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CONCENTRATION OF RADIOACTIVE ELEMENTS (U, Th) IN COMPONENTS OF NATURAL ENVIRONMENT

The research resulted from the necessity of constant radio-ecological monitoring of the territory of Pavlodar region being under the influence of natural and technological factors. The tense radioecological situation in the region is due to the presence of uranium mining areas, rocks with increased radioactivity (granites, granosienites), geological exploration, groundwater and underground with a high content of radionuclides, and polygon activities. The purpose of the work is to assess the content of radioactive elements in natural environments (water, salt deposits, the aboveground part of wormwood, leaves of poplar, children's hair) of Pavlodar region of the Republic of Kazakhstan. Objects: components of natural environment – drinking waters (groundwaters and surface waters), salt deposits (limestone), the aboveground part of wormwood (*Artemisia absinthium*), leaves of black poplar (*Populus nigra*), children's hair. The main methods of the research of chemical content of environments studied are the instrumental neutron activation analysis and inductively-coupled plasma-mass spectrometry. Content of uranium and thorium in five components of environments of Pavlodar region were studied. Halos of higher concentration of radioelements are of natural and technological genesis. The uranium content in drinking waters is mainly typical for the central and northern parts of the region. The concentration of the studied radioelements in salt deposits of drinking waters is typical for the northern part of the region. On the territory of the Lebyazh, May and Bayanaul areas, there are an increased content of uranium in the leaves of the black poplar. Human hair is characterized by a predominant uranium content in the southern and eastern parts of the region. Higher content of thorium in all components of natural environments, except for leaves of black poplar, is indicated in the northern part of the region, which results from the geological component. Thorium-uranium ratio varies from 0,0003 to 2,3.

Key words: uranium, thorium, radioactivity, components of natural environment, regional features.

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Табиғи орта компоненттеріндегі радиоактивті элементтердің (U, Th) концентрациясы

Зерттеу табиғи және техногендік факторлардың әсерінен болатын Павлодар облысының аумағын радиоэкологиялық бағалау қажеттілігімен анықталады. Облыстағы шиеленісті радиоэкологиялық жағдай уран кені аудандарының, радиоактивтілігі жоғары тау жыныстарының (граниттер, граносиениттер), геологиялық барлау жұмыстарының, радионуклидтері жоғары жер асты және жер грунт суларының, полигон қызметінің жүргізілуімен байланысты. Жұмыстың мақсаты – Қазақстан Республикасының Павлодар облысының табиғи орталарындағы (су, тұзды шөгінділер, жусан, жапырақтар, балалар шаштары) радиоактивті элементтердің құрамын бағалау. Нысандары: қоршаған орта компоненттері – ауыз су (жер асты және жер үсті), тұзды шөгінділер (қак), жусан (*Artemisia absinthium*), қара терек жапырақтары (*Populus nigra*), балалар шаштары. Зерттелген орталардың химиялық құрамын зерттеудің негізгі әдістері аспаптық нейтронды активтендіру талдауы, индуктивті байланысқан плазмамен масс-спектрометрия болып табылады. Павлодар облысының қоршаған ортасының бес құрамдас бөлігіндегі уран мен торийдің құрамы зерттелді. Радиоэлементтердің жоғары концентрациясының аймағы табиғи-техногендік генезиске ие. Ауыз судағы уранның мөлшері негізінен облыстың орталық және солтүстік бөліктеріне тән. Ауыз судың тұзды шөгінділеріндегі зерттелетін радиоэлементтердің

солтүстік бөлігіне тән. Лебязі, Май және Баянауыл аудандарында қара терек жапырақтарында уранның жоғарылауы байқалады. Адам шашы облыстың оңтүстік және шығыс бөлігінде уранның басым болуымен сипатталады. Қара терек жапырақтарын қоспағанда, табиғи ортаның барлық компоненттерінде торий мөлшерінің жоғарылауы облыстың солтүстік бөлігінде байқалады, бұл геологиялық құрамдас бөлікке байланысты. Торий-уран арақатынасы 0,0003 пен 2,3 аралығында.

Түйін сөздер: уран, торий, радиоактивтілік, табиғи орта компоненттері, аймақтық ерекшеліктері.

