

## RADIOACTIVITY AND MEASUREMENTS OF SEDIMENT DEPOSITION RATE OF THE DRENOVA RESERVOIR (B&H)

by

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This work presents the first estimate of the radioactivity and sediment deposition rate of the Drenova reservoir. The radioactivity and sedimentation rate were computed applying the <sup>210</sup>Pb and <sup>137</sup>Cs methods. Samples of <sup>210</sup>Pb and <sup>137</sup>Cs were taken from four boreholes drilled in the Drenova reservoir in June 2010. Vertical distribution of the natural and artificial radionuclides in four boreholes was examined using a gamma spectrometry measurement with HpGe detectors, Gamma X type (10 keV-3 MeV). Activities ranging from 122-8 Bq/kg were found for <sup>210</sup>Pb, and from 140-0.8 Bq/kg for <sup>137</sup>Cs. The sedimentation rate in the Drenova reservoir varied from 1.96 to 2.90 cm per year for unsupported <sup>210</sup>Pb and 0.47 to 5.33 cm per year for <sup>137</sup>Cs.

*Key words: radionuclide, <sup>210</sup>Pb, <sup>137</sup>Cs, sedimentation rate, Drenova reservoir*

### INTRODUCTION

Investigating the process of recent sedimentation in a reservoir is very significant as the sedimentation rate is one of the most important parameters of the dynamics of a reservoir. The sediments deposited in reservoirs represent an important piece of data, which may be used to reveal the watershed erosion history, but also to validate various types of erosion [1].

The radionuclide <sup>137</sup>Cs (30.2 year half-life) is of a particular environmental concern because it is a by-product of atomic energy production and nuclear weapons tests performed during the 1950s and 1960s. <sup>137</sup>Cs released into the atmosphere becomes strongly absorbed by clay minerals and tightly bound to the organic soil particles, and appears to migrate in soils rather slowly. After its deposition, its redistribution is mainly associated with physical processes in soil, such as soil erosion. Particularly significant was atomic weapons testing during 1963. Because <sup>137</sup>Cs is strongly associated with minerals the 1963 weapons testing peak has been preserved in most sediments. In

Europe, deposition of <sup>137</sup>Cs originating from the Chernobyl reactor accident in 1986 provides a further time marker that may be even more pronounced than the 1963 weapons peak <sup>137</sup>Cs has been widely used as a valuable tracer in soil erosion and sediment delivery in catchment for the past 50 years [2-5]. Recently, many investigators use <sup>137</sup>Cs technique for testing distributed soil erosion and sediment delivery models, estimated sedimentation rate in reservoir, and for measurements of catchment sediment budgets [6].

The use of natural and artificial radionuclides for estimation of sediment deposition rates have been reported by several authors to be taking place in many countries. However, this is for the first time this method has been used on the territory of the Republic of Srpska, Bosnia and Herzegovina. In that respect, we show the first results of examinations of the Drenova reservoir from the aspect of radioactivity, as well as the results of deposition rate.

### STUDY AREA

The Drenova reservoir is located at 44°52'13"N, and 17°31'13"E, with elevation of 170 m above the sea

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level. The intended purpose of the structure built in 1978 was flood waves attenuation. However, due to the increase in water demand of the municipality of Prnjavor, the accumulation has also been used for water supply. The design reservoir storage capacity is  $9.40 \cdot 10^6 \text{ m}^3$ , with  $0.5 \cdot 10^6 \text{ m}^3$  area designed for deposition of sediment. The Drenova reservoir was created by a damming of the Vijaka river, with a catchment area of  $68.30 \text{ km}^2$ . The average annual flow rate downstream the dam is  $1.72 \text{ m}^3/\text{s}$ , while the average flow rate of the Vijaka river is  $3.29 \text{ m}^3/\text{s}$ . Other tributaries to the storage reservoir are smaller, but only the river Drenovica is significant in terms of siltation of the reservoir. The composition and age of the geological structure of the catchment of the Drenova reservoir are of following nature: Jurassic formation, Diabase-chert formations, Upper Cretaceous formations, and Neogene sediments. Based on the analysis of surface representation of certain lithologic members, it was found that 73% of the basin accumulations are Neogene sediments, which is significant in terms of erosion processes and material import in the accumulation, and thus the siltation of the reservoir is of special importance [7].

