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Geochemistry of radioactive elements (U, Th) in coal and peat of northern Asia (Siberia, Russian Far East, Kazakhstan, and Mongolia)

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Geochemistry of radioactive elements in more than 5000 coal and peat samples of Northern Asia has been investigated by quantitative methods, such as the instrumental neutron-activation analysis, the method of delayed neutrons, and the X-ray-fluorescence analysis. The average U content in the coals of deposits and basins of the region ranges from 0.6 to 32.8 ppm and, for Th, from 0.8 to 9.2 ppm. Within the boundaries of basins, deposits and some coal beds lateral and vertical changes of the distribution of radioactive elements have been studied. The high concentrations of U and Th in coal deposits are spatially related to rock blocks within the basin frames, which are enriched in elements, or are connected with a volcanism of a period of coal formation. A consistent changing of the role of pyroclastic material in the radioactive elements accumulation was determined in the direction from the West to the East.

1. Introduction

The strengthening of social and state environmental control at the end of the 20th century and at the beginning of the 21st century has led to toughening the requirements to the environmental safety for the fuel power industry. Coal-fired power plants are traditionally considered as one of the main sources of environmental pollution. Along with the emissions of carbon dioxide, nitrogen and sulfur oxides, coal combustion at power plants causes release of great amounts of potentially toxic and radioactive elements into the biosphere. It is known that coals sometimes include high contents of natural radioactive elements (U, Th, and their decay products) and, in some cases, concentrate considerable resources of the uranium. At the beginning of the "nuclear era", some efforts were made to use U-bearing coals as a source of the U. However, with the discovery of higher-quality raw sources, the interest in U-bearing coals diminished and now they are considered to be potentially dangerous source of radioactive contamination of the environment (Bauman and Horvat, 1981; Bride and Moore, 1978; Eisenbud and Petrow, 1964; Mazuricheva and Kiselev, 2004). The world coal industry focuses on coals with low concentrations of radioactive elements. However, while combusting such coals, the radioactive elements concentrate in the combustion products such as slag and fly ash. The concentration of U and Th in fly ash usually increases three- or four-fold in relation to the original fuel and in some cases a ten-fold increase is observed.

Since the emissions of radioactive elements from thermal power plants into the environment first of all depend on their concentration in the fuel, forecasting of the environment contamination with radionuclides is only possible in terms of the objective knowledge of their concentrations, distribution, and accumulation conditions in the original fuel (coalbeds). Nevertheless, radio-geochemical and radio-ecological studies have been carried out by now at a very limited number of exploited coal basins and deposits, generally in the EU, USA, and Russia.

We conducted systematical investigations of coal basins and deposits in Russia (Siberia and Russian Far East) and over recent years in neighboring states (Kazakhstan and Mongolia) in an effort to fill this gap and to determine the main regularities of the formation of coals with different radio-geochemical characteristics. These issues have been partially considered in the monographs (Arbuzov et al., 2000, 2003, 2008; Arbuzov and Ershov, 2007) and in a number of articles. This work represents new data on the geochemistry of U and Th in coals and peat of Northern Asia and an effort to generalize the extensive knowledge on the radio-geochemistry of coals of the region.

2. Study objects

Radio-geochemical studies have been carried out in Asian part of the Russian Federation, in Mongolia, and partly in Kazakhstan (Fig. 1). Detailed geological–geochemical surveys were conducted in three Siberian basins: Kuznetskiy, Minusinskiy, and Kansko-Achinskiy. The Gorlovskiy, Tungusskiy, Zapadno-Sibirskiy, Ulugchemskiy, and Irkutskiy basins have been studied to a lesser degree, but also have sufficiently representative data. The total number of the analyzed samples in Siberian region is 2933 for coal and 1927 for peat.

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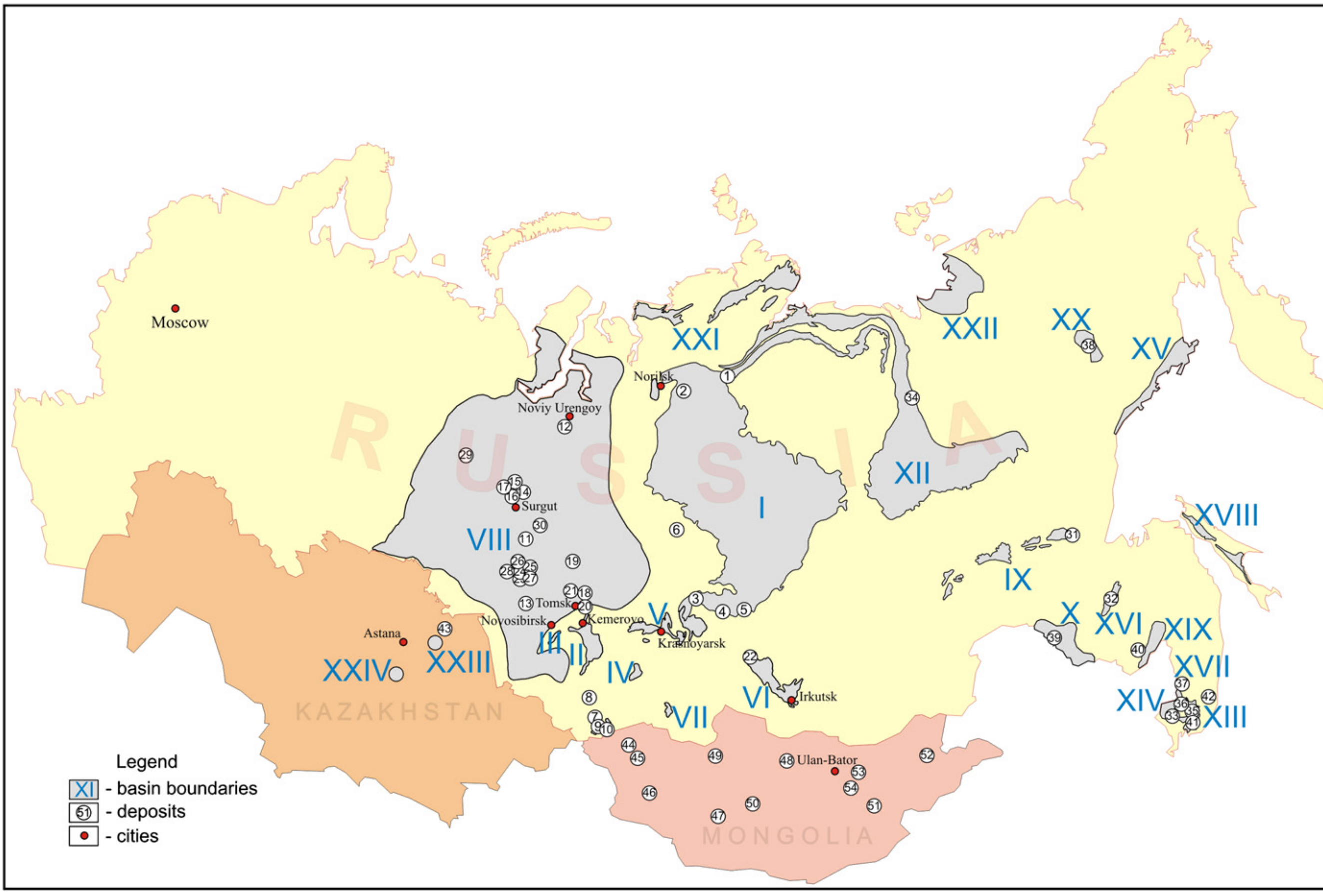

