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RADIOECOLOGY MONITORING SYSTEM FOR MOUNTINE AREAS OF THE TISZA RIVER BASIN (TRANSCARPATHIA, UKRAINE)

O.I. Symkanich[1],*, N.I. Svatiuk[2], V.T. Maslyuk[2], O.S. Glukh[1], S.N. Sukharev[1], A.A. Sherehiy[1]
[1]Uzhhorod National University,
Uzhhorod, Ukraine;
[2]Institute of Electron Physics of the National Academy of Sciences of Ukraine,
Uzhhorod, Ukraine
****E-mail:Olesjasi123@gmail.com***

Regularities of distribution, migration and accumulation of natural gamma-active nuclides ^{40}K, series of uranium ^{238}U (^{214}Pb, ^{214}Bi) and thorium ^{232}Th (^{212}Pb, ^{212}Bi, ^{228}Ac, ^{208}Tl), as well as technogenic ^{137}Cs in the bottom sediments of the Tisza region in Transcarpathia (Ukraine) have been established. It is shown that the content of these radionuclides in the bottom sediments is relatively stable along the entire length of the Tisza river and is proportional to the level of its siltation, which largely depends on the morphology of the river. It was found that the total specific activity of natural gamma-active radionuclides (excluding ^{40}K) in bottom sediments ranges from 110…139.7 Bq/kg. Areas of accumulation of gamma-active nuclides in the bottom sediments of the studied river have been identified, and the probable causes of this phenomenon have been substantiated. The averaged data of the content of specific activity of radionuclides in the bottom sediments of the Tisza river were used for mapping and forecasting of migration processes, as well as for identification of the studied sections of the river taking into account the geochemical features of the region.

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INTRODUCTION

It is known that the Tisza is one of the important rivers of Central Europe and the largest left tributary of the Danube; therefore, its ecological status affects the surface waters of large surrounding areas in several EU countries [1, 2]. The Transcarpathian region is the only one in Ukraine where its administrative borders coincide with the Tisza river basin's boundaries. All rivers in Transcarpathia flow directly into the Tisza, or its tributaries, and none flows into the rivers of another basin. Therefore, the relevance of ecological studies of the Tisza basin is due to the following criteria: first, mainly the right-bank part of the Tisza river basin, which is located in two geomorphological regions on the southwestern slope of the Carpathians and the southwestern Transcarpathian lowlands, is located in Central Europe. Ukraine, where its administrative borders coincide with the river basin's boundaries (the catchment area of the Tisza within Ukraine is $12.8 \cdot 10^3$ km^2) [3]. Secondly, the combination of physical and geographical conditions with the size and types of anthropogenic load within different basin systems is ideal. Third, the transboundary position of the Tisza river (Romania (51%), Ukraine (25.6%), Hungary (10%), and Slovakia (13.4%)) is of international interest in conducting such studies.

Given the river's ecological significance, it is essential to organize monitoring studies of its condition, in particular, according to the parameters of the content of the specific activity of gamma-active nuclides (GAN). GAN research in the environment, particularly water systems, is due to the involuntary transformation of unstable nuclei of atoms into more stable ones, which is accompanied by the release of intra-nuclear energy and radioactive radiation. The release into the environment of alpha-, beta- and gamma-rays leads to changes in the genome of the cell and other disorders of living systems. In this case, the shorter the half-life of the radioactive element, the more active and dangerous it is, both for the environment and the humans. Also, a good reason in favor of radio-ecological monitoring research of the Tisza is that research on the distribution, migration, and accumulation of radionuclides and heavy metals is practically not conducted, and the data available in the literature are fragmentary and relate exclusively to the study of heavy metals [4–6].