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Концентрация радиоактивных элементов (U, Th) в компонентах природной среды

Исследование обусловлено необходимостью радиоэкологической оценки территории Павлодарской области, находящейся под влиянием природных и техногенных факторов. Напряженная радиоэкологическая ситуация в области обусловлено наличием урановодородных районов, горных пород с повышенной радиоактивностью (граниты, граносиениты), геологоразведочных работ, грунтовых и подземных вод с высоким содержанием радионуклидов, деятельности полигона. Целью работы является оценка содержания радиоактивных элементов в природных средах (вода, солевые отложения, полынь, листья, волосы детей) Павлодарской области Республики Казахстан. Объекты: компоненты окружающей среды – питьевые воды (подземные и поверхностные), солевые отложения (накипь), полынь горькая (*Artemisia absinthium*), листья тополя черного (*Populus nigra*), волосы детей. Основными методами исследования химического состава изученных сред являются инструментальный нейтронно-активационный анализ, масс – спектрометрия с индуктивно связанной плазмой. Изучено содержание урана и тория в пяти компонентах окружающей среды Павлодарской области. Ореолы повышенных концентраций радиоэлементов имеет природно-техногенный генезис. Содержание урана в питьевых водах преимущественно отмечается в центральной и северной частях области. Концентрация исследуемых радиоэлементов в солевых отложениях питьевых вод характерно для северной части области. На территории Лебяженского, Майского и Баянаульского районов отмечается повышенное содержание урана в листьях тополя черного. Для волос человека характерно преимущественное содержание урана в южной и восточной частях области. Повышенное содержание тория во всех компонентах природных сред, за исключением листьев тополя черного, отмечается в северной части области, что обусловлено геологической составляющей. Торий-урановое отношение колеблется от 0,0003 до 2,3.

Ключевые слова: уран, торий, радиоактивность, компоненты природной среды, региональные особенности.

Introduction

Radiological situation on the territory of Kazakhstan has been studied from the end of the '40s and has been concerned with the increased need for uranium exploration. A row of deposits such as Kurday, Botaburum, Kyzylsay, Kubasadyr, Manybay, Zaozernoye and Ishim, were discovered in the '50s of the past century. First customized uranium explorations also allowed discovery of two uranium ore districts. In the second period (the '60s) a row of undertaken expeditions allowed formation of the idea of a possible connection between uranium mineralization and boundaries of ground and formation oxidation zones. The third period of works had been

lasted from the end of the '60s to the '90s, which is concerned with implementation of a method for development of water-flooded deposits, which was called as the in-situ leaching (ISL).

To date extensive research [1,2], including radio-hydrolytic survey [3] based on the study of water sources and bottom sediments, has been conducted on the territory of the Republic of Kazakhstan. Maps of radionuclide activity, reflecting radio-ecologic reality defined by the presence of six uranium ore provinces (Shu-Sarysu, Ili, Syrdarya, North Kazakhstan, Betpakdala-Shu-Ili, Mangistau) [4], activity of former Semipalatinsk testing site [5], higher radioactivity of water sources [6], activity of research reactors, have been constructed.

Radiologic research conducted [7] allowed zonation of the territory of Kazakhstan regions by the sum of radiation factors and identification of areas for the detailed monitoring. Pavlodar region was not an exception in such assessment of radiological situation. The tense radiological situation in the research territory is formed by:

- areal contamination by artificial radionuclides from numerous nuclear explosions on the territory of former Semipalatinsk testing site and a number of other nuclear incidents;

- high radiation background in the regions accommodating uranium ore districts and separate areas of development of rock with higher radiation;

- presence of sites of radiation poisoning in the form of radioactive dumps of rock, generated in geological prospecting of uranium, polymetals and rare earth occurrences;

- wide distribution of natural groundwaters and surface waters with high content of radionuclides, often attached to cavities of salt lakes.

Since the research has been involved identification of radionuclide activity (Cs_{137} , U_{238} , Th_{234} , $^{228}Ra_{226}$, K_{40}) only in two components of natural environments (groundwaters and surface waters, sediments), this did not allow complex radio-ecologic assessment, which must include research of various objects of environments, notably research of chemical composition of bio-substrates, because exactly the human body is under the polyfactor influence of environment.

Material and methods of research

Territory of the research. The research was conducted on the territory of Pavlodar region, located in the northeastern part of the Republic of Kazakhstan. The total area of the region amounts to 124 755 km².