Fig. 1. Location of the studied coal basins and deposits in Northern Asia. Basins: I — Tungusskiy, II — Kuznetskiy, III — Gorlovskiy, IV — Minusinskiy, V — Kansko — Achinskiy, VI — Irkutskiy, VII — Ulugchemskiy, VIII — Zapadno-Sibirskiy, XI — Yuzhno-Yakutskiy, X — Nizhnezeyskiy, XI — Khankaiskiy, XII — Lenskiy, XIII — Partizanskiy, XIV — Razdolnenskiy, XV — Okhotskiy, XVI — Bureinskiy, XVII — Bikino-Ussuriyskiy, XVIII — Sakhalinskiy, XIX — Sredne-Amurskiy, XX — Arkagalinskiy, XXI — Taimyrskiy, XXII — Yano-Omoloyskiy, XXIII — Ekibastuzskiy, XXIV — Karagandinskiy. *Deposits*: 1 — Kayakskoe, 2 — Kayerkanskoe, 3 — Kokuyskoe, 4 — Gavrilovskoe, 5 — Kodinskoe, 6 — Podkamenno-Tungusskoe, 7 — Kuraiskoe, 8 — Pyzhinskoe, 9 — Taldu-Dyurgunskoe, 10 — Balkhash, 11 — Myldzhinskoe, 12 — Urengoyskoe, 13 — Verkhtarskoe, 14 — Kavrinskoe, 15 — V-Trom'eganskoe, 16 — Konitlorskoe, 17 — Ai-Pimskoe, 18 — Tuganskoe, 19 — Kolpashevskoe, 20 — Lagernosadskoe, 21 — Talovskoe, 22 — Tulunskoe, 23 — Tabaganskoe, 24 — Shirotnoe, 25 — Gerasimovskoe, 26 — Luginetskoe, 27 — Severo-Kalinovoe, 28 — Archinskoe, 29 — Yakhlinskoe, 30 — Trassovoe, 31 — Elginskoe, 32 — Urgalskoe, 33 — Lipovetskoe, 34 — Zhiganskoe, 35 — Shkotovskoe, 36 — Pavlovskoe, 37 — Bikinskoe, 38 — Arkagalinskoe, 39 — Sergeevskoe, 40 — Ushumunskoe, 41 — Avangard, 42 — Voznovskoe, 43 — Karazhira, 44 — Nuurs-khotgor, 45 — Hartarvagatai, 46 — Ho'ndlon, 47 — Zeegt, 48 — Saykhan-Ovoo, 49 — Mogoin gol, 50 — Bayanteeg, 51 — Alagtogoo, 52 — Uduunchuluun, 53 — Baganuur, 54 — Tugriknuurskoe.

The Far-East region has been studied to a lesser degree. The Late-Jurassic, Cretaceous, Paleogene, and Neogene coals in this region are younger than in Siberia. The number of coal samples from 14 deposits of this region is 217. In addition, 32 peat deposits on the West Siberian plate, containing about 40% of the world peat resources, were sampled (Arbuzov et al., 2009; Mezhibor et al., 2009).

Radio-geochemical studies in Mongolia were carried out for the first time, with a total of 116 coal samples from 11 deposits of Pennsylvanian, Jurassic, and Cretaceous ages (Mashenkin et al., 2009). A minor data on the Pennsylvanian coals of Kazakhstan were also obtained.

In total, this work is based on the results of quantitative analysis of U and Th in the more than 5200 coal and peat samples and more than 2000 samples of coal-hosting rocks of different deposits.

3. Methods

The sampling of coal beds was carried out by the channel method by interval sampling of coals of different types and coal interbeds in coal open-pits and mines, in natural exposures, and also in core samples. The length of a sampling interval was selected depending on thickness and structure complexity of a bed and ranged from 0.15 to 2.0 m on average. Small-thick coal patches separated by rock bands, rock bands, clastic “dikes”, sulfide inclusions, carbonate concretions, and other mineral formations were studied separately. In certain sections, the detailed sampling in intervals from 0.5 to 10 was made. The spatial variety of the element concentrations was estimated according to the system of sections in a bed.

Several physical nondestructive methods were used to determine radioactive elements (U and Th) directly in coal without preparatory ashing as the latter could result in losses of radionuclides. The gamma-spectrometric measurement of U and Th was made with standard field instrumentation directly in coal open-pit mines. Laboratory measurements of U and Th were carried out at the Nuclear-Geochemical Laboratory (NGL) of the Department of Geoecology and Geochemistry of Tomsk Polytechnic University (TPU) (analysts A.F. Sudyko and V.M. Levitskiy) using the research nuclear reactor IRT-T at the Scientific Research Institute of Nuclear Physics of TPU.

Both the conventional method of instrumental neutron-activation analysis (INAA) for the weight of 100–200 mg and the delayed neutron activation analysis (DNAA) for the weight of 5–10 g was used. DNAA is considered as one of the most accurate and also rapid and high-sensitive methods for the determination of low contents of U (Rikhvanov, 2002). The detection limit of the INNA for U is 0.1 ppm and for Th is 0.1 ppm. DNAA detection limit for U is 0.1 ppm. For a group of samples a parallel determination of U in coal ash was carried out by the laser-luminescent method (detection limit is 0.1 ppm). In addition, *X*-ray-spectral fluorescence was used for the U and Th determination (detection limit for U is 1 ppm and for Th is 2 ppm) (analyst N.A. Charikov). The precision of the results of different analysis methods was satisfactory. For control purposes, the parallel measurements of the radioactive elements in coals and coal ash were carried out with further conversion of their contents in ash to those in the coal and vice versa. The quality of the neutron-activation analysis was controlled using different standards for coal ash and rocks, including the standard CLB-1 (Table 1).