It is known that the mountain ranges of the Carpathians are an essential source of clean water and air. On the other hand, they are natural air filters that accumulate human products (including heavy metals and radionuclides) carried by winds from industrial areas of Europe. As a result, they can lead to changes in the micronutrient and isotopic composition of highland soils. The chemical and microelement composition of mountain soils and water resources of Transcarpathia are formed under climatic, geochemical (natural), and man-made factors [7]. The same factors are decisive for assessing the ecological status of the Tisza river basin. It is also worth noting that the change in the Earth's radiation background has changed significantly in recent years, caused by large-scale nuclear disasters. According to official data [8], the largest nuclear catastrophe of our time, which occurred at the Chernobyl nuclear power plant in 1986, led to the release into the environment of $14 \cdot 10^{18}$ Bq (14 EBq) of the total activity of substances. As a result, more than 200,000 km^2 were contaminated in the form of aerosols, of which approximately 70% were in Ukraine, Belarus, and Russia [8].

The appearance of a significant number of man-made radionuclides, which led to an increase in the Earth's radioactive background, is also due to no less large-scale radiation accident of the maximum 7th level at the Fukushima-1 nuclear power plant, which occurred in 2011 due to the earthquake. Emissions of ^{137}Cs were estimated at $6.1 \cdot 10^{15}$ Bq and ^{131}I $1.3 \cdot 10^{17}$ Bq, comparable to 10% of the Chernobyl accident. Water areas are one of the main reservoirs that ultimately receive artificial radionuclides formed during nuclear explosions and nuclear power plants' operation. One of the factors of radionuclides of natural (products of radioactive decay of natural series of uranium and thorium) and anthropogenic nature in reservoirs is their migration from the soil under the influence of wind, erosion, and agricultural land use.

It should be noted that water status's hydrochemical and radioecological indicators do not always reflect the features and dynamics of chemical changes, trace elements, and radionuclide composition. Hence, the state of bottom sediments of these objects is more informative [9–12]. The bottom sediments of the Carpathian water basins can be considered natural markers of their quality, as they are formed under the influence of natural, in particular, geochemical factors and anthropogenic meteorological and seasonal factors due to soil erosion, etc. [7]. So why bottom sediments are an accumulation environment for both the most common priority and specific pollutants, play an essential role in shaping water quality and are an important component in the environmental safety of regions, and are indicators of the anthropogenesis dynamics.

The composition of the bottom sediments of mountain rivers can be divided into three components: sand (quartz-feldspar), fine (clay), and organic matter. These components are in different ratios, which obviously depends on the flow rate, determining the deposition conditions. The mixture of cluster components has the same radioisotope composition (decay products of uranium-238 and thorium-232) as the original rocks. The silty component of water systems is formed and changed under the influence of river water composition. It is a good sorbent of its chemical content, which depends on the soils' physicochemical characteristics of their catchment areas. The peculiarity of mountain rivers is a significant amount of solid runoff (suspended silt and sand particles), the capacity of which is directly related to the erosion of forest and agricultural land, the state of river valleys (especially riverbanks), i.e., is undesirable. It is important to note that even a small concentration of radionuclides that enter water bodies from the soil's surface layers and atmosphere causes its radioactive contamination, which poses a danger to the environment.

Thus, the data of chemical and radioecological monitoring of bottom sediments of rivers and reservoirs may indicate the degree of pollution of water resources and the ecological condition of large adjacent areas [9]. For more effective control over the pollution of environmental objects with the ability to model and predict any processes and changes in the system under study, it is vital to use modern information technology. Today, using such technologies for the compilation, analysis, and interpretation of thematic maps has become a daily necessity and tool for scientists in processing environmental information. Here, special attention should be paid to geographic information systems (GIS), which allow you to quickly process large amounts of information, which is important in statistical research and identify sources and boundaries of the geological environment's contaminated areas to develop the necessary environmental methods of control.

This paper presents radioecological studies of bottom sediments of the Ukrainian part of the Tisza river basin and maps the areas where sampling was carried out. This approach allows a more objective approach to assessing the ecological status of environmental objects and the organization and reliability of research methods. Besides, mapping the Tisza river's floodplains according to radioecological monitoring will predict the migration process of distribution and accumulation of these ingredients and rationally plan economic activities, including tourism and recreation.

