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# Statistical regularities in the distribution of radionuclides in sediments of transcarpathia mountain rivers

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ABSTRACT

The results of the low-background *gamma*-spectrometric measurements of sediments from three Western Carpathian mountain rivers during a three year term (2006–2009) are presented. These sediments are dynamic and very informative for environmental monitoring of a large watershed. Distances between the river sample points were 10–20 km and the total change of the altitude was 200–400 m. The proposed sampling scheme allows to investigate the changes of seasonal and spatial distributions of the radionuclides in sediment. The statistical correlations between the sampling points, the water level of the river and the contents of natural or anthropogenic radionuclides in sediment were studied. The effect of the intensive leaching of radionuclides from sediments during the flood season is shown and this can be considered as the principal self-purification mechanism.



## 1. Introduction

Transcarpathia is a region important from the point of view of water resources and wind flows in Eastern and Central Europe. It is known that the mountain ridges effectively accumulate heavy chemical elements transferred by the wind flows from industry (Grabowski et al., 2006; Pourcelota et al., 2003). Therefore, it is very important to investigate the accumulation and distribution of these pollutants. The monitoring of the Carpathian mountain area for an accumulation of heavy elements in lowland and highland (mountain) ecosystems and investigation of the way system self-purification occurs are attracting enormous attention (Cristache et al., 2009; Kubica et al., 2002; Maňkovska et al., 2008).

Such monitoring can be made by using an appropriate choice of sampling procedure and investigation of selected sets of markers, e.g., gamma active nuclides (GAN), which can be referred to both natural (or geochemical) characteristics of the region and intensity of anthropogenic activities in the neighboring territories (Krmar et al., 2009; Titajeva, 2002; Walling, 2002).

The mountain rivers have more intensive water flow in their highland area than in their lowlands. Their water level can change drastically during the rainy season or flood. There are a limited number of works which have studied the effect of high magnitude floods on the characteristics of river bottom sediments (Symader and Roth, 2002).

The bottom sediments are good sorbents and in the case of mountain rivers they are a dynamic, but very informative substance. Their chemical and microelement composition are influenced by many factors such as the geochemical composition of the nearby soils, meteorological conditions and technogenic activities in the region. Therefore, the spatial and seasonal monitoring of selected sets of GAN in the bottom sediments provides valuable information on intensity and types of anthropogenic activities.

In this paper we present the results of investigations of spatial and seasonal changes of the GAN in mountain river sediments (MRS) of the three largest Carpathian rivers, namely the Borgava, Latorytsa and Uzh.

## 2. The sampling and measuring procedures

We chose several (four or five) sampling points along each river-bed (see Fig. 1). The first sampling point (1T) was situated at the highest elevation of the river-bed where the human influence on the ecological system is minimal. Other sampling points (2T, 3T, 4T and 5T) were chosen downstream where the population level and anthropogenic influence are increased. Distances between the

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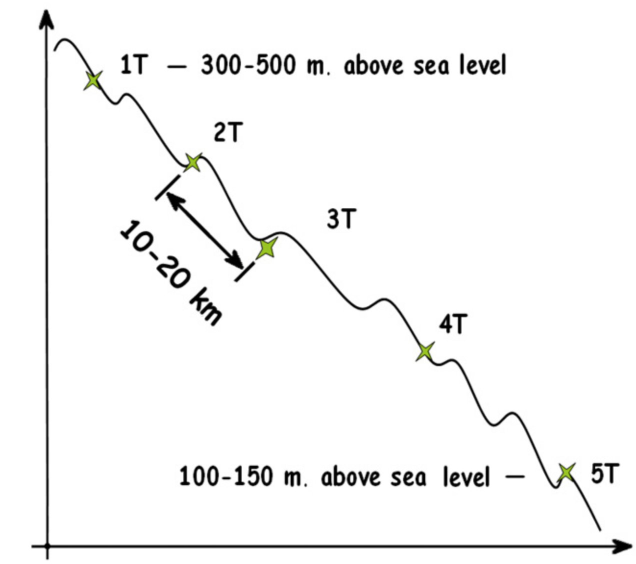


**Fig. 1.** Scheme of a slope long profile with locations of sampling points along the riverbed.

nearest sample points were 10–20 km and the total change of the altitude was 200–400 m. Each sample point was defined by GPS-coordinate. The monitoring was performed for fixed sample points and at each, sampling was done up to 4 times for seasons during three year term (2006–2009).

Sediment samples were taken from water depths of 2–15 cm. They were cleaned from organic substances, dried at room temperature, stored in airtight plastic containers and sealed into a plastic foil to prevent cross contamination. The samples were kept for at least one week before counting, for achieving of the equilibrium between long-lived isotopes and their short-lived products.

