SOME ASPECTS OF THE INFLUENCE OF URANIUM EXPLOITATION ON THE ENVIRONMENT

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Abstract: Gamma-ray spectrometric measurements of samples of riverbed sediments and soil samples taken along the valley of a river, which runs very close to a uranium mine retaining dam are performed. The content of ^{238}U, ^{226}Ra, ^{210}Pb, ^{232}Th, and ^{40}K is analyzed. Up to a distance of about 6 km away from the retaining dam, ^{238}U, ^{226}Ra and ^{210}Pb have high concentrations and the content in the sediments samples is consistently higher than the content in the soil samples. In the same interval are observed considerable fluctuations in the contents related to the swamping of the river. Receding at a greater distance from the retaining dam, the concentration of ^{238}U, ^{226}Ra and ^{210}Pb decreases and has values close to the average ones. A very close correlation is established between the contents of the three radioactive nuclides. Regarding ^{232}Th and ^{40}K, the distribution characteristics along the profile are different in comparison with those of the ^{238}U family members. The performed research contributes to the estimate of the radioactive contamination in a specific area situated in the vicinity of a uranium deposit exploited through underground mining.

Keywords: mineral, uranium, radioactivity

1. Introduction

In the last decades the exploitation of mineral resources has caused serious ecological problems. The exploitation of uranium is giving practically the same waste products as the other branches of the mining industry. Something more, all waste products from the uranium exploitation (with small exceptions) are radioactive. During the process of exploitation of uranium deposits in the environment are entering radioisotopes from the three radioactive families - ^{238}U, ^{235}U, ^{232}Th. But the radioactivity of the waste products is determined mainly by ^{238}U and the products of its decay, among which most active are ^{230}Th, ^{226}Ra, ^{222}Rn. The uranium itself has low radioactivity, but its chemical activity is very dangerous. It should be mentioned that the activity of the waste products form the exploitation of uranium is lower in comparison with the radioactivity of the waste products from the other stages of the nuclear energy production cycle. Anyway, their estimation is imperative and of public health significance, as they can cause radiation intensities above the accepted tolerance levels for a very long period of time - hundreds and thousands of years.

The performed studies are aimed towards obtaining an estimate of the radioactive contamination in a specific area situated in the vicinity of a uranium deposit exploited through underground mining.

2.Main aspects of the influence of uranium exploitation on the environment

In the case of uranium deposits exploited through underground mining, the direct sources of contamination of the environment are the drifts and shafts through which ore is extracted, transported and hoisted, the ventilated air and the mine water. In separate occasions, the radioactive contaminators are going into the environment through channels of leakage - for example radon entering the atmosphere through zones of broken rocks, system

of joints and unplugged drill-holes, or drainage of radioactive water through faults, systems of joints, unplugged drill-holes and filtration.

Generally, the level of radioactive contamination of air within the boundaries and especially outside the boundaries of the industrial areas of the nowadays mines for uranium exploitation is low, as both, the radon and the aerosols, are mixing very quickly with large volumes of air. For the majority of mines, a radioactive pollution of air, which is higher than the background one, can be observed only at a distance up to 100-200 m from the source of contamination and therefore it is practically not going outside the boundaries of the industrial areas. The industrial water from the uranium mines is usually exporting into the environment two contaminators - uranium, which is entering the hydrographic system or the ground water, and radon, which is going into the atmosphere. The quantity of the exported uranium depends strongly upon the mine water level of acidity, which, on its side, is attributable to the presence of sulphides in the ore and in the host rocks. Most frequently, the uranium minerals are in association with pyrite. Pyrite (FeS_2) is the mineral, which is the most active producer of sulphurous acid (H_2SO_4) in nature. It is oxidized by hemolithotropic bacteria in aerobic medium ($4FeS_2 + 15O_2 + 2H_2O \rightarrow 2Fe_2(SO_4)_3 + 2H_2SO_4$). The sulphurous acid dissolves the six-valence form of uranium U^{6+} and the result is UO_2SO_4 , which is very agile. Like this the uranium migrates into the environment.