## METHOD-SAMPLING AND MEASURING

### Sampling and measuring

Samples for determination of  $^{137}\text{Cs}$  were collected from four boreholes excavated in the Drenova reservoir in June 2010. The first samples were taken from a borehole  $B_1$   $17^\circ 37' 22,5'' \text{ E } 44^\circ 48' 31,5'' \text{ N}$ , the second from a  $B_2$   $17^\circ 38' 03,0'' \text{ E } 44^\circ 48' 51,7'' \text{ N}$ , the third from a  $B_3$   $17^\circ 38' 24,4'' \text{ E } 44^\circ 48' 50,0'' \text{ N}$ , and the fourth samples were taken from a  $B_4$   $17^\circ 38' 23,3'' \text{ E } 44^\circ 48' 40,9'' \text{ N}$ . The samples were taken on a 20 cm profile depth, at distances ranging from 6.8 to 7.6 m in the case of the borehole  $B_1$ , *i. e.* from 2. 4 to 5. 1 m in the case of the borehole  $B_2$ , *i. e.* from 6. 2 do 7 m in the case of the borehole  $B_3$ , and from 2 to 3 m in the case of the borehole  $B_4$ .

Vertical distribution of  $^{137}\text{Cs}$  and natural radionuclides in four coring sites were determined by gamma spectrometry measurements in the Laboratory for Nuclear Physics, Faculty of Sciences, Novi Sad, Serbia. Sediment samples were dried at  $105 \text{ }^\circ\text{C}$  until they became a constant mass. After that all the mechanically contaminated particles, mainly small rocks and pieces of plant material were removed. The dried sediment samples were mechanically fragmented and homogenized as fine powder. The prepared sediment samples were packed in cylindrical measurement utensils 62 mm in height and 67 mm in diameter. The typical mass of samples was between 200 and 300 g. There was no radioactive balance established in the samples.

Activity concentrations of radionuclides gamma emitters were determined by the method of low-level gamma spectrometry on actively and passively shielded germanium detectors with maximum background reduction. Time for measurement of these samples was 70,000 s.

Two high-resolution HPGe detectors were used. The first one, produced by CANBERRA has nominal efficiency of 36% and resolution of 1.79 keV. The detector was operated inside the 12 cm thick lead shield with 3 mm Cu inner layer. The second one, germanium detector made by ORTEC was an extended range GMX type detector (10 keV-3 MeV) with nominal efficiency of 32% and resolution of 1.9 keV. The acquired gamma spectra were analyzed using the Canberra Genie 2000 software.

The program calculates the activity concentration of an isotope from all prominent gamma lines after peak background subtraction. All measurement uncertainties are presented at 95% confidence level, meaning that the probability of errors in a repeated measurement of the same sample would be less than 5%.

## Method

If we know the migratory length, that is migration of radionuclides per depth within a period of one year, as well as the age of accumulation, we can determine the value of material import in the accumulation, provided we know the surface density of samples. In such case, we apply the classic equation of radioactive decay

$$A = A_0 e^{-\lambda_{\text{ef}} z} \quad (1)$$

in addition, determine  $\ln A$ , curve inclination determines  $\lambda_{\text{ef}}$  parameter that represents the effective decay constant which is presented as  $\lambda/w$ , where  $\lambda$  is the physical constant of decay (30 years for  $^{137}\text{Cs}$  and 22 years for  $^{210}\text{Pb}$ ) while  $w$  represents migratory length. If the inclination of the function  $\lambda_{\text{ef}}$  is determined using function  $\ln A = f(z)$  that represents the graph of concentration value change of  $^{137}\text{Cs}$  with depth, then it is plausible to directly calculate migratory length in cm per year. This parameter shows radionuclide migration in depth during a year. If the age of the accumulation and surface density of sample is known, this method can be used for determination of preceding material import in the accumulation.

For all the gathered samples, a basic statistical analysis was performed. The analysis included calculation of correlation factor given by [8]. The analysis showed that an apparent correlation between the specific activities of detected radionuclides exists

$$r_{xy} = \frac{\sum_i x_i y_i - \frac{1}{n} \sum_i x_i \sum_i y_i}{\sqrt{\left( \sum_i y_i^2 - \frac{1}{n} \left( \sum_i y_i \right)^2 \right) \left( \sum_i x_i^2 - \frac{1}{n} \left( \sum_i x_i \right)^2 \right)}} \quad (2)$$

## RESULTS AND DISCUSSIONS

The measurement results for the concentration of natural radionuclides in the profile of boreholes B<sub>1</sub>, B<sub>2</sub>, B<sub>3</sub> and B<sub>4</sub> are given in tab. 1. The decrease in the value of radionuclide concentration per depth is visible in all boreholes. In most of these samples, the concentration of natural radionuclide <sup>238</sup>U was under the detection limit. Due to the technical inability to take samples from the same profile depth, it was not possible to compare values of radionuclides concentration between boreholes.

