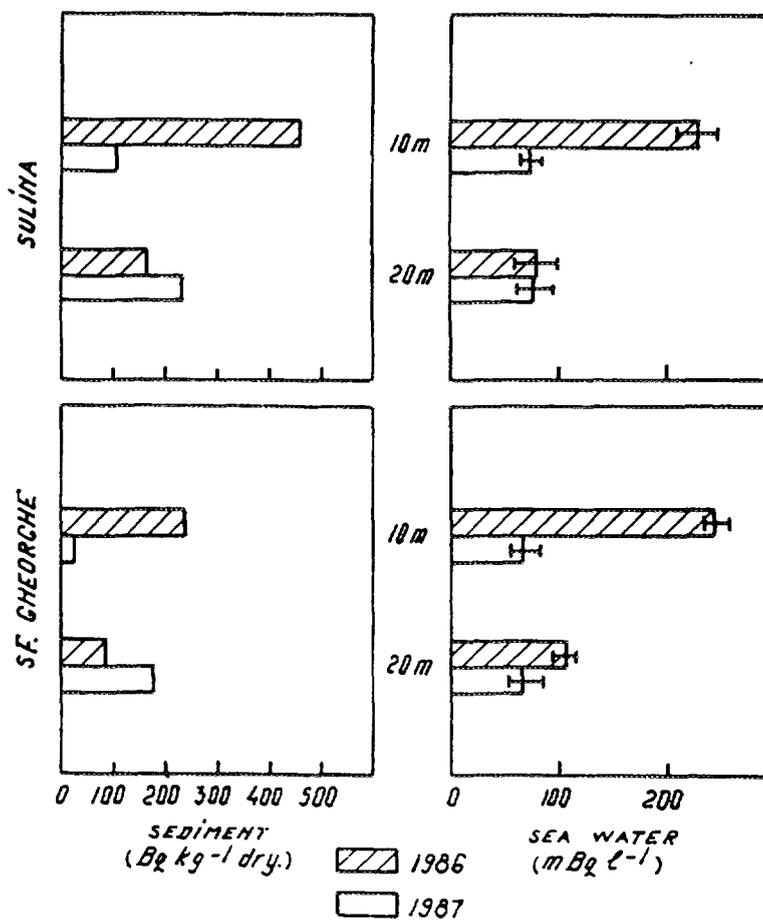


Fig.1 Cs-137 concentrations in sediment and seawater off the predanubian coast during 1986-1987.

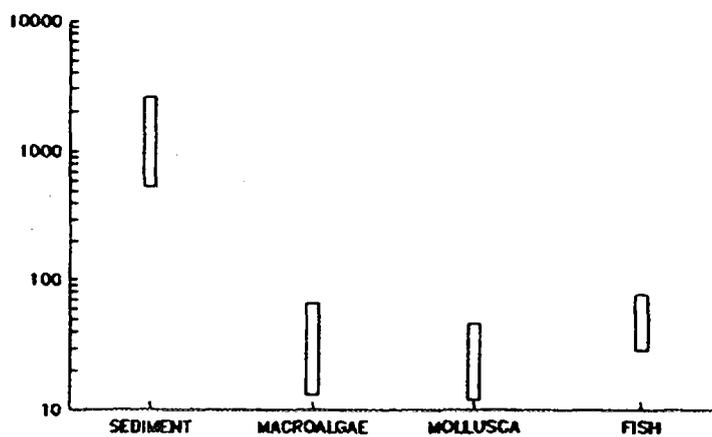


Fig.2 CFs intervals for marine components.

Chernobyl accident is an important component of the Romanian monitoring program; the highest Cs-137 and Cs-134 concentrations in edible marine biota (fish, molluscs), in the Romanian sector, allowed for food by the United National Food and Agriculture Organization (FAO) in 1987 and the following years did not exceed the "action levels" or maximum admissible limits.

The data on gamma radioactivity of the marine littoral ecosystem permitted estimates of annual individual external and internal radiation doses before and after the Chernobyl accident (Fig.3) [14, 15, 16].

The systematic indigenous research activities and results in the field of marine radioactivity have been partially reevaluated within the IAEA research contract No. 4805/ROM/RB/1987-92 "Monitoring of marine water, sediment and biota radioactivity in samples from the Romanian sector of the Black Sea by means of gamma spectrometry" [8]. Certain results obtained under the above mentioned research contract were used through the IAEA research agreement No. 302-K4-ROM/1989-92 "Dose assessment from marine radioactivity in the Romanian sector of the Black Sea" for the IAEA/MEL CRP "Sources of radioactivity in the marine environment and their relative contributions to overall dose assessment from marine radioactivity - MARDOS" [4].

More recent gamma radioactivity measurements along the Romanian shore have been used for the computation of the radiological exposure of the European Community to radioactivity in the framework of the MARINA-MED project [9].

Marine radioactivity data will also complete the international Co-operative Marine Science Program in the Black Sea (CoMSBlack), launched as first regional oceanographic program after 1990, by its working group on marine radiochemistry and radioecology [17].

A Romanian data base for radionuclides in the north-western Black Sea has been developed to include the results of the monitoring program performed by RMRI and RLER beginning with 1984 [18]. This data base was also used for ICSEM/GIRMED and IAEA/GLOMARD marine radioactivity data bases [5].

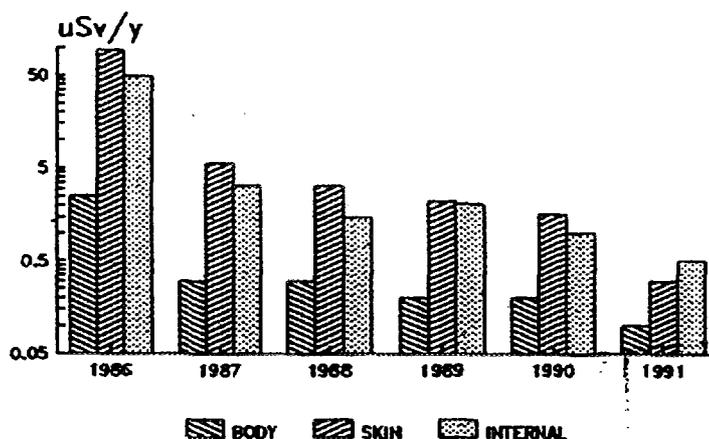


Fig.3 Maximum total doses for external and internal exposure.

#### 4. CONCLUSIONS

-The occasional or continuous monitoring of alpha, beta and gamma radioactivity in the Romanian sector of the North-Western Black Sea has enabled the establishing of reference values for all categories of marine components.

-The Chernobyl nuclear accident caused increases of artificial radioactivity for the abiotic and biotic components, with maximum values in 1986 and 1987.

-Nowadays there is a tendency of coming back to the environment condition before 1986.

-The Danube mouths produce an additional impact zone owing to the Danube contribution.

-The sediment, algae, molluscs and fish significantly concentrate artificial radionuclides, in accordance with environmental concentrations.

-There is a remarkable remanence of Cs-137 in the substratum, as a witness of recent human nuclear history (1987-1994).

-The level of maximum individual exposure to the marine external and internal irradiation is inferior to the national and international standards for 1986-1991.

-National (RADMAR) and international (GIRMED, GLOMARD) data bases have been made up for various uses.

-The knowledge on the actual marine environment condition is required for the characterization of the impact of some future nuclear activities and objectives near the Romanian Black Sea coast (Cemavoda, Danube-Black Sea canal, Constantza-Agigea).

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