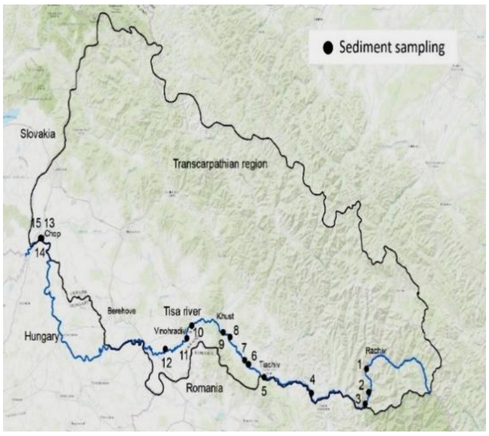
1. MATERIALS AND METHODS

1.1. SAMPLING

The object of the study is radioecological monitoring of floodplains of the Tisza River. Due to the transboundary location of the Tisza, such studies are relevant because they identify both sources of river pollution in Ukraine and abroad with the transfer of pollutants to Transcarpathia. A sampling of the Tisza river's bottom sediments was carried out from a depth of 0…15 cm using dredges at fixed points of the river. The weight of one sample was 1500…2000 g. Transportation, storage, and preparation for analysis were carried out according to current regulations [13, 14]. The Tisza river's bottom sediments were studied quarterly for one year, taking into account landscape and hydrological features in fixed points of settlements (Fig. 1). which correspond to the areas presented in Table.

The proposed sampling scheme of the Tisza river's bottom sediments (see Fig. 1) allows us to solve many tasks. There can assess both the migration processes of radionuclides along the river and identify areas of accumulation of gamma-active nuclides within it, which is important for modeling and forecasting the studied system and adjacent territories.

Before the radiological examination, the prepared bottom sediment samples were placed in sealed containers for three weeks to ensure equilibrium conditions for radionuclides both the uranium ^{238}U as ^{214}Pb, ^{214}Bi, and thorium ^{232}Th. A short half-life characterizes these isotopes, and their decay is accompanied by gamma radiation, which is detected during the analysis.



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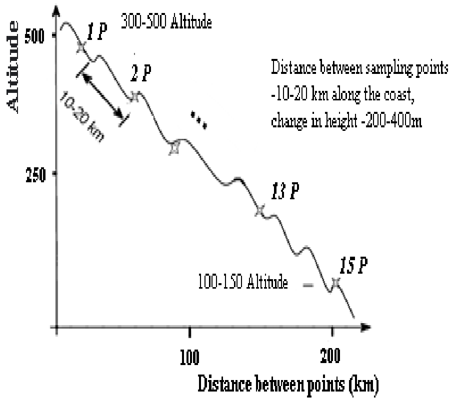


b

Fig. 1. Location of sampling points, 1–15, along the Tisza riverbed (Ukraine – Transcarpathia region) (a); scheme of mountain relief and position of siltation points, 1P - 15P, along the Tisza riverbed (conditionally) (b)

The studied areas of the Transcarpathia region (Ukraine) and their settlements, where the sampling of bottom sediments was carried out

The studied areas of the Transcarpathia region														
№1′ - Rakhiv district			**№2′ - Tyachiv district**			**№3′ - Khust district**			**№4′ - Vynohradiv district**			**№5′ - Uzhhorod district**		
Sampling point														
P1	P2	P3	P4	P5	P6	P7	P8	P9	P10	P11	P12	P13	P14	P15
Settlement														
city Rakhiv	vl. Krugliy	vl. Dilove	vl. Dolotvino	city Tyachiv	vl. Bushtino	vl. Vishkovo	vl. Sokyrnytsya	vl. Khust	vl. Korolevo	vl. Tekovo	vl. Trosnik	city. Chop	city Chop	city Chop

Note. №1′, 2′, 3′, 4′, 5′ – correspond to the study areas where the sampling was performed