The average contents of U and Th in coals of Siberia was estimated by the method of gradual data averaging. The average contents of U and Th in coal beds were calculated as a weighted mean for the thickness of sampling intervals, at deposits – as the weighted mean for the thickness of beds, at basins – as the weighted mean for the coal mass (resources) at deposits (Tkachev and Yudovich, 1975). A similar method was used in a work on the estimation of mean concentration of U in the coal of China (Ren et al., 2006; Yang, 2007). Unlike this estimation, we have calculated the average content not only for epochs of coal accumulation but also for every basin and deposit.

The estimations of the mean values for the main coal formation epochs in the Russian Far East, Mongolia, and Kazakhstan were made without notice of the coal resources at deposits and basins. In this connection, the obtained data should be considered as preliminary.

4. Results and discussion

4.1. U and Th concentration in coal

The U content in the dry coals of Northern Asia ranges from 0.6 to 0.7 ppm (Karagandinskiy basin, deposits of Sakhalin Island) to 32.8 ppm (deposit Aduunchuluun, Mongolia) (Table 2). The peat of Western Siberia contains 0.4 ppm U (Arbuzov et al., 2009). The weighted mean content of U in the coals of Siberian region, including the immense resources of Western-Siberian basin, is 1.5 ppm (Arbuzov and Ershov, 2007). This number is lower than the present estimation of the mean content of U in the coals of the world (Ketris and Yudovich, 2009) and corresponds to the mean exponential content of U in the brown coals of the world (Bouška and Pešek, 1999). The proximity of the regional mean concentration of U to the mean value for the brown coals of the world correlates well with the larger weight quantity of brown coals of Western-Siberian basin in the total balance of coal resources of the region.

The average estimations of the U and Th contents for other regions have not been conducted due to the insufficient samples, but the examination of the data from Table 2 allows us to speculate that they would be close to the average estimation for Siberia. Thus, for the Russian Far East region, the coals related to general stages of the coal accumulation have an average of 1.7 ppm U. Kazakhstan coals have <1 ppm U. The Pennsylvanian coals of Mongolia have low U concentrations and only younger coal varieties of individual deposits are notable for anomalous high-U concentrations.

The average Th contents range from 0.6 ppm in the modern peat deposits of Western Siberia and 0.8 ppm in Jurassic and Cretaceous coals of Tungusskiy basin to more than 9.2 ppm in the coals of the Mogoin gol deposit (Mongolia). The average Th content for coals of Siberia is 2.4 ppm, lower than the mean content of Th in the coals of the world but higher than the geometric mean for the brown coals of the world according to Bouška and Pešek (1999). The low Th–U ratio, typical for coals, is a function of selective U accumulation by the organic matter of coal. For the eastern regions, a higher average content of Th and higher Th–U ratio than for Siberia was noticed. These data correlate with the estimations of the average content of U (3.3 ppm) and Th (7.6. ppm) in the coals of northern China (Dai et al., 2006). As followed from this data the Th–U ratio of 2.3 is significantly higher than that obtained for Siberia (Table 2). Coals of different age differ in their radio-geochemical characteristics.

4.1.1. Devonian coals

The Devonian period, insignificant on the scale of the coal accumulation, is represented by Barzasskoe deposit of liptobiolithic coals in the northeastern part of Kuznetskiy basin and by the Mountain Ubrus deposit in the northeastern part of the South-Minusink depression. A special feature of this stage of the coal formation is the high content of radioactive elements in the coal. Moreover, if the Th content slightly exceeds the mean content of Th in the coals of the world (Ketris and Yudovich, 2009) and can be conditioned by the high ash yield of the coals, then the U content is significantly higher than the mean content of U in the coals of the world. Similar contents of Th in coals of both deposits can be connected with a source area of the same type of the regions of the Devonian peat accumulation. Both deposits are associated with Devonian volcanism, a possible source of U and Th in the coalbeds (Rikhvanov, 2002).

In addition, the beds of liptobiolithic coals overlay Lower Devonian volcanogenic deposits. Mountain Ubrus coals occur among sandy-siltstone sediments in close proximity to the enclosing volcanics of different composition.

The occurrence of liptobiolithic coals among products of the clay hydrolysis of volcanic tuffs suggests the syngenetic nature of Th and U accumulation. Clay, coal-bearing beds, and numerous intrastratal clay interbeds were limited to the additional penetration of radioactive elements in a coal bed at the diagenesis and catagenesis. The Mountain Ubrus coals occurring among permeable sandy-siltstone series are defined by an anomalously high U concentration and lower Th–U ratio than the coals of Barzasskoe deposit. Probably, a more permeable coal-bearing stratum made it possible for the aquatic input of U into the coal bed.

4.1.2. Pennsylvanian-Permian coals

Carboniferous-Permian coal-bearing deposits have been noted from western Mongolia in the south to Taimyr in the north, and from Karagandinskiy basin of Kazakhstan in the west to Tungusskiy basin in the east. Late Paleozoic coals were also detected in the basement of Western Siberian Plate. A specific feature of the Late Paleozoic coal accumulation is the wide distribution of the products of volcanic

Table 1
Determination of uranium and thorium in standard samples (ppm).

Standard sample	Uranium, ppm		Thorium, ppm	
	Certificate data	NGL TPU	Certificate data	NGL TPU
Coal ash ZUK-1 GSO 7125-94	3.3 ± 0.4	3.2 ± 0.71	5.8 ± 1	5.6 ± 0.23
USGS CLB-1	0.55	0.55	1.4	1.62
Marine sediments IAEA SD-M-2/TM	2.49 (1.44–3.5)	2.27 ± 0.19	8.15 (7.2–9.1)	8.69 ± 0.29
Marine sediments IAEA 315	3.2(2.4–3.5)	3.47 ± 0.17	6.64 (6.24–6.92)	6.5 ± 0.35
Baikal silt BIL-1 GSO 7126-94	12 ± 1.1	12 ± 0.83	12.7 ± 1.3	12.7 ± 0.34
Garnet-biotite plagiogneiss GBP-1	0.8 ± 0.1	0.72 ± 0.14	11.3 ± 1.5	11.2 ± 0.4
Dark shale GSL-1	1.65 ± 0.23	1.8 ± 0.2	7.1 ± 1.1	7.3 ± 0.45
Rhyolite JR-1	8.88	9.4 ± 1.1	26.5	26.7 ± 0.84
Granodiorite JG-3	2.21	2.6 ± 0.71	8	8.28 ± 0.36

Table 2
Uranium and thorium contents in coals and coal ash of Northern Asia (ppm).