In metallogenic zonation by uranium on the territory of the Republic of Kazakhstan Turgay-Pavlodar uranium-bearing (potentially ore-bearing) one occupies the northwestern quarter of the region. The leading type of uranium deposits is exodiagenetic in paleo-valleys (ground-infiltration). Schiderty ore occurrence in the area of Tortuy settlement is known within the belt on the territory of the region.

Spurs of the Kazakh hills, which are a peneplain ancient folded mountain country, are located in the southeast of the region. Hill lowlands are isolated in the south of the hills – Bayanaul (maximal height is 1022 m above sea level) and Kyzyltu (maximal height is 1055 m above sea level). Alaskite granites of Bayanaul mountains, where Bayanaul small uranium ore occurrence is, possess higher radioac-

tivity among them. Massifs of alaskite and normal granites, rhyolites with specific activity of uranium (62-125 Bq/kg), thorium (41-123 Bq/kg) are located in the area of Burkitty settlement, Zheltau mountain, the right bank of Aschisu. Small uranium and thorium ore occurrences are also located here.

Ulken-Tuz uranium ore node, located in the northwestern end of Chingiz lifting, on the junction with Central Asian fracture zone, is attached to the northern part of a meridionally stretched volcanic-tectonic structure, folded by volcanics of neutral-acid composition and sediments. Here uranium mineralization is identified in greisen (Shommakol ore occurrence), in albites and albitized rock (Karakuduk, Aschikuduk, Erirbaysor, Ushkara); nasturan-sulfide and molybdenum-nasturan mineralization is identified in beresites and argillites (Ulken-Tuz, Karazhir, Tleumbet, Batyrsha). Ore occurrences of Ulken-Tuz uranium ore node also tend to ways to surface of granites, granosienites [8].

Maykuben brown coal basin is located in the southern part of the region between Shoptykol and Birlik settlements, and is widely stretched on the length of 60 km. Shoptykol large deposit is developed, Tamdy and Taskuduk small deposits are present. In the west of the region, on the left bank of the Olenty river Koitas small deposit is known. Brown coal can present potential radio-ecological hazard in oxidized formations, exposed at surface or revealed by mine workings [9].

North Kazakhstan province of alluvial titanium content occupies the entire north-eastern half of Pavlodar region. Two small iron-titanium-zirconium deposits are known. One is on the left bank of Olenty, another, Druzhba, is to the north-west of Taldy settlement, attached to Druzhba mass of middle and late Eocene [10,11]. Titanium-zirconium deposits possess higher radioactivity due to thorium containing minerals.

On the territory of Pavlodar region there are no uranium mining and processing enterprises, nevertheless there are sources of radioactive contamination. The main source is former multi-year activity of Semipalatinsk nuclear testing site from 1949 to 1989.

Methods of the research. The work is based on the extensive factual material – 450 samples on the territory of Pavlodar region. Chemical composition of samples (limescale, leaves, the above-ground part of wormwood, children's hair) is identified by the instrumental neutron activation method in nuclear geochemical laboratory of "Uranium geology" International Innovative Scientific and Educational Center named after L.P. Rikhvanov on the base

of IRT-T research nuclear reactor of National Research Tomsk Polytechnic university (certificate of accreditation №RA.RU.21AB27 dated 27.05.2015, personnel, conducting analytical research – A.F. Sudyko, L.V. Bogutskaya). Content of radioactive elements in drinking waters was defined by the method of inductively-coupled plasma-mass spectrometry.

Objects of the research. Salt deposits of drinking waters (limescale) were collected in accordance with patent № 2298212 [12] from the walls of enamelware by the method of accurate knocking with a stainless steel knife. Sample preparation: limescale was dried at room temperature, rubbed to powder in an agate mortar, packed on 100 mg.

Drinking water was collected in accordance with GOST R 51592-2003 [13] in the amount of 0.5 liters in containers from polymer materials.

Leaves of black poplar were collected in accordance with the method [14] on 50-100 g without stalks from the lower part of a crown from the branches growing in various directions at a height of no more than 2 meters. When collecting samples of wormwood, its above-ground part was used. Gen-

eral sample preparation of vegetation: washing with distilled water, drying at room temperature, manual grinding, rubbing to powder in an agate mortar, weighing, ashing in a muffle furnace (550 °C), ash weighing, packing on 100 mg.

Children's hair was collected by the method of envelope from five points of head [15]. Sample preparation: washing with distilled water, drying, grinding, packing in packets from foil on 100 mg.