**Table 1. Interval concentration of detected radionuclides (Bq/kg) in borehole samples**

Location	Sampling site-depth [m]	<sup>137</sup> Cs	<sup>210</sup> Pb	<sup>226</sup> Ra	<sup>232</sup> Th	<sup>214</sup> Pb
B <sub>1</sub>	6.8-7.6	3.3-140	60-122	8-23	11-38	6.3-21
B <sub>2</sub>	2.4-5.1	0.8-32	8-105	13-18	2.7-28	2.4-16
B <sub>3</sub>	6.2-7.0	1.6-81	70-100	21-29	40-45	24-31
B <sub>4</sub>	2.0-3.0	5.3-53	44-104	22-25	33-42	24-28

Table 2 shows the values of linear correlation coefficient determined by eq. 2. The correlation concerning borehole B<sub>2</sub> was not determined for radionuclide <sup>226</sup>Ra and other radionuclides, because the values of concentration for <sup>226</sup>Ra were under detection limit. There are small variations in the value of concentration of <sup>226</sup>Ra at all boreholes, which, given the origin of the radionuclides are anticipated.

It is reasonable to assume that the content of radionuclide <sup>210</sup>Pb in the soil derives from two components – one, which is natural and has its origins in <sup>238</sup>U series, and the second component, which is a result of nuclear explosions during the period from 1950 to 1980. This is the reason why linear coefficient of correlation was determined including both total values of <sup>210</sup>Pb concentration and <sup>210</sup>Pb which is a result of nuclear explosions (value of <sup>226</sup>Ra is subtracted from the total values of <sup>210</sup>Pb concentration to get unsupported <sup>210</sup>Pb) [9].

**Table 2. Linear correlation coefficient between radionuclides concentration in the boreholes**

Radionuclides	Linear correlation coefficient			
	Borehole B <sub>1</sub>	Borehole B <sub>2</sub>	Borehole B <sub>3</sub>	Borehole B <sub>4</sub>
Total <sup>210</sup> Pb and <sup>232</sup> Th	0.86	0.95	-0.02	-
Unsupported <sup>210</sup> Pb and <sup>232</sup> Th	0.77	0.65	-0.30	-
<sup>226</sup> Ra and <sup>232</sup> Th	0.64	-	0.08	0.06
<sup>226</sup> Ra and total <sup>210</sup> Pb	0.89	-	-0.72	0.36
<sup>226</sup> Ra and unsupported <sup>210</sup> Pb	0.81	-	-0.78	0.32
Total <sup>210</sup> Pb and <sup>137</sup> Cs	0.19	0.70	0.95	0.14
Unsupported <sup>210</sup> Pb and <sup>137</sup> Cs	0.03	0.64	0.93	0.14

In the case of borehole B<sub>2</sub>, the unsupported <sup>210</sup>Pb was determined by subtraction of the concentration value of <sup>214</sup>Pb from the total concentration value of <sup>210</sup>Pb. This was possible because values for <sup>226</sup>Ra were mostly under the detection values.

The values of linear coefficient show a strong correlation between total concentration values of <sup>210</sup>Pb and <sup>232</sup>Th in the B<sub>1</sub> and B<sub>2</sub> boreholes, while such correlation was not detected in the B<sub>3</sub> and B<sub>4</sub> boreholes. High values of linear coefficient correlation were also detected between the concentration values of radionuclides <sup>226</sup>Ra and <sup>232</sup>Th and concentration of total and unsupported value <sup>210</sup>Pb and <sup>226</sup>Ra only in borehole B<sub>1</sub>. There were no correlation detected between the concentration values of radionuclides <sup>226</sup>Ra and <sup>232</sup>Th and concentration total and unsupported value <sup>210</sup>Pb and <sup>226</sup>Ra in the boreholes B<sub>3</sub> and B<sub>4</sub>.

The values of concentration of <sup>232</sup>Th do not change per depth, unlike the values of concentration of <sup>210</sup>Pb, which decrease in a linear pattern per depth, so it is evident that there is no linear correlation. The reasons may lie in the position of sampling sites, *i. e.* in the characteristics of import sediments. The larger sediments are deposited in the delta, advancing from the upstream end of reservoir towards the dam. During the process of washing out the import, by opening the inlet in the dam, the water in the reservoir gains critical speed, resulting in the vast amounts of the finest sediment being taken out through the inlets, which happens near the borehole B<sub>3</sub>. Furthermore, the borehole B<sub>4</sub> is largely influenced by the torrents of the tributaries, which accumulated great amounts of rocky sediments, enabling sampling only up to one meter in depth. The characteristic of the material import at this sampling site definitely influences the concentration of radionuclides, which was reflected in the nature of linear correlation relation.