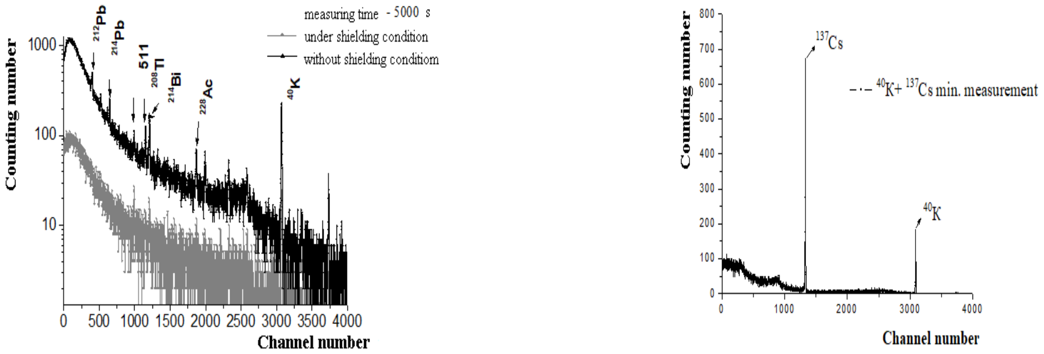
1.2. ANALYSIS OF RADIONUCLIDES

Determination of the specific activity of GAN was performed in low-background conditions on a gamma-spectrometric complex "SBS" with a cooled 100 cm^3 and 175 cm^3 HP Ge-detector, which was in combined protection shield, that allowed to reduce own radioactive background of the setup in ~ 50 times concerning background conditions of the laboratory. When identifying gamma lines in the studied samples' experimental spectra and performing calculations of specific activity, a catalog of gamma spectra of the GAS was used. Measurements were performed according to the recommendations of [15, 16].

Calibration of the gamma-spectrometric complex "SBS" was performed according to the reference source (KOUHC), which is a Marinelli vessel filled with a standard sample (^{152}Eu and combined samples ^{40}K-^{137}Cs) with clearly defined energies. When calibrating the device, the reference source's energy must be compared with the centroid of the corresponding peak of full absorption on the instrument spectrum [17]. Calibration adjustment is performed according to the "channel-energy". The example of the spectrum of reference sources used for calibration of the detector and fragments of the gamma-spectrum of background measurement conditions for two cases (1 – the detector in combined protection, 2 – detector without protection) is presented in Fig. 2.

As shown in [18], increasing the time and statistics of measurements reduces the confidence intervals of GAN activity's measured values. However, the established errors at 5000 and 21600 s do not differ significantly. Thus, we can say that the optimal time that provides sufficient reproducibility and, possibly, GAN determination accuracy is 5000 s, which we used to analyze the measurement of radionuclides' specific activity by gamma-spectrometry in the studied samples of bottom sediments of the Tisza river.

It should also be noted that, if necessary, the measurement time can be extended, which will improve the accuracy of the analysis results, but in monitoring studies, the excessive time is inexpedient. Values of nuclear physical constants were used to identify gamma-lines in the spectra of bottom sediment samples and calculate specific activity [19].



a b

Fig. 2. The spectra of low-background measurement conditions for two cases: 1 – detector in combined protection, 2 – detector without protection (a); the instrumental spectrum of standard sources used to calibrate the detector (b)

The absolute activity of the radionuclide A obtained for total absorption peak for gamma line with energy E is given by the relation [20]:

$$A = \frac{S_p}{\varepsilon_E \times t \times l_\gamma}, \qquad (1)$$

where S_p – is the peak area of total absorption with energy E; ε_E – efficiency of the detector for energy E; t – "live" measurement time; $l\gamma$ is the quantum yield (number of gamma-quanta per decay) for a given energy E.

The specific activity of the radionuclide is calculated according to formula (1):

$$A_m = \frac{A}{m}, \qquad (2)$$

where A is the absolute activity of the radionuclide, m is the mass of the sample.

For processing, full absorption peaks were used, which belonged to specific radionuclides and met condition (3) – the criterion of elimination of peaks [21]:

$$S_p \geq 3 \times \sqrt{S_B}, \qquad (3)$$

where Sp is the peak area, S_B–background area.

After measuring the specific activity of the studied sample of bottom sediments, the experimental installation was also calibrated. A standard radioactive source ^{60}Co (1332.5 keV-characteristic line gamma-radiation), certified by SE Kyiv Regional Research and Production Center for Standardization, Metrology, and Certification, was used. The drift of channels and resolution (for ^{40}K not more than ± 3.2 keV; for ^{137}Cs not more than ± 2.3 keV) were monitored continuously throughout the measurement. Optimal conditions for the following studies: temperature (20 ± 3) °C, and relative humidity – 50%, which in this work was provided by the use of dehumidifier (Pd 3000).