Coal basin, deposit	Number of samples	A^d,%	Content of elements, ppm				Th/U
			Coal		Ash*		
			U	Th	U	Th	
Siberian region							
Devonian coals							
Barzasskoe	14	32.5	3.2 ± 0.8	3.9 ± 0.4	9.8	12	1.2
Ubrusskoe	6	49.2	27.2 ± 1.0	3.1 ± 1.0	55.3	6.3	0.1
Average	20	35.3	7.2 ± 0.8	3.8 ± 0.5	20.4	10.8	0.5
Carboniferous-Permian coals							
Gorlovskiy	24	7.0	1 ± 0.1	1.9 ± 0.2	14.3	27.1	1.9
Kuznetskiy	1350	13.5	2.4 ± 0.2	3.3 ± 0.2	17.8	24.4	1.4
Minusinskiy	490	16.9	2.4 ± 0.3	3.0 ± 0.2	14.1	17.9	1.3
Tungusskiy	35	12.0	3.3 ± 0.8	3.7 ± 0.6	27.5	30.8	1.1
Taimyrskiy	10	Not determined	3.2 ± 0.8	2.1 ± 0.4	–	–	0.7
Kuraiskoe	12	25.2	1.1 ± 0.2	4.2 ± 0.7	4.4	16.7	3.8
Average	1921	12.4	3.1 ± 0.7	3.5 ± 0.5	24.8	27.9	1.1
Mesozoic coals							
Pyzhinskoe	6	6.5	0.95 ± 0.3	0.9 ± 0.4	14.6	13.8	0.9
Kansko-Achinskiy	517	9.8	3.2 ± 0.7	1.0 ± 0.2	32.7	10.2	0.3
Irkutskiy	117	14.5	2.7 ± 0.5	4.1 ± 1	18.5	28.5	1.5
Ulugchemskiy	45	9.3	1.2 ± 0.4	1.2 ± 0.2	12.9	12.9	1.0
Zapadno-Sibirskiy	172	10.6	1.2 ± 0.1	2.4 ± 0.2	11.3	22.6	2.0
Tungusskiy	30	12.6	2.1 ± 0.5	0.8 ± 0.2	16.7	6.3	0.4
Kuznetskiy	3	17.3	2.1 ± 1.0	2.2 ± 0.5	12.1	12.7	1.0
Average	890	10.5	1.2 ± 0.3	2.3 ± 0.3	11.4	21.9	1.9
Paleogene coals							
Zapadno-Sibirskiy	73	30.7	4.6 ± 0.4	3.5 ± 0.2	15.0	11.4	0.8
Taldu-Dyurgunskoe	29	19.8	1.7 ± 0.7	1.1 ± 0.1	8.6	5.6	0.6
Average	102	30.7	4.6 ± 0.4	3.5 ± 0.2	15.0	11.4	0.8
Peat							
Zapadno-Sibirskiy	1927	7.3	0.4 ± 0.1	0.6 ± 0.1	5.1	8.5	1.7
Average for Siberia	4860	11.5	1.5 ± 0.4	2.4 ± 0.4	13.0	20.9	1.6
Russian Far East region							
Late Jurassic — Lower Cretaceous coals							
Elginskoe	47	18.4	1.2 ± 0.1	3.7 ± 0.4	6.7	25.5	3.8
Urgalskoe	57	24.1	2.9 ± 0.3	8.1 ± 0.6	120	34	2.8
Lipovetskoe	4	32.7	2.2 ± 0.2	7 ± 1.1	6.7	21.5	3.2
Average	108	22.7	1.7 ± 0.3	5.1 ± 1.4	7.6	22.6	3.0
Paleogene — Neogene coals							
Shkotovskoe	9	20.8	1.4 ± 0.3	3.6 ± 1.2	6.7	17.4	2.6
Pavlovskoe	27	12.7	2.7 ± 0.5	6.6 ± 1.0	20.9	52.1	2.5
Bikinskoe	16	17.4	1.3 ± 0.4	3.8 ± 0.9	7.6	22.0	2.9
Ushumunskoe	9	10.1	1.0 ± 0.2	2.4 ± 0.8	10.3	23.6	2.3
Yano-Omoloyskiy	16	33.0	2.0 ± 0.4	3.2 ± 0.6	6.0	9.8	1.6
Sakhalinskoe	14	16.2	0.7 ± 0.2	2.0 ± 0.3	4.3	12.5	2.9
Voznovskoe	6	18.1	1.8 ± 0.6	4.4 ± 1.4	9.9	24.6	2.5
Sergeevskoe	7	15.8	1.2 ± 0.2	3.7 ± 0.9	7.6	23.4	3.0
Average	104	18.8	1.7 ± 0.3	3.9 ± 0.7	8.9	20.9	2.3
Kazakhstan							
Carboniferous coals							
Karagandinskiy	3	9.8	0.6 ± 0.1	1.1 ± 0.3	6.4	11.4	1.8
Ekibastuz	12	31.8	0.9 ± 0.4	1.1 ± 0.1	2.5	3.1	1.3
Average	15	20.9	0.8 ± 0.4	1.1 ± 0.3	3.8	5.3	1.4
Jurassic coals							
Karazhira	7		0.5 ± 0.2	1.0 ± 0.2			2.0
Mongolia							
Carboniferous coals							
Nuurs-khotgor	31	16.9	1.8 ± 0.3	4.6 ± 0.9	10.5	27.2	2.6
Hartarvagatai	10	18.7	1.0 ± 0.1	2.7 ± 0.3	6.0	14.3	2.6
Ho'ndlon	7	–	1.0 ± 0.1	2.6 ± 0.5	–	–	2.7
Zeegt	10	–	1.3 ± 0.4	2.0 ± 0.3	–	–	1.5
Average	58	17.8	1.3 ± 0.2	3.0 ± 0.6	7.2	16.7	2.3
Jurassic coals							
Saykhan-Ovoo	6	9.7	3.7 ± 1.3	2.2 ± 0.4	38.8	23.2	0.6
Mogoin gol	15	–	18.3 ± 3.0	9.2 ± 1.7	–	–	0.5
Bayanteeg	8	–	22.0 ± 13	7.2 ± 0.9	–	–	0.3
Average	29	–	14.7 ± 5.6	6.2 ± 2.1	–	–	0.4
Lower Cretaceous coals							
Alagtogoo	10	–	3.0 ± 1.1	4.6 ± 0.7	–	–	1.5
Aduunchuluun	10	11.1	32.8 ± 5.5	1.0 ± 0.1	296	9.2	0.03
Baganuur	2	8.4	3.7	1.6	44.2	13.0	0.4
To'grognuur	7	–	0.7 ± 0.1	1.5 ± 0.2	–	–	2.1