A strong correlation between total and unsupported value of <sup>210</sup>Pb and <sup>137</sup>Cs was detected in the boreholes B<sub>2</sub> and B<sub>3</sub>, while in the other two boreholes a linear correlation of those radionuclides was not detected. High values of linear coefficient correlation between concentration value of unsupported <sup>210</sup>Pb and <sup>137</sup>Cs indicate their common origin in the sample soil, which means they are a result of nuclear explosions. The radionuclide <sup>137</sup>Cs, as mentioned before, was used as a parameter for age determination of accumulation. Figures 1 and 2 are showing allocation of <sup>210</sup>Pb and <sup>137</sup>Cs by depth for all boreholes.

The values of migration constants, which were calculated using eq. 1 are presented in tab. 3. Differences between obtained values for <sup>210</sup>Pb migration constants were not determined between boreholes B<sub>2</sub> and B<sub>3</sub> or between boreholes B<sub>1</sub> and B<sub>4</sub>.

The greater values of migration constants were obtained for boreholes B<sub>2</sub> and B<sub>3</sub> in comparison with boreholes B<sub>1</sub> and B<sub>4</sub>, for both <sup>210</sup>Pb, total and unsupported.

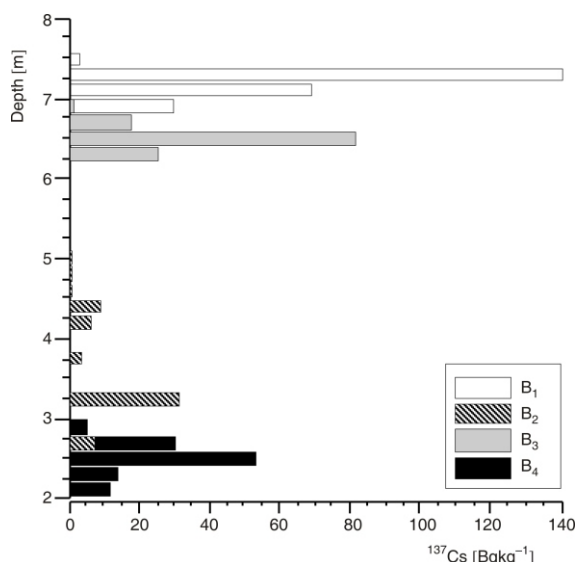


Figure 1. Allocation of  $^{137}\text{Cs}$  by depth

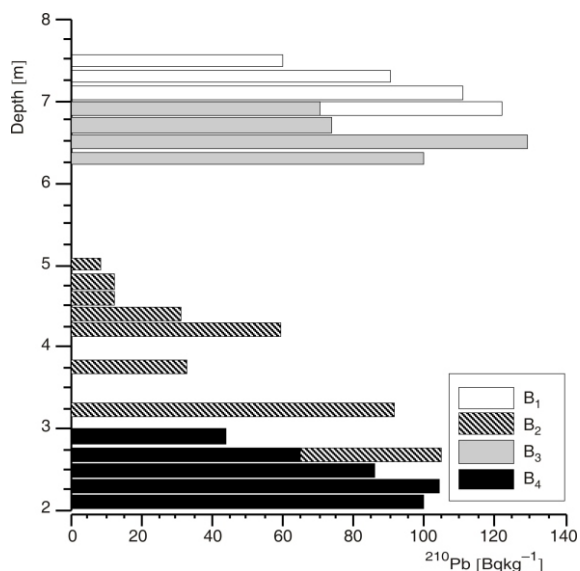


Figure 2. Allocation of  $^{210}\text{Pb}$  by depth

Table 3. Migration constants for  $^{210}\text{Pb}$  and  $^{137}\text{Cs}$  (cm per year)

Location	Total $^{210}\text{Pb}$	Unsupported $^{210}\text{Pb}$	$^{137}\text{Cs}$
Borehole B <sub>1</sub>	2.69	2.88	0.79
Borehole B <sub>2</sub>	3.54	2.90	2.13
Borehole B <sub>3</sub>	3.88	2.31	0.47
Borehole B <sub>4</sub>	2.98	1.96	5.33

Low values of migration constants for  $^{137}\text{Cs}$  were obtained for boreholes B<sub>1</sub> and B<sub>3</sub>, and significantly bigger for the borehole B<sub>4</sub>. The graph indicating the change in the value of  $^{137}\text{Cs}$  shows that in the case of boreholes B<sub>1</sub> and B<sub>3</sub> there is a significant leap in the value of concentration when it reaches its maximum/peak value, and then it suddenly drops to a large

degree. This leap influences the linearity of the curve, which results in small value of migration constant. In addition, in the case of borehole B<sub>4</sub> there is a pronounced maximum of the curve indicating the value of concentration of  $^{137}\text{Cs}$ , which is a parabola, and thus a great deviation from linearity. The values of migration determined by this research show no signs of any significant deviation from the values determined on other accumulations throughout the world, where a research of similar nature was carried out [9-11].