1.3. MAPPING

According to chemical and radioecological monitoring, mapping of areas was performed using the computer program "ArcGIS 10.2.1" regarding GPS coordinates [22]. ArcGIS platform is a world leader in the construction and use of geographic information systems (GIS) and is used by people worldwide to apply geographical knowledge in the practical field of public administration, science, education, and business. The use and application (GIS) of ArcGIS to analyze radioecological processes in the study areas will effectively process large amounts of information needed to address the rehabilitation of contaminated sites. The ArcGIS package functions are accessed via a graphical interface, the set of tools of which depends on the software module. The ArcMap version 10.2.1 module was used for the analysis. The input data for the ArcMap module was presented in an accessible tabular format.

2. RESULTS AND DISCUSSION

Radioecological monitoring was performed on natural GAN, particularly ^{238}U and ^{232}Th series, as well as ^{40}K and man-made ^{137}Cs by low-background gamma spectrometry. It should be noted that the GAN of natural and artificial origin contained in the soils and bottom sediments of mountain rivers can serve as "labels" for both geochemical indicators of the region and the intensity of urbanization processes [23, 24]. Analysis of radio spectroscopic analysis data showed that maximum value of ^{40}K is set for №1′ (in particular, №2 – 461 Bq / kg, №3 – 454 Bq / kg) – mountainous area, the lowest – №5′ (№14 – 331 Bq / kg) – lowland area. This distribution is due to the geomorphological, geochemical, and tectonic features of the studied areas and the sediment's nature. In general, the data show that the content of ^{40}K in bottom sediments does not exceed acceptable levels [7].

It was found that the specific activity of individual natural GAN series ^{238}U and ^{232}Th in the bottom sediments of the Tisza river is relatively stable within the studied areas. In contrast, the sum of natural GAN (excluding ^{40}K) increases with the transition from №5′ to №1′ sampling points (mountainous terrain). According to [24], the increased content of natural radionuclides in mountainous and foothill areas may be due to the young Carpathian Mountains' seismic activity, where earthquakes occur periodically. This fact may contribute to Radon's release (^{222}Rn and ^{220}Rn) – the decay products are ^{214}Bi, ^{214}Pb, ^{212}Bi, and ^{212}Bi, forming the natural radioactivity of soils, and hence bottom sediments.

The standards of natural and man-made GAN contents in environmental objects may serve [25] as marks for whole investigated areas. Examples for these marks can be the ^{214}Bi natural GAN for the ^{238}U series and ^{212}Pb for the ^{232}Th series, and the integrated activity of the natural GAN.

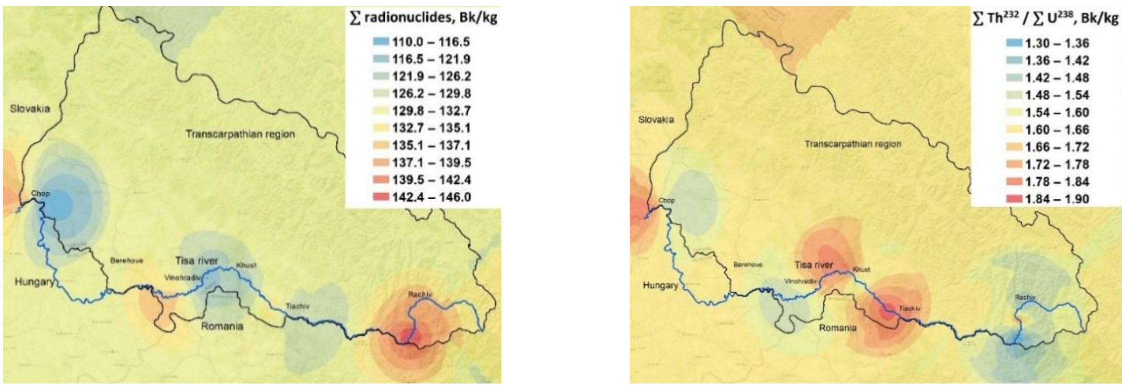
An important indicator that can also be used to identify areas is $\Sigma\,^{232}$Th / $\Sigma\,^{238}$U [25]. The ratio of the total activity of natural GAN of the ^{232}Th series to the ^{238}U series in the studied bottom sediments for the Tisza river is relatively stable between samples from the same macro location, and it varies from 1.3…1.9 along the river within the region. In the bottom sediments, sampling point №1′, the GAN of the series ^{238}U has a more dominant component, while in other studied areas of the Tisza river – the GAN predominates in the series ^{232}Th. If the Th/U ratio in bottom sediments within a local area differs significantly from this ratio within the region, we can talk about a geochemical anomaly. The study of the conditions of imbalance of the genetic GAN series ^{232}Th and ^{238}U is a difficult task for scientists, which needs more sufficient statistics, the number of studied isotopes, and the knowledge about chemical conditions and trace elements formation in the sediments.

