

RADIOACTIVITY OF BOTTOM SEDIMENTS SAMPLED IN 1996 FROM THE ROMANIAN SECTOR OF THE DANUBE RIVER

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Abstract

Man-made and natural radioactivity of 23 bottom sediments sampled in spring 1996 along the Danube river from its entrance in Romania to the Black Sea and on the Black Sea coast was investigated by gamma-ray spectrometry. Specific activities of ¹³⁴Cs, ¹³⁷Cs, ²²⁶Ra, ²²⁸Ra and ⁴⁰K were determined. The highest natural and artificial radioactivity was found at Orsova upstream to the Portile de Fier (Iron Gates) dam and in the Black Sea off Razelm Lagoon at Gura Portita. A comparison of the radioactivity levels of Danube bottom sediments collected from the same sites over the last few years was made.

Key-words: radionuclides, Danube delta,

Introduction

The aim of this work was to investigate the presence of man-made and natural radionuclides of the U-Ra and Th families in the bottom sediments along the Danube river in Romania and on the Black Sea Romanian coast, during spring 1996. The sediment samples were collected by the National Institute of Meteorology and Hydrology, Bucharest from the principal cross-sections of the Danube river and from the same stations in the Black Sea which had been studied before 1977 by the Polytechnical Institute of Bucharest [1]. For radioecological purposes, the survey of the Danube river and Black Sea radioactivity was generally carried out during specific hydrological periods, i.e. in spring, summer and autumn seasons at high and relatively low flow rates, under conditions of high and low water dilution.

More recently, data on ¹³⁴Cs and ¹³⁷Cs radioactivity in sediments collected from the same areas during 1993, and on the ¹³⁴Cs, ¹³⁷Cs, ²³⁸U, ²³²Th and ⁴⁰K radioactivity in sediments collected in the summer and autumn of 1994 and 1995 from 20 and 21 locations, respectively, have been presented in previous papers [2, 3]. Twelve of the sediments collected during 1994 were analyzed for ²¹⁰Po radioactivity [4]. The annual averages of ¹³⁷Cs specific activities in sediments from some of these locations during 1986-1990 [5], and a ¹³⁷Cs vertical profile in some selected lake sediments of Danube Delta during 1994 [6, 7] have also been determined.

Experimental

The bottom sediment samples were collected from the principal cross sections along the Danube river, from the entrance in Romania to its mouths (delta included), and from the Romanian coast of the Black Sea, during spring 1996 (22 samples). An additional sample was collected at Cernavoda (km 300) from about 300 m along the Danube-Black Sea man-made channel in the vicinity of the CANDU Nuclear Power Plant.

The sediment samples of 1.5-2 kg each have been collected from depth of 0-15 cm using a stainless steel cup and then placed in plastic bags. Associated *in situ* hydrological measurements were made according to the procedures of the National Institute of Meteorology and Hydrology of Bucharest. Sediments were dried in an electric oven at 110°C and then homogenized in an agate mortar. About 100 g of each sample were enclosed in a standard polyethylene box (7.3 cm diameter) and kept inside for one month to permit ²²⁶Ra (U-Ra radioactive family) to establish radioactive equilibrium with its decay products.

The gamma ray measurements were carried out by means of a high resolution, low background PC multichannel spectrometer, using a HPGe EG&G ORTEC detector of 30% efficiency and 2.1 keV resolution for the ⁶⁰Co 1332.5 keV line. As standards, the reference materials IAEA-135 (Radionuclides in Irish Sea sediment) and IAEA-306 (Radionuclides in Baltic Sea sediment) with certified radioactive concentrations were used. Counting times ranged between 14-24 hours.

Results and Discussion

Table 1 presents the ¹³⁴Cs, ¹³⁷Cs, ²²⁶Ra, ²²⁸Ra and ⁴⁰K specific activities (Bq/kg dry) in Romanian Danube and Black Sea coastal sediments. In this table, the sampling sites in the Danube delta start with Ceatal Sfântu Gheorghe (km 63), i.e. the split of the Danube into the Sulina and Sfântu Gheorghe branches; Sтамбул Vechi (km 4) is

located on the Chilia branch while Sulina (km 4.6) and Sfântu Gheorghe (km 8) are located on the Sulina and Sfântu Gheorghe branches, respectively; on the Black Sea coast the sampling sites are noted from north to south with (1), (2) and (3).

Table 1. Specific activity of the bottom sediments sampled in spring 1996 along the Danube river from the Romanian border to the Black Sea and on the Black Sea coast [Bq/kg dry].