Results and discussion

Statistical data on content of uranium and thorium, uranium-thorium ratio in various components of natural environments are presented in table 1. Analysis of the data received demonstrated unequal distribution of the elements studied in most environments on the territory of the region (Table 1). Homogeneity of thorium distribution in leaves of black poplar in Pavlodar region indicates accordance with the law of normal distribution. Thorium-uranium ratio in components of natural environments varies in a sufficiently wide range from 0,0003 to 2,3.

Table 1 – Statistical data U, Th, Th/U in components of environment on the territory of Pavlodar region

Components of natural environment	Content of uranium $\frac{X \pm \lambda}{\min \dots \max} V$	Content of thorium $\frac{X \pm \lambda}{\min \dots \max}$	Thorium-uranium ratio Th / U
Drinking water (mg/l)	$\frac{0,015 \pm 0,01}{0,000001 \dots 0,13} 234$	$\frac{0,000004 \pm 0,0000007}{0,000001 \dots 0,000009} 58$	0,0003
Limescale (mg/kg)	$\frac{33,5 \pm 1,7}{0,01 \dots 479} 72$	$\frac{0,15 \pm 0,03}{0,001 \dots 4,9} 263$	0,005
Leaves of black poplar (Populus nigra) (mg/kg)	$\frac{0,4 \pm 0,04}{0,01 \dots 1,1} 59$	$\frac{0,37 \pm 0,05}{0,02 \dots 1,9} 100$	0,89
Wormwood (Artemisia absinthium) (mg/kg)	$\frac{0,8 \pm 0,1}{0,002 \dots 2,9} 84$	$\frac{1,8 \pm 0,2}{0,3 \dots 5,9} 75$	2,3
Children's hair (mg/kg)	$\frac{0,29 \pm 0,02}{0,02 \dots 1,2} 79$	$\frac{0,01 \pm 0,001}{0,001 \dots 0,096} 101$	0,03

Note: in numerator – arithmetic mean \pm standard error, in denominator – minimal and maximal values.

Natural waters. On the territory of the research groundwaters and surface waters are predominantly hydrogen carbonate and sulfate-hydrogen carbonate, sodium-calcium with mineralization 750 mg/l. Average content of uranium in natural waters of Pavlodar region amounts to 0,015 \pm 0,01 mg/l. Maximally high concentrations of radionuclides are not-

ed in waters of Pavlodar district, predominantly on the right bank with the epicenter in Naberezhnoye settlement. It should be mentioned that according to the research conducted previously in this settlement maximal content of uranium was recorded in the above-floodplain terrace of the right bank of Irtysh in 2004.

The given part of the region was allocated by the researchers as Ertis-Kulundy potentially radioecological danger zone. Concentration of uranium in sediments amounted to 50-74 Bq/kg. Here it should be noted that zircon-ilminite deposits, which possess higher radioactivity, localize in the northeastern part of the region, in Paleogene mass in the valley of the Irtysh River.

Spatial distribution of thorium in drinking waters is preferentially noted in the northeastern part of the territory the research. Higher concentrations of uranium and thorium in natural waters affected composition of salt formations (limestone).

Limestone. Salt deposits (limestone), forming on the inner walls of heating devices in the process of boiling, are sufficiently informative environment,

used in ecological-geochemical research [16-18]. Average concentration of uranium in salt formations of Pavlodar region amounts to $33,5 \pm 1,7$ mg/kg. Maximal content of radionuclides is identified in salt formations of Terenkol district ($43,2 \pm 7,7$), (picture 1), which is in good accordance with the data received by specialists of "Volkovgeology". The text continues here (Figure 1).

Higher concentrations of uranium in limestone ($39,7 \pm 6,1$) of the southern part of the region are of natural and artificial genesis, which is connected with development of deposits of Ekibastuz and Maykuben brown coal basins. Content of uranium in Maykuben coal amounts to $5,0 \pm 2,0$ mg/kg, which exceeds the Clarke level more than 2.1 times [19].

