ON A MONITORING METHOD FOR RADIOACTIVE POLLUTION IN A FLOW CROSS SECTION ROMANIAN SECTOR OF DANUBE RIVER

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Abstract: Some years ago, a research has been carried out in view to determine the radioactive pollution in a cross section of the Danube river, taking into account (considering) the specific behavior of this river, i.e. the Romanian sector of the Danube is not a regularized river as against others. The following fission and induced radio nuclides have been chosen: ^{144}Ce - ^{144}Pr, ^{125}Sb, ^{106}Ru - ^{106}Rh, ^{137}Cs, ^{54}Mn, ^{110}Ag, ^{65}Zn, ^{60}Co and ^{22}Na, found at the Mile 34, that marks the beginning of the Danube Delta, where the Danube river branches into Sulina and Saint George. The origin of these radio nuclides is from fallout, no special injection being effected for our study.

- The distribution of radio nuclides on carriers (filtered water, suspended matter and sediments) has been discussed.
- "Significative verticals" have been proposed in order to reduce the vertical number for radioactive measurements.
- The experimental results obtained are in agreement with some mathematical analytical relations, concerning the solid discharge and this fact permits us to design a physico-mathematical model of longitudinal dispersion with radioactive pollutants.

Comparison is made with the natural and artificial radio nuclides found at the entrance of Danube river in Romania, as well as in the biota during February-March 2000, from Moldova Veche till Giurgiu.

Keywords: Danube river, "significative verticals", radioactivity, sediments, water, heavy metals, fish.

UBER EINE METHODE DER UBER WACHUNG DER RADIOAKTIVEN VERSCHMUTZUNG IN EINEM TEIL DER DONAU, RUMANISCHES SEKTOR

Zusammenfassung: Vor Jahren, wurden Untersuchungen gemacht um die Radioaktivitat zu bestimmen, in einen Teil der Donau, die spezifische Benehmung dieses Fluβ berucksichtigend, daβ der rumanische Sektor der Donau nicht wie andere Flusse regularisiert ist. Folgende Isotopen die aus Fission oder durch zufugen von Neutronen wurden auserwahet: ^{144}Ce - ^{144}Pr, ^{125}Sb, ^{106}Ru - ^{106}Rh, ^{137}Cs, ^{54}Mn, ^{110}Ag, ^{65}Zn, ^{60}Co und ^{22}Na, identifiziert (gefunden) bei Mila 34, Anfang der Donau Delta, das heiβt wo sich die Donau in den Armen Sulina und Sf. Gheorghe im "fallout" (die atmosphrische einfallen, daβ heiβt daβ es kein besonderer Zufug fur dieses Studium gemacht wurde).

- Es wurde die Distribution der Isotopen auf den Trager studiert (filtriertes Wasser, Suspensionen und Sedimenten).
- Es wurde die Bennenung "beteutende Vertikalen" vorgeschlagen, um die Anzahl der Vertikalen fur radioaktive Messungen zu mindem.
- Die erhaltene experimentelle Ergebnisse, uber stimmen mit den mathematischen analytischen Verhaltnisse uber den den festen Durchfluβ, und dieses erlaubt uns einen physisch-matematishen Modell der langsgerichteten Zerstreuung der radioaktiven Umweltverschnutrer zu banen.

Es wurde ein Vergbich zwischen den radioisotops gefunden beim Eingang der Donau in Rumanien, im Wasser und die Bios in Februar - Marz 2000 von Moldova Veche bis Giurgiu.

Schluesselworte: der Fluβ Donau, "bedeutende Vertikale", Radioaktivitat, sedimenten, Wasser, schwere Metale, Fisch.

