DANUBE SEDIMENT RADIOACTIVITY INVESTIGATION

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Abstract
About 50 sediment samples taken from locations evenly distributed along the Vojvodina part of Danube have been collected. The radionuclide content of the samples was determined by means of low-level, high-resolution gamma-spectroscopy.
Beside the members of the natural radioactive chains of ^{238}U and ^{232}Th and the natural ^{40}K, in all samples the Chernobyl origin ^{137}Cs was detected. The data obtained will be compared with data collected in four years after the Chernobyl accident, and the elimination rate of ^{137}Cs from the river ecosystem will be discussed.

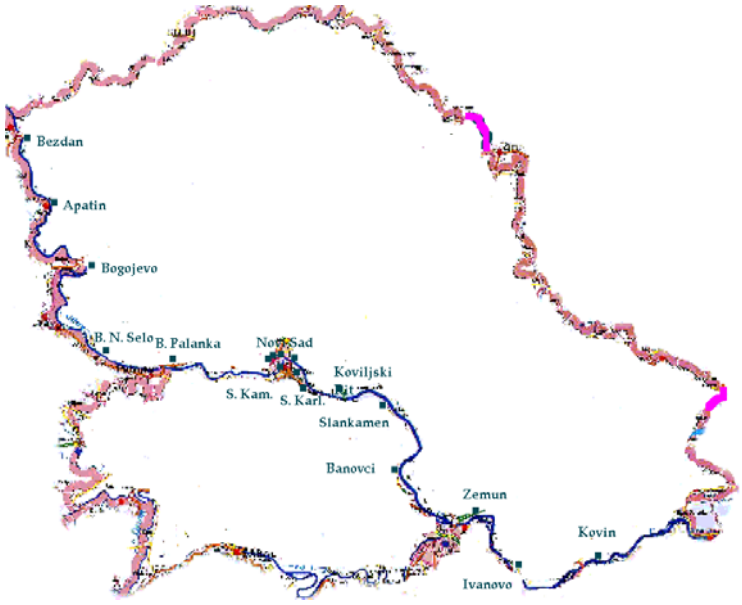
Introduction
There are several components affecting on the radioactivity of the River Danube. The water of the river cools the reactors of the nuclear power plants on Danube (1). Therefore, the energy production in nuclear power plants is always accompanied by pollution of the river with radionuclides. The artificial phosphate fertilizers (2), (3) with high uranium concentration may cause a gradual increase of the uranium series activity concentration in soil that gets to the river. The radioisotopes from atmospheric nuclear weapon tests and the additional fallout from the accidents like Chernobyl can be also the cause of pollution. The fact that the Paks Nuclear Power Plant is near to Yugoslav border is also a reason to investigate the Danube sediment radioactivity to extract the information on the influence of the power plant operation on water pollution.
The river sediment is considered as a durable and reliable registrator of the river pollution by radionuclides. In the sediment the water pollution components are deposited. A long-term radioactive pollution is accumulated in the sediment and when the sediment is mixed up the radionuclides reenter into ecological nutrition chains.
The dislocation of the radionuclides is very complicated process but following the logical sense the higher concentration of radionuclides can be expected at locations where river flow is slowed down.
Unfortunately there are no systematical data of sediment radioactivity measurements on the Yugoslavian reach of Danube in Vojvodina, especially after 1990.

Sampling technique and sample preparation
The sediment samples were taken from the surface layer at three locations close to the riverbank from the left or the right side. The sediment samples were taken at 54 locations at the Yugoslavian reach of the River Danube in Vojvodina. The locations are presented in *Table 1*, and marked at the map at *Fig. 1*. The wet sample mass was about 1 *kg*.
The sediment samples were prepared by drying at 105^{o}C to a constant mass and after homogenization transferred to sample holders, cylindrical containers (67 mm diameter and 62 mm height), and sealed. The samples were kept hermetically closed for about 40 days in order to reach the equilibrium due to radon ($T_{1/2}$ [^{222}Rn]=3.8 days) emanation. When keeping the samples in sealed sample holders for about ten half-lives the ^{222}Rn equilibrium will be restored. The measurements where performed after this period.