Investigations were carried out with the low-background *gamma*-spectrometry method. This method is the most suitable for studying large numbers of samples and is often used for environmental monitoring (Dimovska et al., 2010). The $\gamma$-spectroscopy apparatus included cooled 100 cm$^3$ Ge<Li> and 175 cm$^3$ HPGe detectors and multichannel amplitude analyzers, housed in a low-background laboratory. The shielded environment reduced external *gamma*-radiation 100 times. To reduce the gamma-ray background the detector was shielded by copper (8 mm), cadmium (1 mm), aluminum (3 mm) and a lead (95 mm) layers. Conditions for the gamma-spectrometric measurements such as channel drift and spectrum resolution were controlled during the investigations. The series of measurements which verify background level were performed periodically (from 4 to 30 h) and showed its stability. To guarantee adequate data statistics and their accuracy the optimal measurement time must be compared with background conditions and their possible fluctuations. This verification for counting of natural *gamma*-activity of MRS was carried out within 5–20 thousand seconds range. Each sample was measured for three consecutive times and the counting intervals were 60–90 min per measurement. The measurement time of 5000s yield us the good accuracy of MRS measurements.

The GAN from the U and Th radioactive series can serve as markers for natural or geochemical characteristics of the area (Lozano et al., 2002). This is true under the assumption that secular equilibrium is achieved or almost achieved. This methodic is often used for environmental studies. In work (Dimovska et al., 2010) it was shown that low-background *gamma*-spectroscopy and NAA can give comparable accuracy for the radionuclide content in soils.

The gamma-ray lines of $^{214}$Bi (609.3 keV and 1120.3 keV) and $^{214}$Pb (295.2 keV and 351.9 keV) were used to determine the activity of $^{238}$U. For $^{232}$Th we obtained an equivalent Th content from the detection of $^{212}$Pb (238.6 keV), $^{212}$Bi (727.3 keV) and $^{228}$Ac (338.3 keV and 911.2 keV). The activity of $^{40}$K (used as a natural marker) was determined from the detection of its single gamma-ray line-1460.8 keV.

The intensity of technogenic factors was analyzed by the investigation of the $^{137}$Cs isotope content in MRS of the rivers. The $^{137}$Cs originated from the atmospheric deposition as a result of previous nuclear tests or accidents at nuclear power plants. The latter includes Chernobyl in 1986 and local accidents from nuclear plants based in Carpathian region: Slovakia (Bohunice and Trnava), Hungary (Paks) and Romania (Cernavodă). The $^{137}$Cs in water is from direct deposition on water surfaces and input of $^{137}$Cs adsorbed onto eroded soil particles. Properties of radioactive $^{137}$Cs make it unique as a tracer for studying erosion and sedimentation (Pourcelota et al., 2007). Activity concentrations of any other fission products except $^{137}$Cs were below the detection limits.

## 3. Results and discussion

Seasonal changes of the main GAN in Borzhava river sediments are shown in Fig. 2 for the 3rd sample point (lower part of the river). One can see that the concentrations of $^{214}$Bi, $^{212}$Pb and $^{137}$Cs in MRS oscillated during the time of monitoring. These dependencies were in antiphase with respect to averaged values of the water level of the river. In dry season, when the river water level was minimal, the concentration of the radionuclides in MRS was higher than during the rainy.

This can be explained by following assumptions. The MRS in normal condition is formed mainly by products of soil erosion from large nearby areas and reflects their chemical and isotopic composition. But in long-term drought season and weak water flow the sediments can effectively concentrate heavy metals and their isotopic compositions can vary. During the flood or rainy seasons the leaching of the light sediments fractions with high concentrations of heavy metals occurs caused by intense water flow. Thus their concentration in MRS is decreased. We found the same trend for other rivers and sampling points. This is the evidence of mountain self-purification during flood or high water periods.

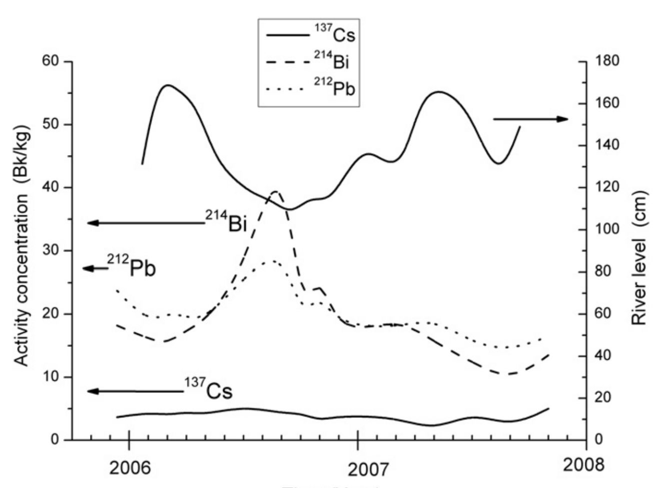


**Fig. 2.** Seasonal oscillations of $^{137}$Cs (dash curve), $^{214}$Bi (dot curve) and $^{212}$Pb (dash–dot curve) concentrations in MRS with respect to averaged values of the water level (solid curve).