The passage of radon into the mine water is a common and permanent event, which is not dependent on the geo-chemical conditions. The process of saturation of mine water with radon complies with the law of gas parcel pressure and that is why the ground water, which carries in itself extremely big quantities of radon, is liberating quickly the supplementary amount. The mine water radon concentration is also depending on the water temperature - the lower it is, the more radon can be dissolved. Contrary to the cases of uranium and radium, the concentration of radon in the mine waters is rapidly decreasing due to its short half-life. In the near past, the contamination of the surface water with natural radionuclides during the exploitation and processing of radioactive ores was a common event. The mine water was running, practically without any purification, into the hydrographic system. Huge volumes of trailing water were filtrated out of the retaining dams. Radioactive contaminators were entering into the hydrosphere also during the export of natural radionuclides from the ore stores. It should be mentioned that the contamination of the surface water, especially the running one, is not as dangerous as the one of the ground water or the non-running water.

This is connected to the fact that the time period of a complete water turn-over, for example in the rivers, is usually days, but not exceeding weeks. The contaminated deposits can be extracted and dumped into deep-seated formations for example, although this operation is very expensive. Much more complicated is the problem of purification and neutralization of ground water, which has a time period of complete turn-over surpassing hundred years.

3.General information on the performed research

Gamma-ray spectrometric analysis was performed. Samples of riverbed sediments and soil samples taken along the valley of a river, which runs very close to a uranium mine retaining dam were counted (fig.1). The total profile length along the river valley is about 9 km. The samples were taken at intervals of 350-450 m. The sediment samples were collected form the riverbed and the soil ones - at a distance of 20 m from the riverbank. On top of this, five samples were taken along each of three profiles, perpendicular to the river course, at

distances 40, 60, 80, 100 and 120 m from the riverbank respectively. The soil samples were picked starting from station 7, as up to this station the river is running through a village.

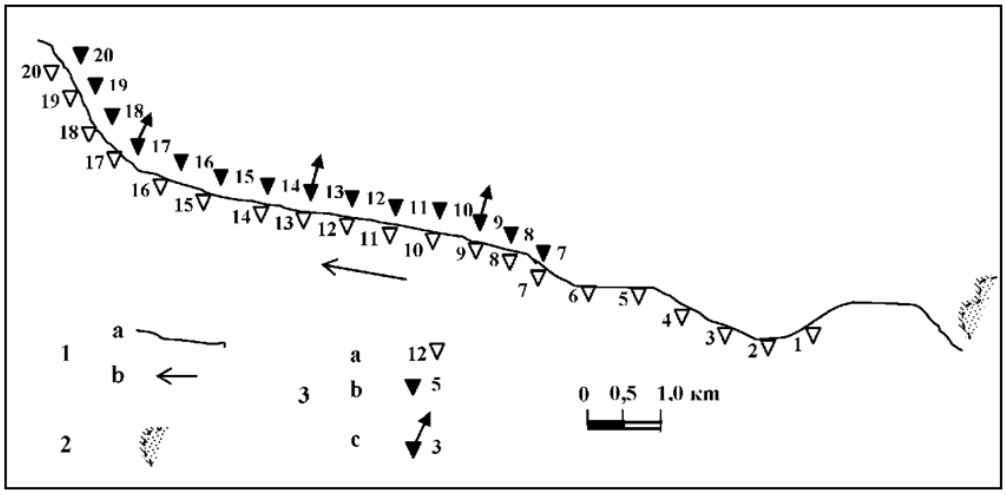

Fig.1. *Scheme of the stations of sample collection:*
1 - river: a - riverbed; b - stream direction; 2 - uranium mine retaining dam; 3 - stations of sample collection: a - samples of river bed sediments; b - cultivation soil samples; c - profiles along which were taken five additional samples at distances 40, 60, 80, 100 and 120 m from the riverbank respectively

The spectrometric analysis includes measuring the activity of the radionuclides which have significant importance as environment contaminators resultant from the uranium exploitation - ^{238}U, ^{226}Ra, ^{210}Pb, ^{232}Th, and also measuring the radioactivity of ^{40}K. The basic information on these radionuclides and on the gamma-ray energy lines utilized in the analysis is systematized in Table 1.