(continued on next page)

Table 2 (*continued*)

Coal basin, deposit	Number of samples	A^d,%	Content of elements, ppm				Th/U
			Coal		Ash*		
			U	Th	U	Th	
Lower Cretaceous coals							
Average	29	9.8	10.1 ± 7.6	2.2 ± 0.8	103	22.3	0.2
Clarke for coals**	8400		2.4	3.3	16	21	1.4

(–) No data.
* Calculated over the ash.
** By Ketris and Yudovich, 2009.

activity in the coals and coal-bearing strata. They are represented by altered ash tuffs (tonsteins) and dispersed pyroclastic material (Van, 1972). Rarely, the traces of ashfalls have been fixed in the form of thick beds of bentonitic clays. The ash material caused a slightly high radio-geochemical background of the relevant coal beds, distinctly influencing the geochemical background of Th. Probably, this explains a relatively moderate content of Th in the Late Paleozoic coals in the whole territory of Siberia and Mongolia in spite of the great variety of the composition of source areas of the coal basins and deposits. Only the Kazakhstan deposits are distinct in low contents of U and Th. In many cases, the role of the terrigenous-aquatic accumulation of radioelements in coals gives way to the role of ash falls. The analysis of world published data suggests that the great influence of the volcanic matter on the formation of the radio-geochemical background of the Late Paleozoic coals is a worldwide characteristic (Dai et al., 2003, 2008, 2011; Guerra-Sommer et al., 2008; Hower et al., 1999; Lyons et al., 1992; Mardon and Hower, 2004; Zhou et al., 2000; Arbuzov and Ershov, 2006).

4.1.3. Mesozoic coals

The Mesozoic stage of coal formation includes several periods, including Triassic, Lower-Middle-Jurassic, Late-Jurassic, and Cretaceous. By the scale of its occurrence and coal resources in the region, it surpasses the Late Paleozoic period of coal accumulation. The largest coal resources of this age are concentrated in the sedimentary cover of Western Siberian Plate and in the Lenskiy coal basin. The radio-geochemical characteristic of the Mesozoic coals in the Siberian region is generally determined by the composition characteristics of source areas of the coal formation regions and by the landscape-climatic conditions of the formation of ancient peat lands (Arbuzov and Ershov, 2007). Volcanism, which probably took place that time, did not play a significant role in the U and Th accumulation in the Mesozoic coals of Siberia as it did in the Paleozoic coals (Van, 2001). For the coals of the Russian Far East and Mongolia, where the volcanic activity was identified from the Mesozoic by present time, the role of ash material was significantly higher (Chekryzhov, 2009). In this connection the distribution of uranium and thorium in the coals of these regions is rather irregular.

The average content of U in the Mesozoic coals ranges from 1.2 ppm in the Elginskoe deposit (South-Yakutskiy basin) to 32.8 ppm in the Aduunchuluun deposit (Mongolia). A significant dispersion of U in coals is not only characteristic for individual deposits but for the individual parts of deposits or basins. In addition, in large basins, U often has an evident connection with blocks of rocks with a certain composition, for example in Kansko-Achinskiy basin (Fig. 2).

In the coals of the eastern part of the basin, the concentration of U does not exceed the mean content of U in the world coals (2 ppm) (Ketris and Yudovich, 2009) whereas in the western part it is 4.9 ppm on the average. In some cases, anomalously high U contents in the Mesozoic coals of Siberia usually have epigenetic nature. Normally, they are confined to outcrops of coalbed under Quaternary deposits, wash-out sites of coalbeds, or sites of ground oxidation. The sites of high-radioactive coals are known in deposits of Irkutskiy and Kansko-Achinskiy basins and also in deposits of Zabaikalie and Mongolia (Smyslov et al., 1996). There are some data on the epigenetic concentrations of U in the Jurassic and Cretaceous coals of in northwestern and northern China, respectively (Dai et al., 2011; Zhuang et al., 2006).

The syngenetic U anomalies are rarely found in coalbeds. In addition, unlike in Siberia, the volcanic matter plays a significant role in the accumulation of the radioactive elements in the Mesozoic coals of the Russian Far East region and Mongolia. This is supported not only by the direct observations of the volcanic material in coal beds (Elginskoe, Urgalskoe, and other deposits) but also by the high Th–U ratio and also the high Th concentrations in coal ash of individual deposits, significantly exceeding its content in rock units of coal-bearing strata.

4.1.4. Cenozoic coals and peat

Cenozoic coal and peat formation is widely present in the territory of northern Asia. The wide basin of the Paleogene coal accumulation arose on the Western Siberian Plate. Coals of Paleogene age are widespread in Gorny Altai, are known in Kuzbass, and probably occur in Tungusskaya tectonic depression. Considerable resources are concentrated along the eastern margin of Euro-Asian Continent (Primorskiy Krai, Sakhalin, Kamchatka, and the north-east of Russia). The coals of this stage are notable for low quality, high-ash yield, and extremely heterogeneous contents of radioactive elements. The contents of U and Th in the studied deposits range from 0.7 ppm in coals of Sakhalin Island to 22 ppm in some coals of the Pavlovskoe deposit of Primorskiy Krai. Deposits located among basite-ultrabasite intrusive-volcanic formations are usually lean in U and Th. The Paleogene coals of Western Siberian basin are notable for the high levels of U and Th accumulation. The study of the U and Th contents in the coal ash points that the high concentration of the radioelements in the coal was caused by its high ash content. In conversion to the average ash yield for the region the coals of this period are significantly leaner with Th than the Mesozoic coals and they are comparable to them in respect of the U content. In addition the low Th–U ratio indicates the U geochemical feature of the Cenozoic Era of the coal formation in Siberia. This was also confirmed by the low Th–U ratio in the modern peat (Table 2).