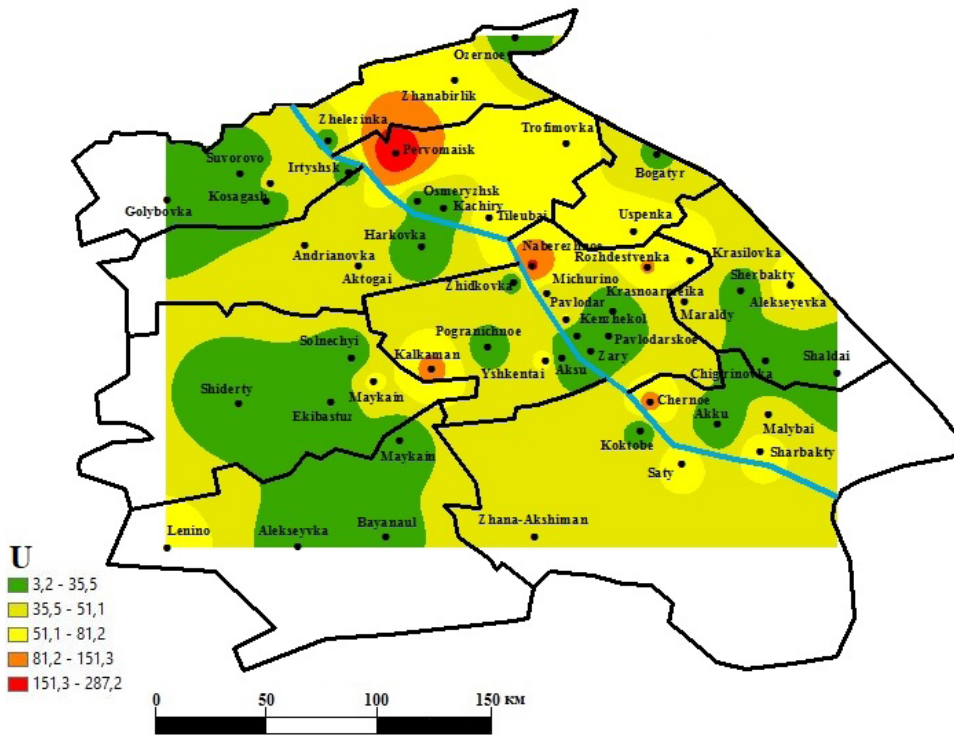


Figure 1 – Schematic map of distribution of uranium in salt formations (limestone) of Pavlodar region, mg/kg

Zhana Akshiman settlement (the southern part of the region) with concentration 46,5 mg/kg in limestone is located on the territory of Ulkentuz uranium ore node at the distance of 7 km from Ulkentuz ore occurrence and within 15 kilometers from Tleumbet and Karazhir ore occurrences. Ore occurrences tend to ways to surface of granites, granosienites, which possess higher radioactivity.

It should be noted that in the southern part of the region higher content of radionuclides in salt deposits is possibly connected with activity of former Semipalatinsk site (1949-1989), accounting for more than 400 explosions.

When comparing the data received with the research conducted previously on the territory of Tomsk region, Baykal area, Altay, the Republic

of Bashkortostan [17,18] it was found that Pavlodar limestone is different in maximal content of uranium. Average content of thorium in limestone amounts to $0,15 \pm 0,03$, minimum – 0,001 mg/kg, maximum – 4,9 mg/kg (Figure 2).

Distribution of thorium in salt formations preferentially in the northern part of the territory of the research. Here high concentrations of rare earth elements are observed, which is probably connected

with formation of rare metal – rare earth occurrences in waste mantle in depths of water or with distribution of titanium-zirconium deposits.

Thorium-uranium ratio in salt deposits (limestone) of Pavlodar region is one of the low – 0,005, for example, it amounts to 0,008 in Baykal area, to 0,2 in Altay, to 0,05 in Tomsk region, and only in the Republic of Bashkortostan this indicator is even lower 0,002 [17,18].