In order to estimate the analysis precision, each second (even numbered) sample has been measured twice. The differences are within the limits of the measurements relative error. The errors statistical distribution is illustrated in fig.2.

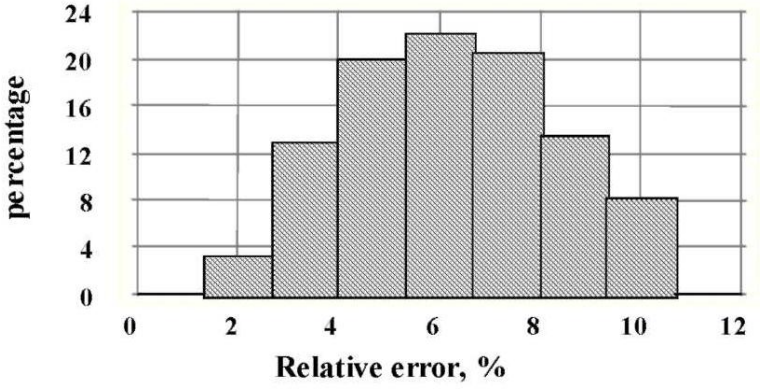

Fig.2. Histogram of the analysis relative error distribution

The data obtained from the measurements are presented as distribution charts for the specific radionuclides along the straightened out studied profile. Statistical analysis is performed in order to establish the relationship between the radionuclides.

Tabel 1. *Basic information on the studied radionuclides -* ***^{238}U, ^{226}Ra, ^{210}Pb, ^{232}Th and ^{40}K***

Radio-active nuclide	Half-life Tl/2, Years	Main biological influence	Content in soil, %	Activity of soil layer 0-10 cm, Bq/kg	Gamma-ray energy lines utilized in the analysis
^{238}U	$4{,}51.10^{9}$	Enters the body system mainly with the food. Causes chronic radiation disease and lung pathology. Has also influence upon the central nervous system and the endocrine glands.	$0{,}1.10^{-4}$ to 7.10^{-4}	5 to 250	92,59 KeV main energy line ; 63,29 keV and 1001,3 keV additional energy lines
^{226}Ra	1601	Enters the body system mainly with the food. About 80% of it are accumulated in the bones and cause increased fragility. Acts as a toxin and causes blindness and cancer disease.	0 to $1{,}2.10^{-10}$	7 to 150	Through its daughter product ^{214}Bi having half-life $T_{1/2}$ = 19,9 min and 609 keV, 1120 keV and 1765 keV energy lines
^{210}Pb	22,3	One of the most dangerous nuclides for the public health. Accumulates in the bones and causes damage to the brain. Stimulates the development of cancer disease.	$0{,}3.10^{-12}$ to 10.10^{-12}	10 to 200	46,5 keV energy line
^{232}Th	$1{,}41.10^{10}$	Enters the body system mainly with the food. Accumulates mainly in the lung, the skeleton and the marrow. Smaller quantities cumulate in the liver and in the spleen. Causes anaemia.	5.10^{-4} to 50.10^{-4}	2 to 120	Through 911 keV energy line, belonging to its daughter product ^{228}Ac
^{40}K	$1{,}28.10^{9}$	In homeo-stochastic equilibrium	0 to $1{,}4.10^{-4}$	110 to 1200	1460 keV energy line

4.Results from the performed research

The distribution charts of the content of ^{238}U, ^{226}Ra and ^{210}Pb along the straightened out studied profile are presented in fig.3, fig.4, and fig.5.