It is shown in [7] that the geochemistry of the progenitors of the decay series U and Th depends only on their chemical properties with similar environmental characteristics. However, for intermediate members of the series, in addition to chemical properties, indicators due to radioactivity should be taken into account: communication with the ancestor of the series, rate of radioactive decay/accumulation, diffusion of recoil atoms, dependence on the presence of carriers such as ^{238}U or ^{232}Th. As expected, the specific activity of man-made ^{137}Cs, which arose as a result of the Chernobyl accident in 1986, changes abruptly in the bottom sediments, both within the study areas and between different areas of the Tisza, with a maximum content of №1′, № 4′–8.9 Bq/kg and the lowest for №5′–2.1 Bq/kg.

This fact may be due to the zonal inflow of this radionuclide by rain and wind flows from remote areas, as mountains are a natural barrier to the spread of air masses and significantly affect man-made waste, especially radionuclides from radioactive clouds. It should be noted the values of ^{137}Cs specific activity obtained by us are close to the same data of this radionuclide for the Tisza river in the Hungarian-Serbian part and lower than the content in the bottom sediments of the Danube and Bega rivers [2, 26]. Thus, the average values of ^{137}Cs activity concentrations (Hungarian-Serbian part) in the Danube river are (28 ± 23) Bq/kg, the Bega River is (45 ± 7.4) Bq/kg, and the Tisza river is (6.6 ± 0.6) and (4.3 ± 2.3) Bq/kg (Ukrainian side). Comparing the concentration of ^{137}Cs radionuclide activity in the Tisza river's bottom sediments (Hungarian-Serbian and Ukrainian part) with the corresponding data for the Danube and Bega river sediments, it is shown that the Tisza river channel sediments are relatively clean of man-made radionuclides.

Based on the data of the specific activity of GAN in the bottom sediments of the Tisza river, we constructed a map of the distribution of the total specific activity of natural GAN (excluding ^{40}K) and a map of the distribution of the ratio $\Sigma\,^{232}$Th / $\Sigma\,^{238}$U, which is presented in Fig. 5. Mapping of the radioecological condition of the bottom sediments of the Tisza river was carried out using the computer program "ArcGIS 10.2.1".

Data analysis of Fig. 3 shows that we can distinguish three zones of accumulation of natural GAN and low radionuclide content zone. This pattern is explained, apparently, by the different accumulating properties of the bottom sediments of small rivers, their chemical and particle size distribution, and the different amount of silt in the rivers. On the other hand, bottom sediments' radioactivity depends on the intensity of radionuclide exchange processes between soils and water resources and water content.



a b

Fig. 3. Distribution map a - the total specific activity of natural GAN (excluding ^{40}K): b – $\Sigma\,^{232}$Th / $\Sigma\,^{238}$U ratio for the Tisza River according to the state of their bottom sediments

It is known [7] that the Carpathian mountains are relatively young and seismically active, and in rocks of volcanic origin, there are more natural isotopes than in sedimentary ones. As a result of their removal from geological or soil horizons with various processes, these isotopes fall into the bottom sediments with their subsequent movement along the river.