Table 1. Sampling locations

Location	*Bank*	*Number of samples*
Bezdán	left	3
Apatin	left	3
Gombos (Bogojevo)	left	3
Bačko Novo Selo	left	3
Bácspalánka (Bačka Palanka)	left	3
Novi Sad (Újvidék) beaches	right	12
Novi Sad, Sremska Kamenica	left	3
Novi Sad, Petrovaradin	left	3
Karlóca (Sremski Karlovci)	right	3
Koviljski rit	left	3
Slankamen	right	3
Stari Banovci	right	3
Zimony (Zemun)	right	3
Sándoregyháza (Ivanovo)	left	3
Kovin	left	3
	total:	54

Figure 1. The sampling locations (marked by circles)



Experimental techniques and method of measurements

The radionuclide content of the samples was measured by means of the reversed electrode "GMX" type HPGe spectrometer made by ORTEC. The nominal efficiency of the detector is 32% and the resolution is 1.9 *keV*. This detector has thin dead layer on outer surface and beryllium entrance window, thus enables the detection of gamma rays below 100 *keV* with excellent efficiency.

The detector was operated inside the 25 *cm* thick iron shield made from pre II World War cast iron. As compared with led shields the iron shields have definite advantages in the low energy region due to the absence of the lead *x*-rays (4), (5). The detector was calibrated by means of reference radioactive materials in cylindrical geometry (NBS Standard Reference Material 4350B). The matrix effects were taken into account by means of a computer code. The typical time of measurements of the samples was about 50 *ks*.

A modified version of the SAMPO program was used to process the spectra, in such a way that, besides the identified gamma-lines, it always presents spectral intensities of 20 selected nuclides. The measurement uncertainties were presented at the 95% confidence level, what means that the probability for obtaining a result laying outside the presented limits in a repeated measurement of the

same sample is less than 5%. The list of selected radionuclides, their half-lives, main production reactions and target abundance are presented in *Table 2*.

The experimental results

The mean values of the average activity concentrations of the radionuclides presented in *Table 2.*
The mean values of the average activity concentrations of the radionuclides measured on sediment samples from the Yugoslav-Hungarian border in 1985, 1990 are presented also in *Table 2*. and compared with the mean values of the present measurements.

Table 2. Average values of activity concentrations of radionuclides

	Bezdan-Mohacs (48 samples) 1985.		Bezdan-Mohacs (48 samples) 1990.		Yugoslavian reach of Danube in Vojvodina (54 samples) 2002.	
	Specific activity $\bar{A}_s$ [Bq/kg]	Number of samp.*	Specific activity $\bar{A}_s$ [Bq/kg]	Number of samp.*	Specific activity $\bar{A}_s$ [Bq/kg]	Number of samp.*
^{75}Se	<0.6	0	<0.9	0		
^{144}Ce	4.7±2.3	7	<7	0	2.5±1.0	3
^{141}Ce	-	-	-	-	0.7±0.3	3
^{125}Sb	0.7±0.5	2	2.9±2.4	5	1.00±0.14	2
^{7}Be	20.0±14.0	20	120.0±150.0	16	13±10	30
^{103}Ru	0.7±0.6	1	<2.9	0	0.25±0.20	1
^{134}Cs	1.7±1.3	22	22.0±14.0	46	0.48±0.16	7
^{124}Sb	<1.2	0	1.5±0.9	1	0.38±0.12	3
^{106}Ru	3.0±1.0	5	6.2±0.3	3	3.7±0.5	3
^{110m}Ag	1.3±1.1	2	0.9±0.5	6	0.38±0.15	4
^{137}Cs	8.0±5.0	1	150.0±100.0	48	28±23	54
^{95}Zr	1.4±0.5	10	2.7±0.6	2	0.8±0.6	1
^{95}Nb	1.1±0.6	18	0.6±0.2	2	0.41±0.14	8
^{58}Co	0.8±0.6	1	<1.7	0	0.40±0.21	2
^{160}Tb	-	-	-	-	1.6±1.1	2
^{60}Co	0.9±0.5	3	0.8±0.6	1	0.37±0.15	6
^{238}U	-	-	36±15	11	42±12	53
^{226}Ra	30±7	48	38±14	48	32±8	54
^{232}Th	32±9	48	37±15	48	36±9	54
^{40}K	490±110	48	530±180	48	445±88	54