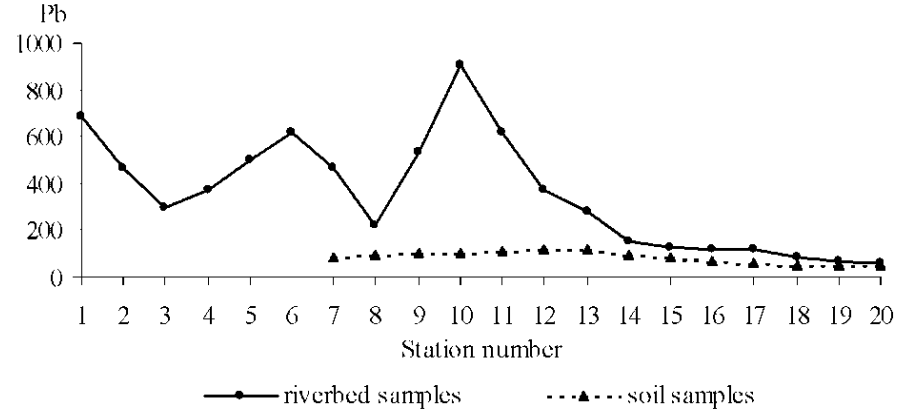

Fig.3. Distribution chart of the content of ^{238}U, Bq/kg along the straightened out studied profile

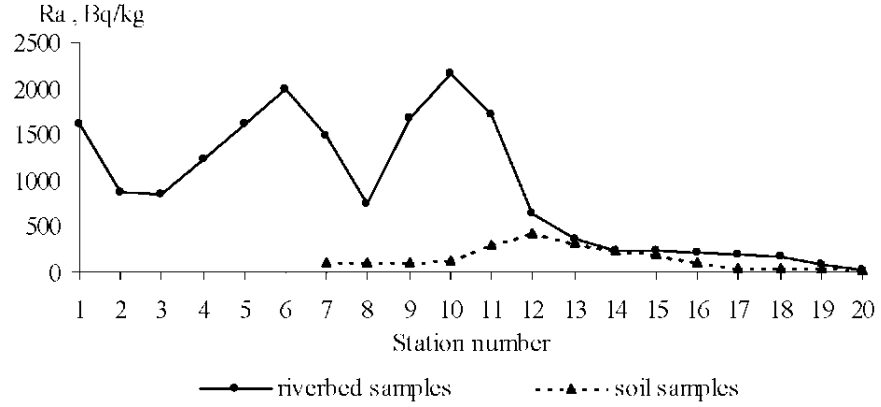

Fig.4. Distribution chart of the content of ^{226}Ra, Bq/kg along the straightened out studied profile

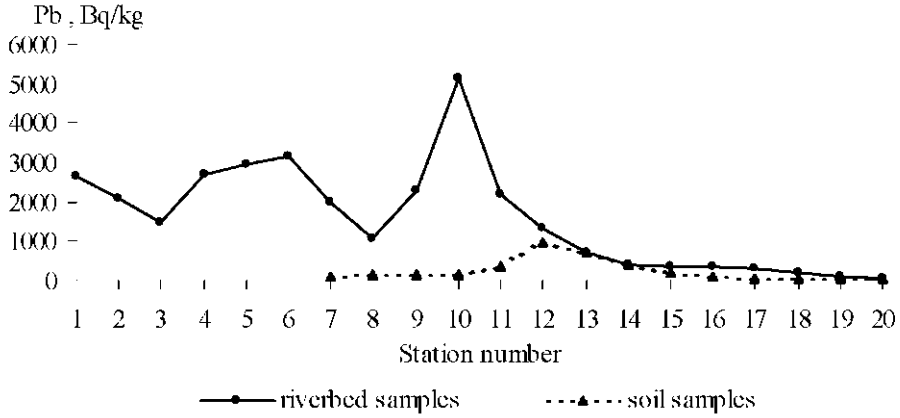

Fig.5. Distribution chart of the content of ^{210}Pb, Bq/kg along the straightened out studied profile

The distribution analysis is showing, that up to a distance of about 6 km away from the retaining dam, all three radioactive nuclides have high concentrations and the content in the sediments samples is consistently higher than the content in the soil samples. Up to this distance, there are considerable fluctuations in the contents in the river bed sediments. This fact is due to the swamping of the river and the developing of favorable conditions for adsorption of the radioactive nuclides. In these anomalous zones the content of ^{238}U, ^{226}Ra and ^{210}Pb is very high. It surpasses the average specific activity of soil layer 0-10 cm in Bulgaria (Table 1) about 4-5 times for uranium, 10-12 times for radium and 20-30 times for lead respectively.