The widespread formation of residual soil in the Paleogene at the peripheries of Western Siberian basin provided the input and concentration of U in the area of sedimentation. The formation of great U anomalies up to 300 ppm in the Paleogene coals is connected with it. The enrichment of the Jurassic and even Late-Paleozoic coals with U is connected with the Cenozoic stage. These processes especially obvious in the western part of Kansko-Achinskiy coal basin (the deposits Itatskoe, Nazarovskoe, Beresovskoe, and Kozulskoe). The superposed feature of the U anomalies is determined by their position in a coal bed and often by the disturbance of the radioactive equilibrium (Arbuzov et al., 2003, 2008; Gavshin and Melgunov, 2006; Gavshin and Miroshnichenko, 1999).

The radio-geochemical characteristics of the Russian Far East coals of the Cenozoic Era differ from Siberian ones. The influence of volcanic-hydrothermal processes on the formation of a contemporary radio-geochemical characteristic of the coals of the Russian Far East caused an extremely irregular distribution of U and Th within them.

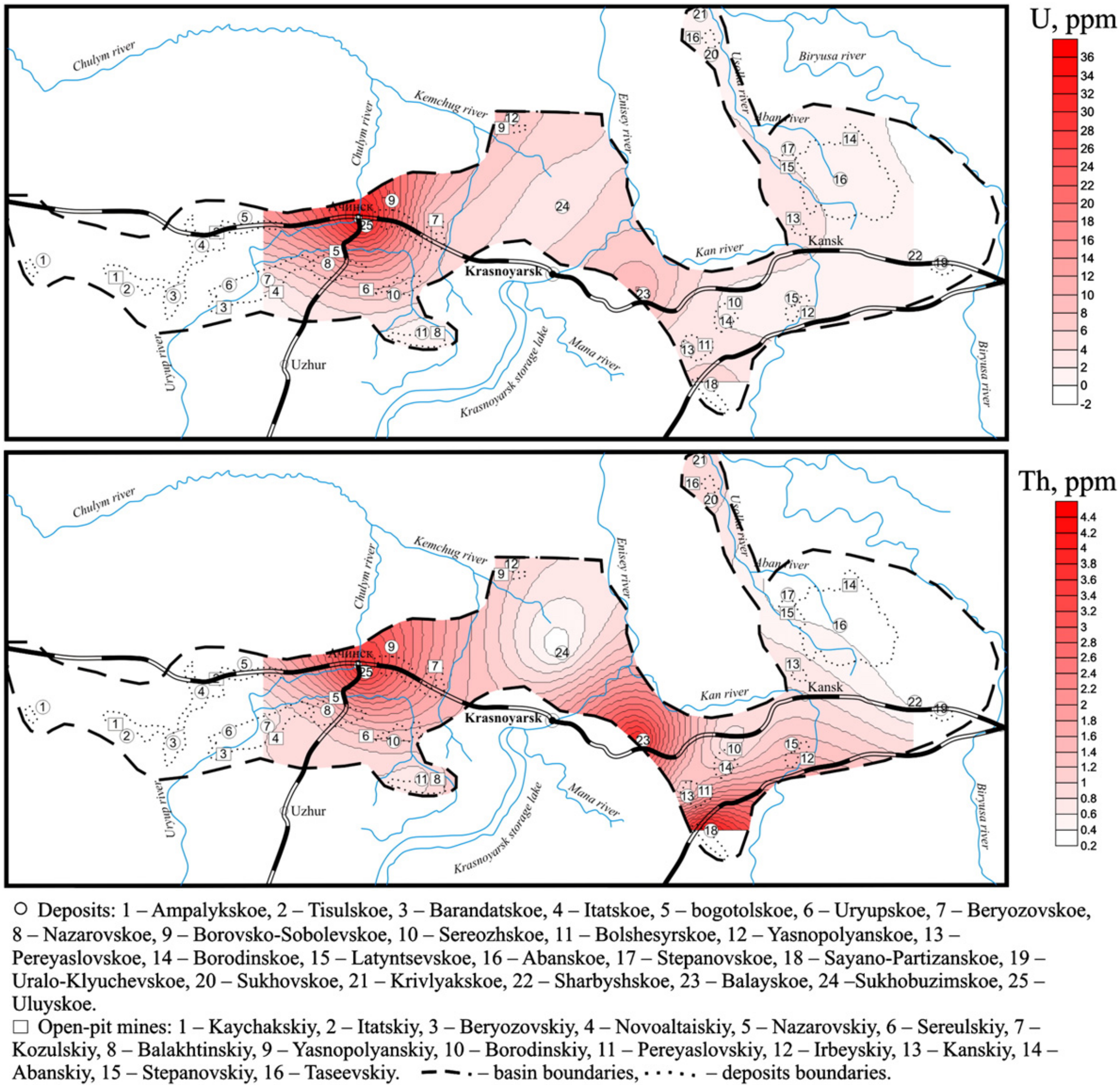

Fig. 2. Distribution of U and Th contents in coals of Kansko-Achinskiy basin.

Together with the low-radioactive coals of the Sakhalin and the Yano-Omoloyskiy basin, coals with the anomalous U and Th contents have been determined there. Unlike the Paleogene coals of Siberia, the Cenozoic coals of Russian Far East are, as a whole, characterized by the high Th–U ratio that indicates the mainly syngenetic clastogene mechanism of the U and Th input in a coal layer. Here the Ge-bearing Primorskiy Krai coals with the anomalous U content are sharply distinct. They differ by an extremely irregular U distribution with the quite normal Th content and in the opinion of Seredin et al. (2006) they have the hydrothermal nature.

4.2. Factors controlling the accumulation of radioactive elements in coals

As a whole, the coals of Northern Asia are defined by the low levels of U and Th accumulation which are close to the mean contents of these elements in the world coals. The studies revealed coal deposits and basins enriched with U and rarely with Th. Within the basins and deposits, individual coal beds or groups of beds and individual patches in beds are defined by high content of the radioactive elements. These differences are caused by different conditions which existed in a wide area of the coal formation in the Phanerozoe. First, the differences are determined by the geological-structural situation of the basins of the coal accumulation, tectonomagmatic, and climatic conditions. They determine the particularities of rock composition and geochemical zoning of the basin peripheries, composition and regime of underground and surface waters, rate of the coal accumulation, and coal properties. Different paleofacies, hydrological, hydro-geochemical conditions were formed based on combinations of these factors. These conditions determined the particularities of the radioelement accumulation in certain environment and also conditions of post-diagenetic distribution of the elements. All these factors were closely interconnected.

4.2.1. Tectonics

Geotectonic conditions, together with climatic factors, determine the rate of the coal accumulation, intensity of denudation processes in a source area, and features and composition of ground and surface waters.