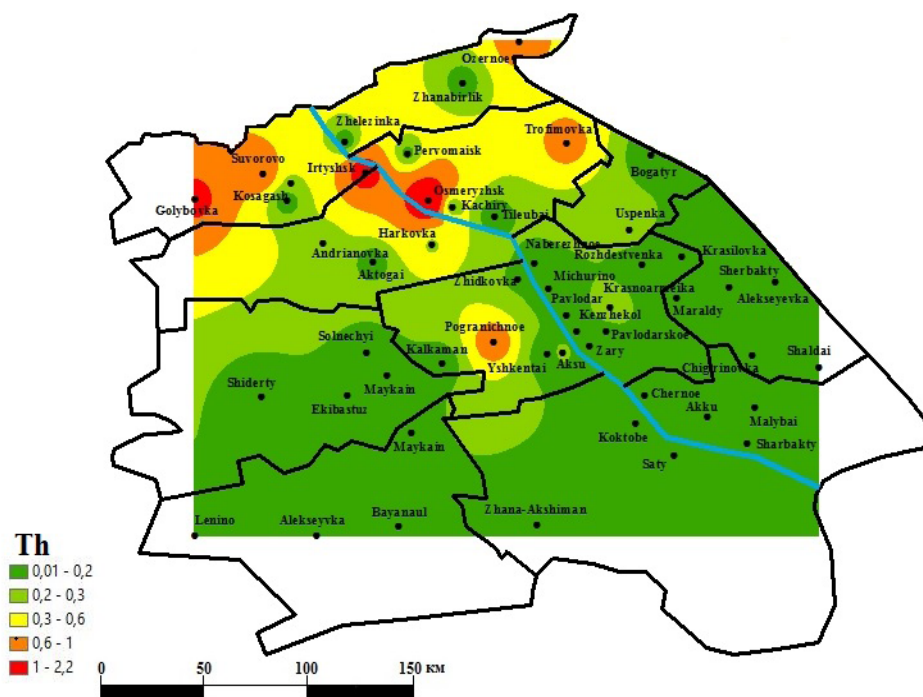


Figure 2 – Schematic map of distribution of thorium in salt formations (limestone) of Pavlodar region, mg/kg

Leaves of black poplar and the above-ground part of wormwood. As is known, plants have the feature of selective concentration of elements, which allows their usage in biochemical research of urbanized territories [20-21]. Thus, for example, leaves of trees can serve as a so called natural tablet, which not only accumulates contamination, but also reflects intensity of the anthropogenic impact on ecosystems [22].

The in-depth analysis of content of radioactive elements in the ash of leaves of black poplar revealed three settlements on the territory of Pavlodar region with maximal values of uranium and thorium. These are Akkuly, May and Bayanaul districts with values Th ($0,37 \pm 0,05$ mg/kg) and U ($0,4 \pm 0,04$ mg/kg) in objects of plant origin. Ways of bedrock to surface

are observed in Bayanaul district. In the given settlement alaskite granites of the Bayanaul mountains, where Bayanaul small uranium ore occurrence is, also possess higher radioactivity. On the area of the right bank of the Aydos and Aschysu rivers, massifs of alaskite and normal granites, rhyolites with activity of uranium 62-125 Bq/kg; thorium 41-123 Bq/kg are located. Here small uranium and thorium occurrences are also located. As is known, Bayanaul district includes Maykuben brown coal basin, which is located in the southern part of the region.

A halo of higher content of thorium in leaves of black poplar of May district is possibly connected with underground explosions, which were conducted on former Semipalatinsk testing site (Figure 3).