Receding at a greater distance from the retaining dam, the content of the radionuclides is displaying a consistent trend towards decrease and has values approximating the average ones for the uncontaminated soil.

A very close correlation is established between the contents of the three radioactive nuclides belonging to the family of ^{238}U. In fig.6 is illustrated the fitted simple linear regression equation ^{226}Ra [Bq/kg] = f {^{238}U [Bq/kg]}.

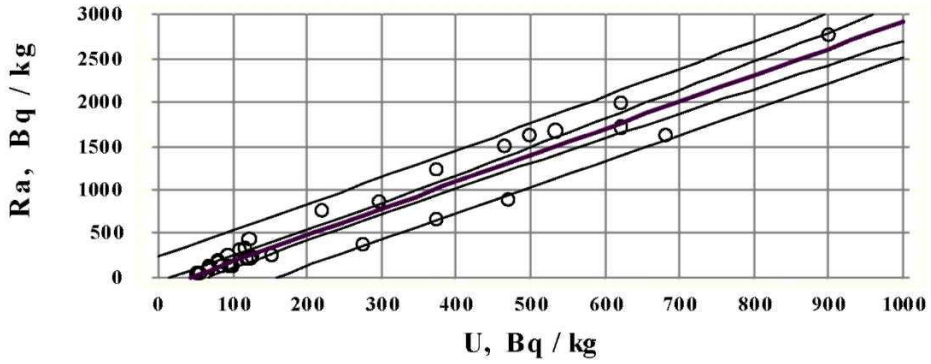

Fig.6. *Plot of fitted simple linear regression equation ^{226}Ra [Bq/kg] = f {^{238}U[Bq/kg]}*
Correlation coefficient = 0,98

The analysis shows that the specific activity and respectively the content of ^{226}Ra are about three times higher than the values in the case of radioactive equilibrium with ^{238}U . This anomaly is due to the fact that uranium first of all has already been separated in the processing, and second - it forms easily soluble complex compounds and in such a way is transported away by the running water.

Very high coefficients of correlation are obtained for the fitted simple linear regression equations ^{210}Pb [Bq/kg] = f {^{226}Ra [Bq/kg]} - fig.7 and ^{210}Pb [Bq/kg] = f {^{238}U [Bq/kg]} - fig.8. Their analysis is showing that the specific activity and respectively the content of ^{210}Pb are about two times higher than the values in the case of radioactive equilibrium with ^{226}Ra, and about six times higher than in the case of radioactive equilibrium with ^{238}U.

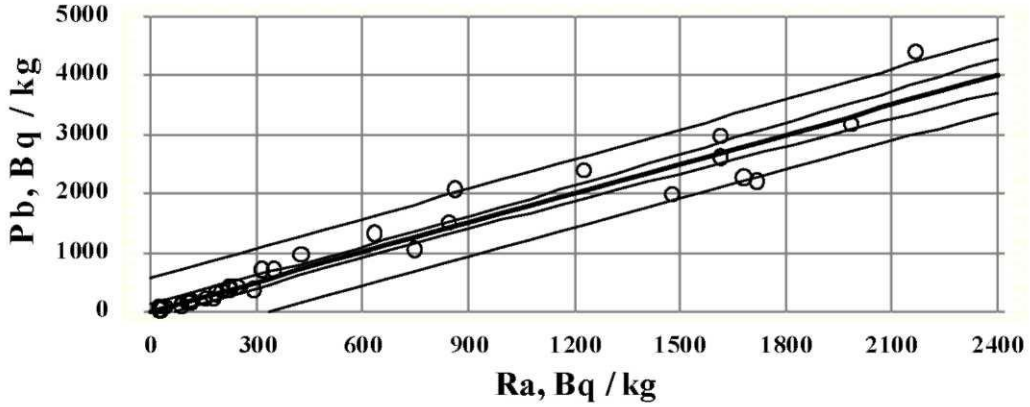

Fig.7. *Plot offitted simple linear regression equation ^{210}Pb [Bq/kg] = f {^{226}Ra [Bq/kg]}*