Hydrodynamics also makes significant adjustments to the accumulation and migration processes of isotopes. In particular, in [7], a study of the distribution of specific GAS activities of natural and man-made origin in the silt of the Borzhava river flows into the Tisza river. It is established that the growth of indicators of specific activities of GAS of natural and man-made origin in the bottom sediments of the Borzhava river during the summer months (the water level in rivers is minimal) and the decrease of their content in the spring months. In other words, this process is oscillating, so the fluctuations of the specific contents of GAN in silt occur in antiphase to the water level in mountain rivers.

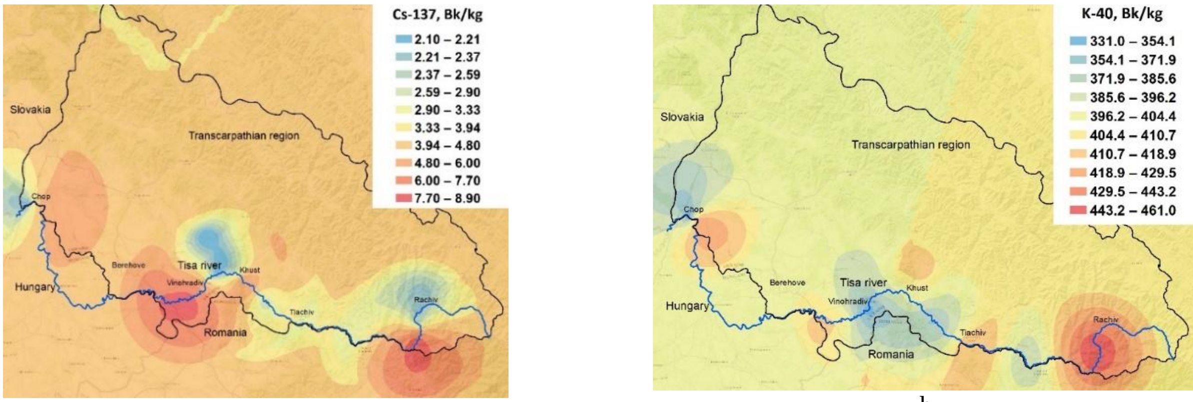


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Fig. 4. The distribution map of the specific activity of ^{137}Cs (a) and natural ^{40}K (b) of the Tisza according to the state of their bottom sediments

The total specific activity $\Sigma\,^{232}$Th / $\Sigma\,^{238}$U in the studied samples shows changes between the samples of the studied areas. In Fig. 4 shows that the zone of accumulation of specific activity $\Sigma\,^{238}$U in the bottom sediments of the Tisza River is more pronounced in the region №1′, and $\Sigma\,^{232}$Th in №2′ №3′ №4′, which may be due to both geomorphological features of the territories and heterogeneity sedimentation.

It should be noted that the accumulation zone of natural GAN for bottom sediments №1′ is the most intense, which is obviously due to the sharp break of the river (slowing down) and, as a consequence, greater siltation. Such zones of radionuclide accumulation in the bottom sediments of rivers in the mountain landscape are undesirable, and such zones should be considered when using these rivers.

Comparing the radioecological state of the Tisza river's bottom sediments in terms of ^{137}Cs and ^{40}K content (see Fig. 4) is somewhat similar

The comparison of the radioecological state of the bottom sediments of the Tisza river in terms of ^{137}Cs and ^{40}K content (see Fig. 4) is somewhat similar. As in the case of the distribution of the total specific activity of natural GAN in the Tisza river's bottom sediments (see Fig. 3), the accumulation zone of ^{137}Cs and ^{40}K is more pronounced in №1′ area with their gradual migration to lowland areas along the river.

That is, the higher the altitude above sea level, the higher the content of radionuclides.

The distribution of ^{40}K in sediments along the river length is more stable within two distinct zones of accumulation compared with the uneven content of ^{137}Cs radionuclide – with the allocation of three zones of increased specific activity areas with morphological changes of the Tisza.

We used cluster analysis to find out the statistical relationship between the content of these radionuclides (^{137}Cs and ^{40}K) depending on the territorial factor. The results of cluster analysis are presented in Fig. 5. It is shown that there is a pronounced territorial distribution of ^{137}Cs and ^{40}K in bottom sediments and depending on the sampling point. There are strong correlations on ^{137}Cs and ^{40}K contents for mountain points, smaller (less than the value of the aggregation parameter) – for lowland.