## CONCLUSIONS

The paper displays the results of the Drenova reservoir radioactivity testing which has been performed for the first time. The sediment sampling, which was later analyzed by gamma emitters' spectrometry, was carried out from four boreholes at various points along certain profile.

The preliminary testing showed high values of linear correlation coefficient between unsupported radionuclides in the case of boreholes B<sub>1</sub> and B<sub>2</sub>, unlike boreholes B<sub>3</sub> and B<sub>4</sub>, where these values were low.

It is a different case with the linear correlation coefficient between  $^{137}\text{Cs}$  and  $^{210}\text{Pb}$ , where there is a strong correlation between boreholes B<sub>2</sub> and B<sub>3</sub>, which indicates their mutual origin, nuclear explosions. In the case of boreholes B<sub>3</sub> and B<sub>4</sub>, there is no significant change in the value of concentration for all natural radionuclides by depth, or this change is within error limit, which explains the non-existence of correlation. However, in the case of B<sub>4</sub> borehole the value of concentration of  $^{210}\text{Pb}$  is strongly connected to the change of depth.

The obtained values of migration constant lie within the interval of 1.96 to 2.90 cm per year for unsupported  $^{210}\text{Pb}$  and 0.47 to 5.33 cm per year for  $^{137}\text{Cs}$ , respectively.

The similar values of migration constant for the unsupported  $^{210}\text{Pb}$  indicate that there is no difference in the speed of migration of this radionuclide for different layers. The values gained in this research do not deviate from the values gained in similar research of accumulations all over the world. However, the results indicate the need for more intense sampling, both by depth and by profile, if possible, because that would provide researchers with a clearer view on radionuclide distribution [11].

Within the research, a bathymetric survey was conducted, in order to determine the volume of sediment deposited in the reservoir. Based on the difference of zero and contemporary condition, and by analyzing the digital models of relief, the value of average depth of the deposit was determined (61 cm). This means that the sedimentation rate, gained by dividing this number by the duration of the process of material import in the accumulation, is 2.03 cm per year, which



is very close to the results obtained by  $^{210}\text{Pb}$  and  $^{137}\text{Cs}$  [12] methods.

Given the money and time needed for bathymetric surveys and the production of digital models of relief of zero and contemporary conditions, the  $^{210}\text{Pb}$  and  $^{137}\text{Cs}$ , dating techniques are a powerful tool for determining the sediment accumulation rate. Taking into consideration the extent to which other reservoirs in Bosnia and Herzegovina are investigated the application of this method, with the improvement in sampling techniques, would provide for a better insight into the conditions of reservoirs and prospects of their further exploitation.

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#### **РАДИОАКТИВНОСТ АКУМУЛАЦИЈЕ ДРЕНОВА (БиХ) И МЕРЕЊЕ ДЕПОЗИЦИОНЕ БРЗИНЕ**

У овом раду дате су прве процене радиоактивности и седиментне брзине у акумулацији Дренова. Радиоактивност и седиментна брзина одређене су коришћењем такозване  $^{210}\text{Pb}$  и  $^{137}\text{Cs}$  методе. Узорци су сакупљени из четири бушотине из акумулације Дренова у току 2010. године. Спектрометријом гама емитера коришћењем HPGe детектора, GMX типа (10 keV-3 MeV), одређена је вертикална дистрибуција природних и вештачких радионуклида у свим бушотинама. Добијене вредности концентрације су у интервалу 8-122 Bq/kg за  $^{210}\text{Pb}$ , и 0,8-140 Bq/kg за  $^{137}\text{Cs}$ . Седиментна брзина у акумулацији Дренова је у опсегу 1,96-2,90 cm по години за коригован  $^{210}\text{Pb}$  и 0,47-5,33 cm по години за  $^{137}\text{Cs}$ .

*Кључне речи:* радионуклиди,  $^{210}\text{Pb}$ ,  $^{137}\text{Cs}$ , брзина седиментације, акумулација Дренова