As a whole the supplies of U and, especially, Th with terrigenous ash were predominate factor for coals, therefore, under other equal conditions, the higher-ash coals contain more U and Th. For instance, Kuraiskoe deposit of Pennsylvanian age (south of Siberia) is notable for higher content of radioactive elements in coals than anthracites of Gorlovskiy basin. This deposit, formed at very unsteady tectonic conditions, is defined by the high ash yield of coals, variable thickness of the coal beds, and their small dimensions in spite of the predominantly basite-ultrabasite composition of the source area. The coal accumulation in Gorlovsky basin occurred at steady tectonic conditions at a distance from the source area, as indicated by the low-ash yield of the coals and the predominance of fine-grained enclosing rocks.

The role of terrigenous ash in accumulation of U and, especially, Th is distinctly defined for all the regions. This factor is commonly confirmed by the direct correlation between Th content and the ash yield. Because of the particularities of the geochemistry of U, this factor for U is less evident but is also important.

Tectonic factor of the radioactive elements accumulation in coals plays a significant role together with the factor of source rock chemistry. Following Yudovich and Ketris (2002), the source rock is the whole group of surrounding rocks of a coal basin.

4.2.2. Source rock chemistry

The influence of rocks of a source area on the formation of coal basins and deposits geochemical background can be expressed in geochemical particularities of coals and in regular changes of the contents of trace elements from the margin of a sedimentation basin to the center. The important role of the factor of source rock chemistry in the accumulation of trace elements in coals is currently accepted as an axiom. The composition of rocks at the peripheries of coal-bearing depressions was apparently the main factor of the syngenetic accumulation of U and Th in coals.

The presence of rocks with enriched U contents at the peripheries of coal basins and deposits provided for the enrichment of coals with U (Arbuzov and Ershov, 2007). Thus, in southern part of Kuzbass, at the vicinity of folded margins it is noted that coals are more enriched with U, up to 3–5 ppm of U and 4–6 ppm of Th, in comparison with the northern part (Arbuzov et al., 2005). Coals of the northern part of Kuzbass (Kemerovo district) are characterized by a low level of U accumulation while the Th content is high. In both cases, the geochemical particularities of coalbeds correlate well with the geochemical characteristic of the surrounding structures. In southern and south-eastern parts of the basin, rare-metal granitoids, acid volcanics, and dark-shale and phosphate-bearing strata enriched with U are abundant. There are hydrothermal deposits and occurrences of U (Labyshskoe, Bazasskoe). The occurrence of the rocks of Th – rare-metal – rare-earth enrichment is peculiar to the north-eastern margins of Kuznetsk basin, which was the source area in the coal formation period in the north of Kuzbass. Hydrothermal deposits of Th are also known (Bogatyrskoe and others).

The coal accumulation in Tungusskiy basin occurred in the activated Siberian platform. The highest concentrations of U and Th were determined in the coals of Kayerkanskoe deposit located at the periphery of the platform. In that area, the activation process was accompanied by the formation of volcanics enriched with U and Th and also ore-bearing differentiated intrusives, all contributing to the geochemistry of the coal deposits.

The factor of the source rock chemistry is evident in the extended Kansko-Achinskiy basin. Numerous epigenetic anomalies in coals of the basin clearly coincide with the blocks of rocks enriched with acid and alkaline effusive rocks, granitoids, U deposits and occurrences. The anomalies associated with Kuznetsko-Alaltausskaya zone in the western part of the basin and Angaro-Kanskaya block in the east (Fig. 2).

The high U-bearing capacity of the Jurassic and Cretaceous coals in Mongolia is closely connected with a wide development of intrusive-volcanic complexes in the region (Mashenkin et al., 2009; Mironov, 2006).

4.2.3. Syndepositional volcanism

The presence of a volcanic material in the Pennsylvanian and Permian coal-bearing deposits of the Kuznetskiy, Tungusskiy, and Minusinskiy basins has been marked by many researchers (Admakin, 1992; Van, 1968, 1972; and others).

It is known that ash material enriched with occluded (mobile) modes of many trace elements (including P; K; and vital trace elements as Cu, Co, Zn, B, etc.) could be a natural "fertilizer" providing the intensive growth of coal-forming plants (Van, 2001; Yudovich and Ketris, 2002). In addition, more intensive ash-falls, which were temporarily capable of interrupting the peat formation, their later argillization turned the layer of volcanic ash into an effective aquiclude which caused the swamp formation and new growth of a peat deposit (Zhelinskiy and Mitronov, 1990). As a result of this the thickest coal beds were formed in advantageous geotectonic conditions.

Thereafter, during formation of tonsteins due to the swamp weathering of ash material free trace elements (including U and Th) could be occluded by the adjacent peat layers. Such a process was especially effective if the initial pyroclastics had acid and alkaline composition. This can be seen in coal beds of Minusinskiy basin, bed IV–V of Kuznetskiy basin, a group of beds of Urgalskoe, Rakovskoe and Pavlovskoe (Chikhezskoe) deposits of the Russian Far East, an extra-thick bed of the Nuurs-khotgor deposit in Mongolia.

The hydrolysis of the volcanic material leads to the loss of Th and rare-earth elements from tonsteins contributing to their accumulation in the organic matter of coal beds (Fig. 3).

These data agree with the investigations of the coal bed C of the Morrison formation in Utah, USA (Crowley et al., 1989), which showed that in the near-contact coal patches, adjacent to tonsteins, the concentration of uranium reaches up to 63 ppm and thorium — 60 ppm, while their contents in the ash of the central lithologies are 20–25 ppm and 25–30 ppm respectively. Since the contents of U (16 ppm) and Th (48 ppm) in the tonsteins are significantly lower than in the ash of the near-contact coal lithologies, the presence of a certain content of the elements connected with the organic matter due to their leaching from the ash horizons is supposed.

The ash material took a special significance for the geochemistry of radioactive elements while accumulating in low-ash coals. Our calculations for the layers Dvukharshinny and Velikan-I of Chernogorskaya suite of Minusinskiy basin, containing the interbeds of tonsteins, showed that the essential part of U and Th in coals could be supplied by the pyroclastics. About 75% of radioactive elements are connected with the pyroclastic material (Table 3).

The studies showed that the factor of syndepositional volcanism occurred in the whole territory of northern Asia. For western regions, the role of Pennsylvanian-Permian volcanism was significant in the accumulation of U and Th in coals. It is noted that to the east the pyroclastics becomes younger: from the Jurassic age for Pribaikalie, Zabaikalie, Yakutia, and central Mongolia; to the Paleogene-Neogene and Quarternary age for Primorie, Sakhalin, and the north-east of Russia.