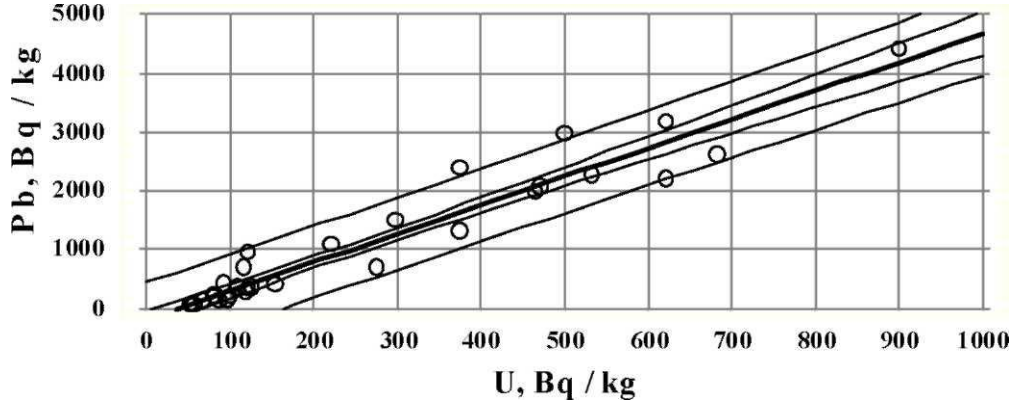

Fig.8. *Plot of fitted simple linear regression equation Pb [Bq/kg] = f { U[Bq/kg]}*
Correlation coefficient = 0,97

Regarding Th and K, the distribution characteristics along the profile are different in comparison with those of the radioactive nuclides belonging to the family of ^{238}U (fig.9 and fig.10). The contents in the soil samples remain without significant changes. In the case of the samples of riverbed sediments, increased contents are observed in the intervals of swamping and a consistent trend towards decrease in the contents along the river course after the village.

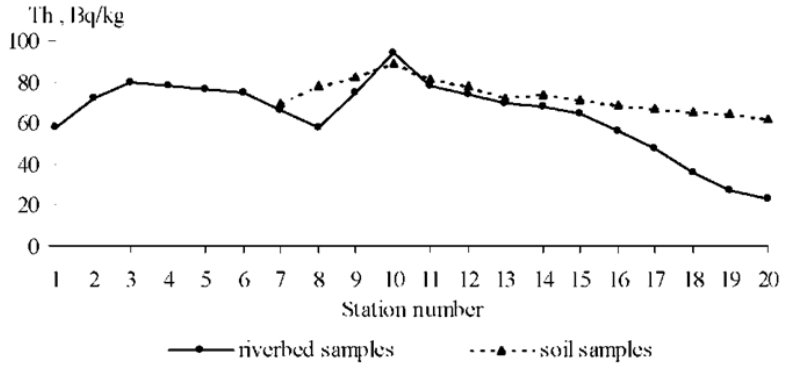

Fig.9. Distribution chart of the content of ^{232}Th , Bq/kg along the straightened out studied profile

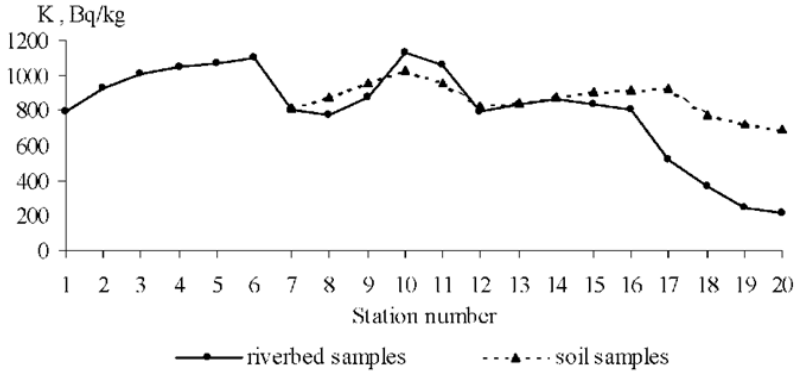

Fig.10. Distribution chart of the content of ^{40}K , Bq/kg along the straightened out studied profile

The specific activity of both radioactive nuclides is within the limits of the contents in uncontaminated soil. A very close correlation is established between the concentrations of ^{232}Th and ^{40}K (fig.11).