Introduction

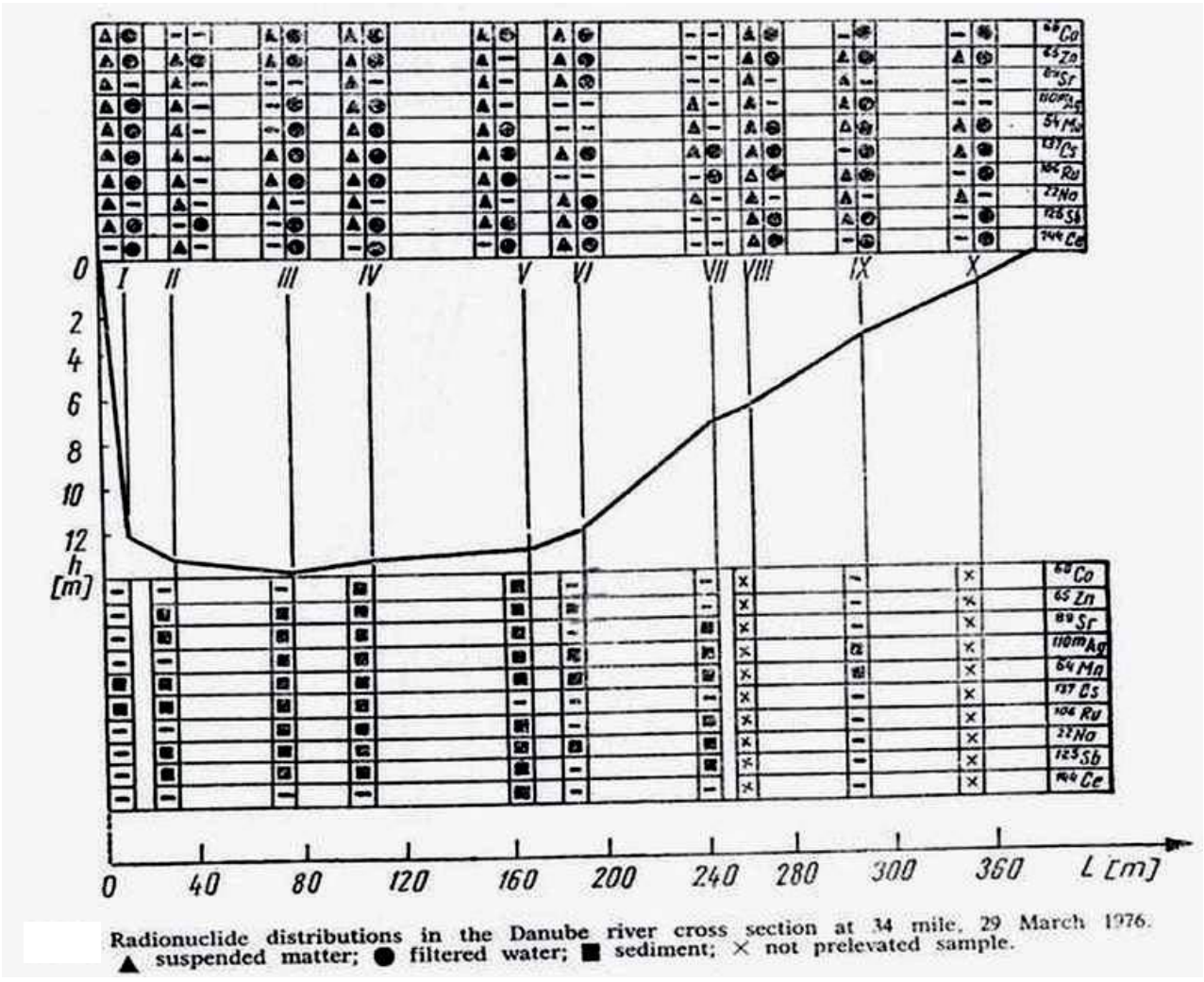
This research was performed under Contract RB/1077 (Georgescu, I.,I., 1977) between the International Atomic Energy, Vienna and the Polytechnical Institute of Bucharest, Romania.
Elaboration of an accurate method for monitoring radioactive pollution of flowing water is a difficult problem from two points of view: radioactive measurements and selected radio nuclide distribution in suspended matter and in water. In addition turbidity repartition in flow cross sections, as well as accurate determination of suspended and sedimented solid debit still posse problems for hydrologists. In hydrographic basins where antierosion and regulation of rivers have been effected, solid debit in suspension can be determined only by one prelevated sample (Hinrich. H., 1972). However, this is not applicable to the Danube river of the Romanian hydrographic basin, that explaines a necessary laborious studies. It must be outlined another investigations were performed after the gold mining events in N - W Romania, in January 2000 (Georgescu, I.,I., et al, 2001).