Samplig site / Date	Distance* (km)	¹³⁴ Cs	¹³⁷ Cs	²²⁶ Ra	²²⁸ Ra	⁴⁰ K
Bacius / 18.05.96	1072.4	—	11.7 ± 0.6	31.7 ± 1.2	32.0 ± 1.6	396 ± 14
Svinia / 19.05.96	995	1.0 ± 0.3	56.4 ± 2.2	43.0 ± 2.1	38.1 ± 1.9	501 ± 18
Orsova / 19.05.96	957	1.7 ± 0.2	79 ± 2.0	44.1 ± 2.2	41.6 ± 2.1	548 ± 17
Iganasi / 21.05.96	889	—	6.1 ± 0.4	22.0 ± 1.1	16.7 ± 1.3	576 ± 17
Calafat / 21.05.96	787.9	—	2.4 ± 0.4	16.7 ± 0.8	16.5 ± 1.3	392 ± 14
Bechet (upstream) / 22.05.96	709	—	1.4 ± 0.3	17.8 ± 0.8	13.7 ± 1.0	400 ± 12
Bechet (downstream) / 22.05.96	678.7	—	1.7 ± 0.3	12.1 ± 0.7	12.4 ± 0.9	366 ± 10
Turnu Magurele / 23.05.96	596.3	—	0.8 ± 0.3	11.1 ± 0.7	11.1 ± 0.8	521 ± 15
Giurgiu / 25.05.96	493	—	0.8 ± 0.3	12.6 ± 1.1	12.7 ± 1.6	401 ± 11
Chicru Călarasi / 27.05.96	379.6	—	0.9 ± 0.4	13.2 ± 1.1	12.4 ± 1.4	393 ± 9
Cernavoda (upstream) / 27.05.96	303	—	1.4 ± 0.3	14.4 ± 0.9	14.9 ± 1.2	373 ± 9
Cernavoda (channel to Black Sea) / 27.05.96	300	1.2 ± 0.3	34.6 ± 0.9	47.1 ± 2.1	47.0 ± 1.7	595 ± 15
Cernavoda (downstream) / 27.05.96	297	—	1.0 ± 0.3	16.9 ± 0.8	15.8 ± 1.0	353 ± 10
Vadu Oii / 29.05.96	238	—	0.7 ± 0.3	16.9 ± 1.1	15.8 ± 1.2	392 ± 10
Braila / 05.06.96	167	—	0.3 ± 0.3	17.6 ± 1.1	19.3 ± 1.1	221 ± 13
Ceatal Izmail / 07.06.96	80.9	—	0.5 ± 0.3	21.6 ± 1.0	18.0 ± 1.1	274 ± 8
Ceatal Sfântu Gheorghe / 14.06.96	63	—	1.0 ± 0.3	17.6 ± 0.8	14.5 ± 1.0	326 ± 10
Sfântu Gheorghe / 14.06.96	8	—	0.3 ± 0.3	18.8 ± 0.9	11.0 ± 0.8	286 ± 9
Sulina / 11.06.96	4.6	—	0.8 ± 0.3	14.3 ± 0.8	12.8 ± 0.9	276 ± 8
Sтамбул Vechi / 10.06.96	4	—	1.3 ± 0.3	43.7 ± 2.2	40.5 ± 2.5	305 ± 9
East Sulina / 27.04.96	(1)	0.6 ± 0.2	16.6 ± 0.7	24.1 ± 1.1	25.9 ± 1.6	504 ± 12
Gura Portita / 25.04.96	(2)	1.4 ± 0.2	86.1 ± 3.0	32.6 ± 1.6	35.8 ± 1.3	638 ± 17
East Constanta / 06.05.96	(3)	—	6.9 ± 0.5	28.7 ± 1.0	34.0 ± 1.2	389 ± 10

* Distance between sampling site and the Danube mouths to the Black Sea: Sulina is considered "km 0", except for the sites placed on the Chilia and Sfântu Gheorghe branches.

(1) In front of Sulina Port, 6.5 km offshore and 26.5 m depth in the Black Sea;

(2) In front of Razelm lagoon, south of the Danube delta, 13.7 km offshore and 21.0 m depth in the Black Sea;

(3) In front of Constanta, 8.6 km offshore and 31 m depth in the Black Sea.

In order to calculate the activities of ²²⁶Ra and ²²⁸Ra, the gamma peaks of 352 keV (²¹⁴Pb) and 609 keV (²¹⁴Bi) for ²²⁶Ra, and 583 keV (²⁰⁸Tl) and 911 keV (²²⁸Ac) for ²²⁸Ra were taken into consideration.

The following conclusions drawn from the data are summarized below:

1) In all samples analyzed, only the fission products ¹³⁴Cs (T_{1/2} = 2.06y) and ¹³⁷Cs (T_{1/2} = 30.17y) were identified as man-made radionuclides. ¹³⁷Cs was present in all the samples with specific activity levels between 0.5 ± 0.3 and 86.1 ± 3.0 Bq/kg dry, while ¹³⁴Cs was measured in the range of 0.6 ± 0.2 to 1.7 ± 0.2 Bq/kg dry only at 5 sampling stations where higher activities of ¹³⁷Cs were recorded. The activity