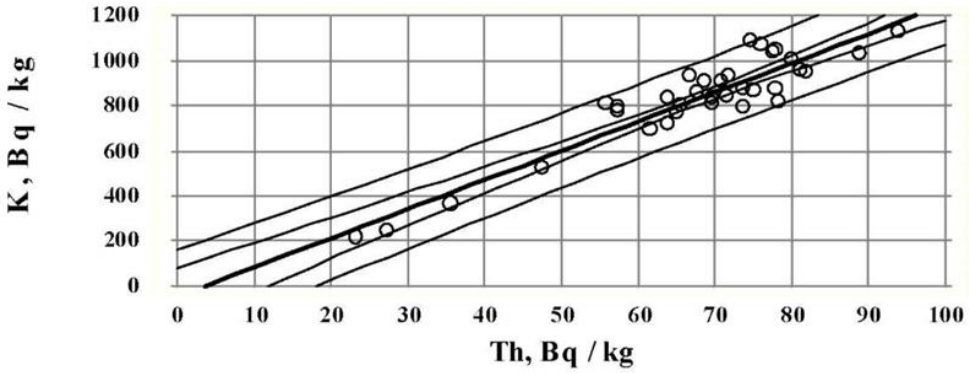

Fig.11. Plot of fitted simple linear regression equation ^{40}K [Bq/kg] = f {^{232}Th [Bq/kg]}
Correlation coefficient = 0,94

There is no correlation between the contents of ^{232}Th and ^{40}K and the specific activity of the three nuclides belonging to the family of ^{238}U. This fact corresponds entirely to the above-analyzed distribution characteristics of the particular radionuclides along the studied profile. The distribution chart of the total specific activity caused by the cumulated effect of all five analyzed radioactive nuclides is illustrated in fig.12.

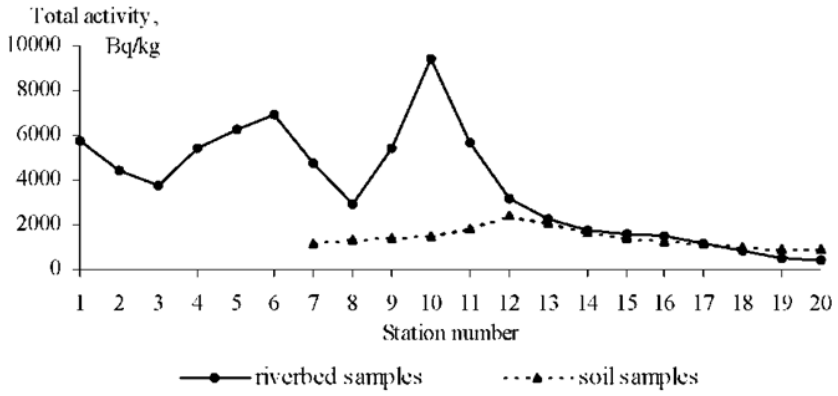

Fig.12. Distribution chart of the total specific activity caused by the cumulated effect of all five analyzed radioactive nuclides along the straightened out studied profile

The total count for the samples of riverbed sediments in the anomalous areas connected to swamping of the river and developing of favorable conditions for adsorption of the radioactive nuclides, is surpassing even more than ten times the average total specific activity. For the soil samples, only in the portion between stations 11 and 13, a better-pronounced migration of the radioactive contaminators is observed. As a result, in this interval the total activity of soil layer 0-10 cm is increased. Near the end of the studied profile, the total count for both types of samples has values close to the average ones.

The results from the analysis of soil samples collected along the three profiles, perpendicular to the river course, are statistically equal to the results from the main samples. The differences are within the limits of the research precision. This fact confirms the conclusion that the transportation of radioactive contaminators is predominantly along the river and that migration in soil has occurred only in areas where water has gone out of the riverbanks.

The distribution chart of the ratio ^{232}Th / ^{238}U along the straightened out studied profile is of definite interest. It is illustrated in fig. 13.