Materials and Method

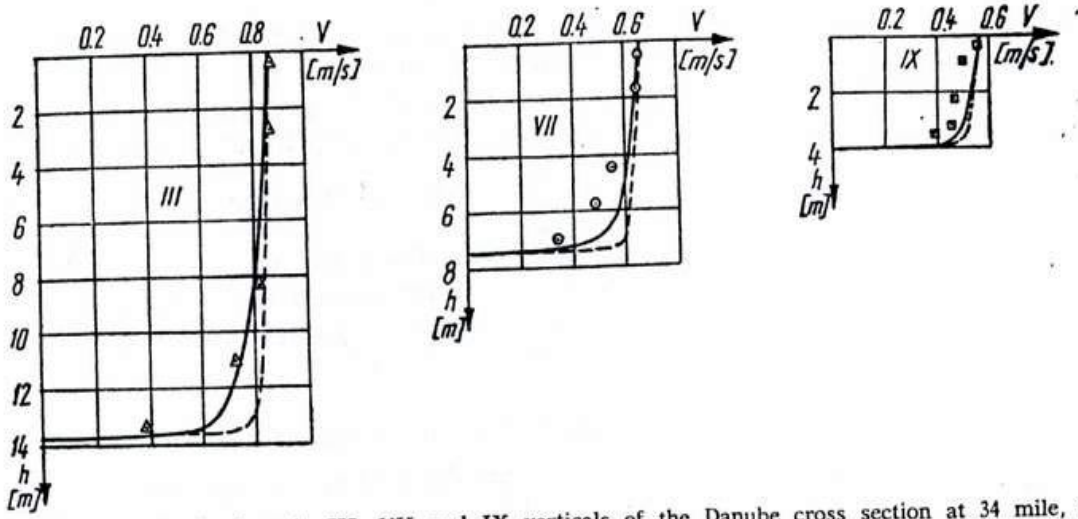
The experimental studies were carried out at the bifurcation of the Danube into Sulina branch and the Saint (Sf.) Gheorghe branch 34 miles of Tulcea. Sample prelevations were performed by the hydrological standards. The number of vertical cross section to be studied, nine to ten verticals.

- Current velocity at five points of each vertical, i.e., at surface, at depth of 0.2 h, 0.6 h and 0.8 h as well as at about 30 cm from the bottom of the upper bed of the river.
- Water sampling with suspended matter at the below mentioned points of odd verticals, designed by I, III, V, VII, IX and only at one point at 0.6 h in even verticals.
- Radiometric measurements were performed on surface water, sampled from each vertical cross section and on river bed sediments prelevated along the same vertical. A sextant was used to determines cross section verticals. A "Jestowski" type hydraulic mill was employed to determine water current velocity; river depth was measured with a cable. One liter bathometer was used to collect water for measurements of suspended matter at different depths. Turbidity water samples was determined by photocolorimetry.

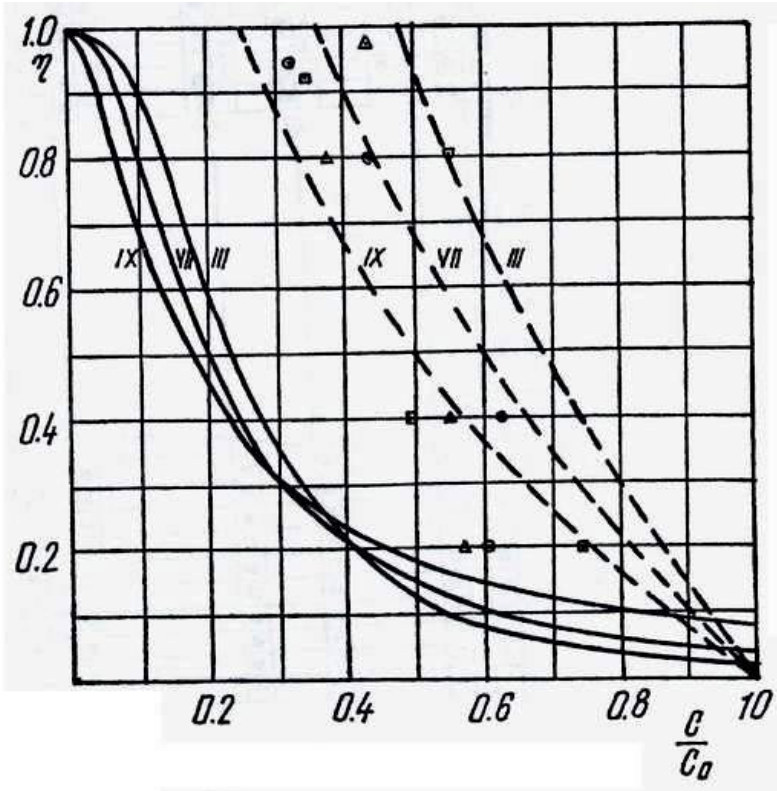
The radio nuclides were identified by a multichannel analyzer SA-42 Intertechnique coupled to a high - resolution Ge (Li) detector of 2.7 keV for ^{60}Co gamma rays. Figures 1 - 5 (Georgescu,I.,I. et al, 1977, 1978) represents the experimental data. After the gold mining event in N - W Romania, January 2000 it was interesting to investigate the pollution of the Danube river at km 1073, the river mouth in Romania, downstream to Giurgiu (km 493), two months after the gold mining accident. Atomic Absorption Spectrometry (AAS) (Varian 250 PLUS spectrophotometer) was to determine heavy metal concentration in water samples (Table 2) (Georgescu, I.,I. et al, 2001) and by γ -ray spectrometry the radioactivity of the sediments (Table 3) (Georgescu, I.,I. et al, 2001).