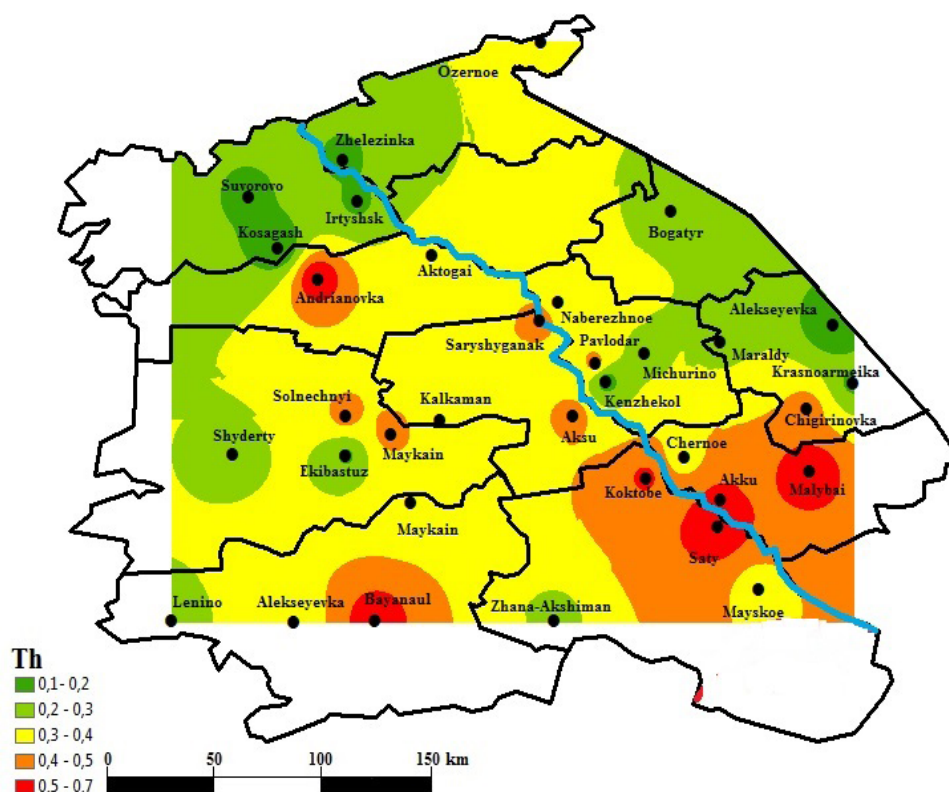


Figure 3 – Schematic map of distribution of Th in the ash of leaves of black poplar on the territory of Pavlodar region, mg/kg

Higher concentration of uranium (0,91 mg/kg) and thorium (2,38 mg/kg) was detected in the ash of the above-ground part of wormwood in Irtyshsk and Pavlodar districts.

The highest content of uranium is noted in samples of vegetation (leaves of black poplar and the above-ground part of wormwood). This is possibly a consequence of location of large coal mines and their dust load.

One of the relevant indicators of industrial sources of contamination is thorium-uranium ratio. In most cases, it is less than 1 or equal to 1, which points to uranium specialization of Pavlodar region. Districts with high Th/U ratio, May, Akku and Aktogay districts, in particular, are singled out. Origin of anomalous zones is connected with glacial erosion of high thorium containing rock and also monacite deposits in Kalba-Narym thorium rare metal ore province, located in East Kazakhstan region along the Irtys'k river.

Thorium-uranium ratio in leaves of black poplar in Pavlodar region amounts to 0,89, by comparison, to 0,42 in Ust-Kamenogorsk, to 0,86 in Taraz, to 0,88 in Aktobe, to 1,78 in Tomsk and Seversk, to

1,88 in Kyzyl, to 0,49 in Krasnokamensk, to 2,88 in Blagoveschensk [21,22].

Human hair. For assessment of modern ecological and geochemical situation of the territory the research human bio-substrates (hair, blood, nails, urine) are used, which are not only instant indicators of supply of chemical elements, but also indicators pointing to the long-term impact on them.

In the study of content of radioelements in children's hair, attention should be paid to high content of uranium in hair of the children living in the territory of Pavlodar, Sharbakty, Akku districts. In general, higher content of uranium and relatively low concentration of thorium in human hair are characteristic of regions of Kazakhstan (North Kazakhstan, Akmola, Pavlodar) by comparison with regions of Russia (Tomsk, Chita, Kemerovo) [23,24].

This points to uranium specialization, which was caused by supply of uranium with drinking water on the example of Pavlodar region.

It should be mentioned that excessive concentration of uranium in children's hair is connected with distribution of zirconium-ilmenite deposits, which are located in the valley of the cross-border Irtys'k

river. This may be reflected in the research of sediments below Pavlodar city according to the data of JSC “Volkovgeology”.

Thorium-uranium ratio in human hair of Pavlodar region (0,03) is considered to be one of the low by comparison with Akmola (0,06), North Kazakhstan (0,06), Tomsk (0,21), Kemerovo (0,01), Chita (0,26) regions [25].

Conclusion

Thus, distribution of uranium and thorium in components of natural environments on the territory of Pavlodar region is unequal.

Halos of higher concentration of uranium in the components of natural environments studied by us correspond to the research conducted previously and confirms allocation of Ertis-Kulundy potentially radio-ecological dangerous zone.

Within Pavlodar region according to geochemical conditions and the radio-ecological situation studied, rich for areal and local sources with anomalous concentration of uranium and thorium (granitoids, carbon-silicon slates, ore nodes of endogenous ore occurrences of uranium, occurrences of thorium – rare metal mineralization of various scale, activity of former Semipalatinsk testing site), districts of region are clearly allocated: Pavlodar, Terenkol, Bayanaul, May, Akkuly.

Thorium-uranium ratio in limescale, leaves of black poplar, human hair is considered one of the low by comparison with settlements of Kazakhstan and Russia.

The data received can be used for ranging of the territory of the region by the complex of radiation factors determining the status of modern radio-ecological situation.

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