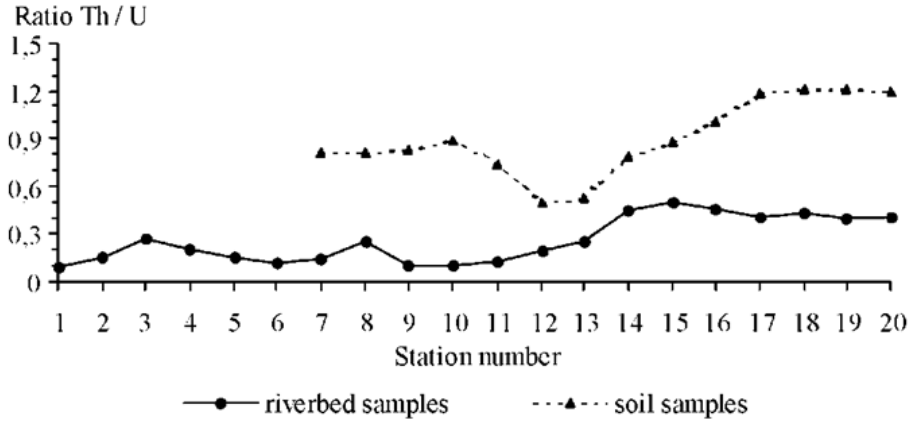

Fig.13. Distribution chart of the ratio ^{232}Th / ^{238}U, along the straightened out studied profile

The values for this ratio in the soil samples surpass without ambiguity the ones in the samples of riverbed sediments. This fact reflects the relatively consistent distribution of thorium in both types of samples and the increased content of uranium in the samples of riverbed sediments.

5.Conclusions

The compound analysis of the results from performed estimation of the content of ^{238}U, ^{226}Ra,^{210}Pb, ^{232}Th and ^{40}K in samples of riverbed sediments and soil samples, taken along the valley of a river, which runs very close to a uranium mine retaining dam, is the reason for the following conclusions:

- Up to a distance of about 6 km away from the retaining dam ^{238}U, ^{226}Ra, and ^{210}Pb have high concentrations and the content in the samples of riverbed sediments is consistently higher than the content in the soil samples. This fact shows that the transportation of the radioactive contaminators is mainly along the river and that migration in soil has occurred only in areas where water has gone out of the riverbanks.
- The observed considerable fluctuations in the contents in the river bed sediments are related to the swamping of the river and the developing of favorable conditions for adsorption of the radioactive nuclides. In these anomalous zones the content of ^{238}U, ^{226}Ra and ^{210}Pb is very high. It surpasses the average specific activity of soil layer 0-10 cm in Bulgaria about 4-5 times for uranium, 10-12 times for radium and 20-30 times for lead respectively.
- Receding at a greater distance from the retaining dam, the content of ^{238}U, ^{226}Ra and ^{210}Pb is displaying a consistent trend towards decrease. A very close correlation is established between the contents of the three radioactive nuclides belonging to the family of ^{238}U. The specific activity and respectively the content of ^{226}Ra and ^{210}Pb are higher than the values in the case of radioactive equilibrium with ^{238}U . This anomaly is due to the fact that uranium first of all has already been separated in the processing, and second - it forms easily soluble complex compounds and in such a way is transported away by the running water.
- The observed specific activity of ^{232}Th and ^{40}K is within the limits of the values for uncontaminated soil. A very close correlation is established between their concentrations. There is no correlation between the contents of ^{232}Th and ^{40}K and the specific activity of the three nuclides belonging to the family of ^{238}U. This fact corresponds entirely to the analyzed distribution characteristics of the particular radionuclides along the studied profile.
- The total count for the samples of riverbed sediments in the anomalous areas connected to swamping of the river and developing of favorable conditions for adsorption of the radioactive nuclides, is surpassing even more than ten times the average values for the total specific activity in the territory of Bulgaria.
- The ratio ^{232}Th /^{238}U in the soil samples surpasses without ambiguity the one in the samples of riverbed sediments. This fact reflects the relatively consistent distribution of thorium in both types of samples and the increased content of uranium in the samples of riverbed sediments.

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