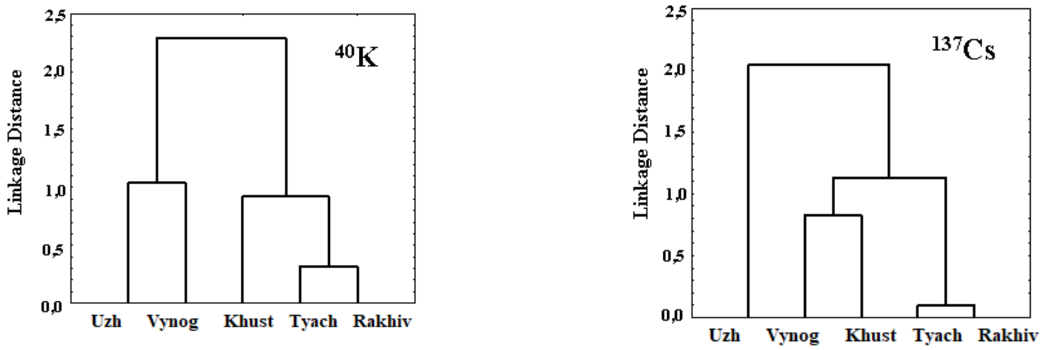

Fig. 5. The cluster analysis results (distribution of GAN natural a – ^{40}K and man-made b – ^{137}Cs) to establish the degree of statistical proximity of sampling points along the Tisza riverbed

This fact may indicate the accumulation of man-made ^{137}Cs in the Carpathians' highlands due to the interaction of mountains with radioactive air currents transferred from remote areas due to the Chernobyl accident, thus the migration of this GAS from soils to bottom sediments.

CONCLUSIONS

Thus, the obtained results testify to the peculiarities of distribution, migration, and accumulation of gamma-active nuclides in the Tisza river's bottom sediments. Mapping makes it possible to establish areas of accumulation of gamma-active nuclides in the Tisza river and areas with the lowest content of these ingredients, allowing forecasting the future state of the studied areas and is an essential aspect of environmental safety. It was found that for the Tisza river, the zones of accumulation of gamma-active nuclides are most pronounced in areas of change in the morphology of the river, especially clearly observed in the mountainous part of the river, which should be taken into account when studying this river. According to the analysis of literature sources, such studies for the Tisza river have not been conducted before and are becoming relevant in today's conditions. Also, there is reason to believe that the same patterns apply to other mountain ranges in Europe.

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РАДІОЕКОЛОГІЧНИЙ МОНІТОРИНГ ГІРСЬКИХ СИСТЕМ БАСЕЙНУ РІЧКИ ТИСА (ЗАКАРПАТТЯ, УКРАЇНА)

О.І. Симканич, Н.І. Сватюк, В.Т. Маслюк, О.С. Глух, С.М. Сухарев, А.А. Шерегій

Встановлені закономірності розподілу, міграції та акумуляції природних гамма-активних нуклідів ^{40}К, рядів урану ^{238}U (^{214}Pb, ^{214}Bi) і торію ^{232}Th (^{212}Pb, ^{212}Bi, ^{228}Ac, ^{208}Tl), а також техногенного ^{137}Cs у донних відкладах р. Тиси, Закарпатської області. Показано, що вміст цих радіонуклідів у донних відкладах є відносно стабільним за всією протяжністю р. Тиси і пропорційний рівню її замулювання, що в значній мірі залежить від морфології річки. Встановлено, що сумарна питома активність природних гамма-активних радіонуклідів (без урахування ^{40}К) у донних відкладах коливається у межах 110…139,7 Бк/кг. Виявлені ділянки акумуляції гамма-активних нуклідів у донних відкладах досліджуваної річки, обґрунтовані ймовірні причини цього явища. Усереднені дані вмісту питомої активності радіонуклідів у донних відкладах р. Тиси використані для картування і прогнозування міграційних процесів, а також для ідентифікації досліджуваних ділянок річки з урахуванням геохімічних особливостей регіону.