*the number of samples in which the radionuclide was detected

Radionuclides that are not present in the selected list were not detected.

Dicussion and conclusions

From the overview of Table 2. we can at first conclude that beside the natural ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K in all samples ^{137}Cs is present. This radionuclide originates mostly (6), (7) from the accident of the nuclear power plant 'Lenin' in Chernobyl in 1986. Due to the long half-life of this radionuclide of (30.174±0.034)*y*, it was relocated, washed out and redistributed, but it is still present in the Vojvodina reach of Danube. The large difference between the minimum and maximum ^{137}Cs activity concentrations on the same location show typical features of a man-made contaminant. The average value of activity concentrations on each location does not show any dependence on the distance form the border or spatial correlation.

Some parts of the Danube, like Šodroš at Novi Sad have very low average ^{137}Cs activity concentrations (Ā=3 *Bq/kg*) while at Ivanovo mean activity concentration is Ā=70*Bq/kg*. In fact the ^{137}Cs concentration varies significantly in samples from the same location. Thus at Ivanovo the concentration varies from 6 to 140. The lowest ^{137}Cs activity concentration in one sample (1.8±0.4) *Bq/kg* is registered at Šodroš in Novi Sad.

The activity concentrations of ^{134}Cs and ^{137}Cs are (31.3±2.7) *Bq/kg* and (206±26) Bq/kg respectively (8) for Bezdan at 1990 while the activity concentrations of ^{134}Cs and ^{137}Cs are (0.6±0.3) *Bq/kg* and (140±6) *Bq/kg* respectively for Ivanovo at 2002. According to the predicted elimination half-life,

$T_{ec}(^{137}Cs)$= (238±52) *d,* (9) the activity concentration of ^{137}Cs should be $<2\cdot10^{-3}$ *Bq/k*g. Our measurements of sediment radioactivity in 2001/2002 show that this prediction is later violated and the elimination rate of the radioisotopes is much slower. The ratio of the activity concentrations of ^{134}Cs and ^{137}Cs in 1990 at Bezdan was 0.152±0.023 while in 2002 at Ivanovo this ratio is $(4.7\pm0.7)\cdot10^{-3}$. Using the listed half-lives of this isotopes and their activity ratio from 1990, their activity ratio in 2002 can be calculated (if there are still of Chernobyl origin). The value calculated $(4.3\pm2.2)\cdot10^{-3}$ is shows that after the Chernobyl accident there was no further, major contamination with these radionuclides.

As the first result of this comparison we can note that some reactor corrosion products (^{110m}Ag, ^{58}Co, ^{60}Co) are detected in bigger number of samples in 2002 than before. This can be explained by the improved detection limits of the new spectroscopy system, but the fingerprints of the reactors cooled by the waters of Danube are clear.

The natural radionuclide levels especially of ^{238}U and ^{226}Ra did not changed statistically significantly. This means that the employment of phosphate fertilizers containing this radionuclides and the NATO bombardment did not affected the radioactivity of the river. However, local contaminations from fertilizing cannot be excluded, due to the poor sampling net.

The cosmic origin ^{7}Be content of the samples is just the consequence of the natural fall-out process. Generally the radioactivity level of Danube sediment is now similar to the pre Chernobyl era, and via the known (10) concentration factors of ^{137}Cs in fish and sediment we can estimate that the health risk from the Danube radioactivity is not significant. However, this optimistic point view must be justified by more detailed investigations involving research teams from both Hungary and Yugoslavia.

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