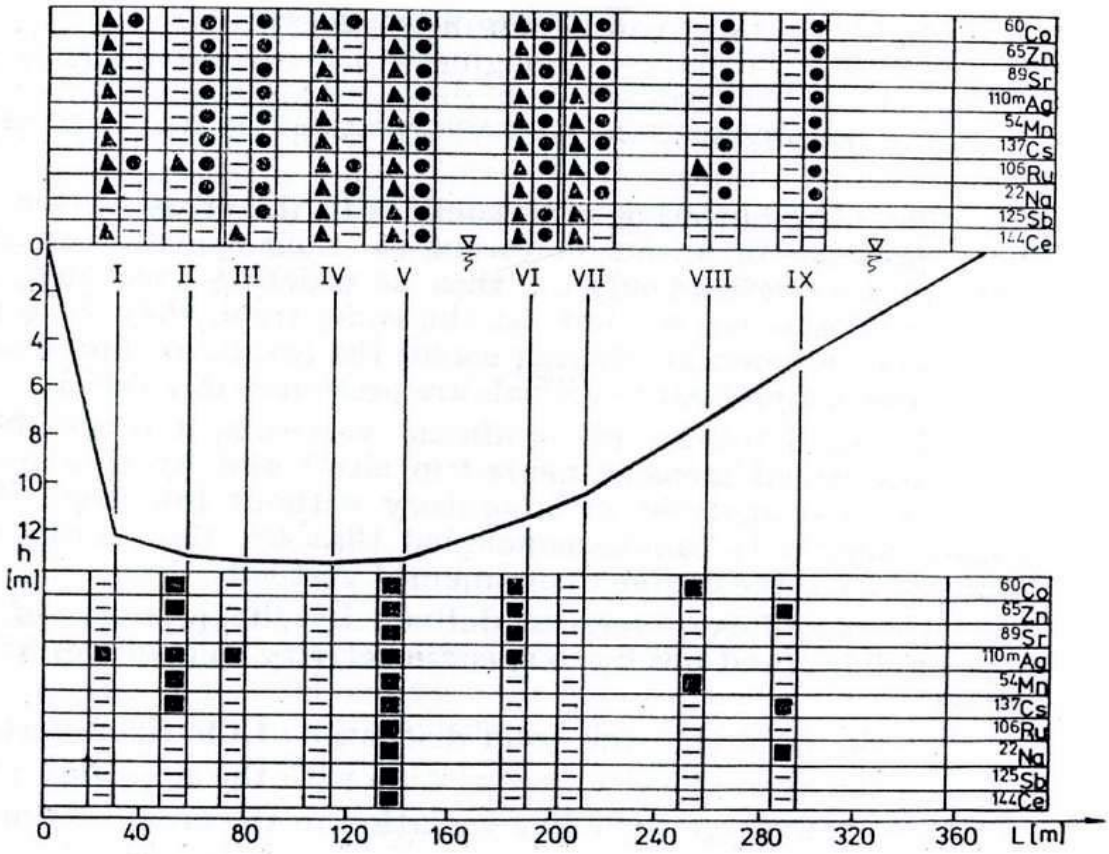
Radionuclide distributions in the Danube river cross section at 34 mile, 29 March 1976. ▲ suspended matter; ● filtered water; ■ sediment; × not prelevated sample.



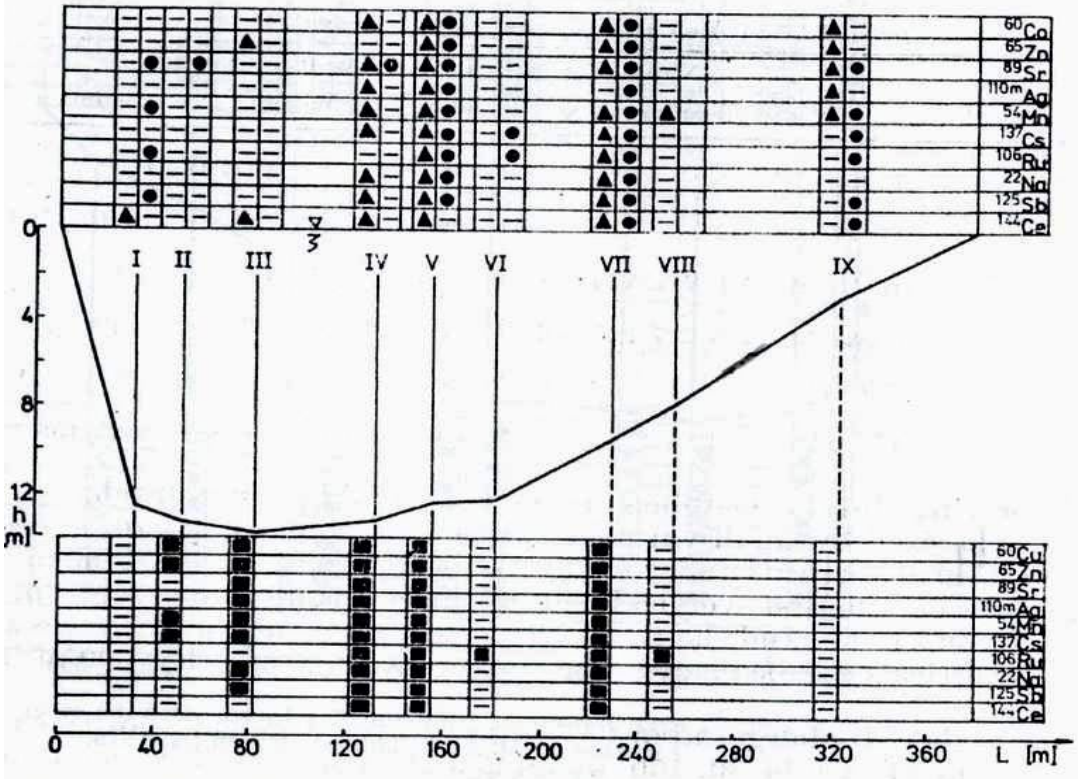
Velocity distributions in III, VII and IX verticals of the Danube cross section at 34 mile, 29 March 1976. —— eq. (7); — — — eq. (9); △ ⊙ □ experimental values.



Turbidity distributions in III, VII and IX verticals of the Danube river cross section at 34 mile, 29 March 1976. —— eq. (8); — — — eq. (10); △ ⊙ □ experimental values.



— Distribution of radionuclides in the Danube river cross section at Mile 34 (Tulcea), July 14, 1976.
▲ Suspended matter :
● Filtered water ; ■ Sediments.



— Distribution of radionuclides in the Danube river cross section at Mile 34 (Tulcea), October 8, 1976.
■ Suspended matter ;
● Filtered water ; ■ Sediments.

Table 1. Summarized activity of I - X verticals at 34 miles cross section of the Danube river on March 29, 1976

Radio nuclide	*Suspended matter pCi / 10 liters*	*Filtered water p Ci / 10 liters*	*Bed-load sediment * p Ci / 8 grams*
^{144}Ce - ^{144}Pr	23.61	205.82	65.50
^{125}Sb	6.17	11.14	21.67
^{22}Na	0.663	absent	0.474
^{106}Ru-^{106}Rh	1.62	8.5	11.15
^{137}Cs	0.185	31.03	3.20
^{54}Mn	0.276	0.41	1.234
^{110m}Ag	0.420	0.282	2.356
^{89}Sr	0.195	0.133	1.01
^{65}Zn	0.568	1.60	2.50
^{60}Co	0.10	0.414	0.26
Total activity	33.807	259.309	109.354

** It was not possible to prelevate samples from VIII th and X th verticals, due to the rocky facies.*

Activity of filtered water, p Ci / l (1 p Ci = 0.037 Bq)

^{22}Na, ^{110m}Ag ,and ^{89}Sr, lack of all samples;

^{144}Ce 0.33 - 6.70 till 86.8 - 89, present in all samples;

^{125}Sb 0.35 - 0.87, 3.85, lack of two samples;

^{106}Ru 0.16 - 5.28 present in all samples;

^{137}Cs 1.5 - 1.6 present in all samples;

^{54}Mn 0.02 - 0.10 present in four samples, lack in two samples;

^{65}Zn 0.04 - 0.37 present in all samples;

^{60}Co 0.02 present in three samples, lack of three samples.

Activity of bottom sediments, pCi / g dry weight

^{22}Na, ^{125}Sb, ^{89}Sr, lack of all samples;

^{144}Ce 0.03 - 1.06 in four samples, lack of two samples;

^{106}Ru 0.02 - 0.14 in four samples, lack of two samples;

^{137}Cs 0.01 - 0.02 in two samples, lack of four samples;

^{54}Mn 0.02 in one samples, lack of five samples;

^{110m}Ag 0.01 - 0.02 in three samples, lack of three samples;

^{65}Zn 0.012 in one sample, lack of five samples;

^{60}Co 0.01 - 0.12 in two samples, lack of four samples.

Table 2. Elemental concentration in Danube water, determined by AAS (μg L^{-1}).

Sample site /Date	*Cd*	*Cr*	*Cu*	*Fe*	*Mn*	*Ni*	*Pb*	*Zn*
km 1031/16.02.00	4	5	34	0	147	0	16	65
km 1073/23.03.00	6	22	11	22	43	2	43	18
km 1073/29.03.00	10	0	10	0	1	17	50	10
km 633/29.03.00	9	14	20	14	65	14	75	30
km 493/28.03.00	14	0	9	0	0	9	64	24
km 493/29.03.00	4	15	8	57	0	20	57	15
km 493/29.03.00	4	43	11	43	11	7	57	7
km 493/30.03.00	22	22	17	24	24	4	22	24

AAS - Atomic Absorption Spectra.
Average concentration values in water samples (in mg L^{-1}): 19.162 (Cl^-), and 0.017 (CN^-)

Table 3. Activity concentrations in Danube sediments, by γ-ray spectrometry (Bq kg^{-1}).

Radio-Nuclide	*km 1073/ 20.03.00*	*km 1072.4/ 24.03.00*	*km 633/ 29.03.00*	*km 629.5/ 29.03.00*	*km 493/ 28.03.00*	*km 493/ 29.03.00*	*km 493/ 30.03.00*
^{137}Cs	4.4 ±0.6	1.8 ±0.3	1.4 ±0.2	2.4 ±0.5	0.6 ±0.2	1.6 ±0.3	1.6 ±0.3
^{226}Ra	35.6 ±1.6	9.6 ±1.1	8.5 ±0.8	13.3 ±1.3	12.2 ±1.1	13.5 ±1.5	12.6 ±1.0
^{228}Ra	33.4 ±2.7	11.6 ±0.9	11.9 ±0.7	16.7 ±1.0	15.5 ±1.0	15.0 ±1.2	13.6 ±0.9
^{40}K	463 ±23	270 ±11	366 ±12	390 ±25	350 ±11	425 ±13	400 ± 14

Conclusions

1. It has been concluded that the activity of fission and neutron induced nuclides transported by Danube river into the Black Sea, is low.

2. Ten fission and neutron induced radio nuclides were investigated in each type of carrier: ^{144}Ce, ^{125}Sb, ^{22}Na, ^{106}Ru ,^{137}Cs, ^{54}Mn, ^{110m}Ag, ^{89}Sr, ^{65}Zn, ^{60}Co.

3. The essential points of the proposed method are: choice of a sufficient number of verticals and velocity and turbidity determinations by radioactive measurements of water and bed load sediments. The method is a suitable and simple tool for determining radioactive pollution.

4. Two months after the gold mining events at Baia Mare (N - W Romania) in January 2000, no pollution with heavy metals and man made radioactivity was observed in the samples collected along the Danube river from river from Moldova Noua (km 1073, the river mouth in Romania) downstream to Giurgiu (km 493).

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