4.2.4. Climatic Factor

At present, the role of a climatic factor in the accumulation of radioactive elements in coals is difficult to estimate due to the absence of necessary paleo-climatic information. Nevertheless, some ideas about the influence of the climatic factor can be expressed. Climate influences the rate of chemical reactions, leaching of radioactive elements from rocks, and their input to ground and surface waters. The level of the U and Th concentrations depends on the temperature and the amount of precipitation. Climate also influences the rate of the biomass growth, decomposition, and accumulation. These two conditions influence the syngenetic accumulation of radioactive elements. Together with the source rock chemistry, climate determines the formation of the radio-geochemical particularities of coals.

During Late Paleozoic accumulation, a stable, warm humid climate was believed to dominate in the whole studied territory, varying from a sub-tropical climate in the Pennsylvanian Period to a temperate one with distinct season changes in the Permian Period (Betekhtina et al., 1988). At the end of the coal accumulation, there was an aridization which more distinctly occurred in Minusinsk basin. The remains of

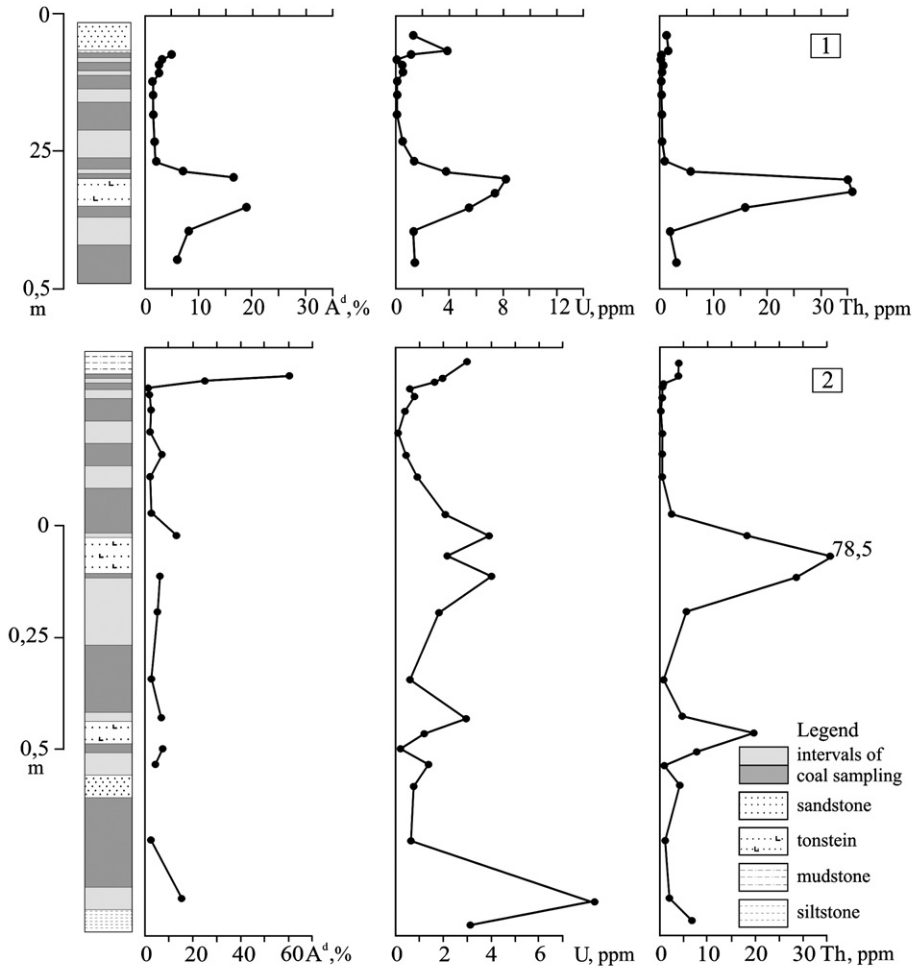

Fig. 3. Uranium and Th distribution in the sections of the coal beds Dvukharshinny (1) and Velikan-I (2) of Chernogorskoe deposit of Minusinsk basin.

xerophitic flora in sedimentary deposits; presence of facies of "drying lakes" in coal-bearing sequences; and horizons of freshwater carbonaceous deposits, testifying the high mineralization of surface and ground waters also indicate this (Bogomazov, 1961).

As follows from the data obtained, the coals of the top part of the sections of Minusinskiy (Izykhskaya suite) and Kuznetskiy (Tayluganskaya suite) basins are defined by the high contents of radioactive elements. It is possible a drier climate of the Late Permian contributed to the increase of U and Th contents in the surface and ground waters and, consequently, to their accumulation in the organic matter of the coal. These deductions agree with the study of two Lower Cretaceous coals in Canada (Van Der Flier and Fyfe, 1985) showing that the coals formed in conditions of a more intensive rock (kaolin-gibbsite) weathering were lean in U in comparison with the coals formed in conditions of a less active weathering.

Climate also influences the nature of recent hypergene changes of coals. The coalbeds in the aeration zone undergo epigenetic transformations. The conditions of a semiarid climate more significantly contribute to the U accumulation than humid conditions due to its higher concentrations in waters. Higher U contents formed in Minusinskiy basin under the contemporary arid climate in Khakassia in comparison with Tungusskiy and Kuznetskiy basins with more temperate climate. More pronounced hypergene anomalies are typical for coals of Mongolia and Zabaikalie.

4.2.5. Hydrological factor

Hydrodynamic conditions determined the particularities of water-mineral feeding of paleo-peatlands. The role of hydrological factor is evident when comparing the average contents of U and Th in modern ombrotrophic and minerotrophic peat deposits (Table 4).

Ombrotrophic peat deposits, generally feed by the atmospheric fallouts, are leaner in radioactive elements than minerotrophic ones. Minerotrophic peat deposits receive sufficiently more mineral

Table 3
Average contents (ppm) of U and Th and ash yield (A^d,%) of some coals containing pyroclastic material.*

Coal layer	Average for the section of a bed			Parts out of the zone of pyroclastics influence		
	A^d,%	U	Th	A^d,%	U	Th
Dvukharshinny	5.0	1.5	3.1	2.5	0.4	0.8
Velikan-I	4.4	1.1	3.1	2.0	0.2	0.5

* Average data for some detailed sections.

Table 4
Average contents of U and Th in peat and peat ash of Western-Siberian Plate (ppm).

Elements	Low-moor peat		High-moor peat		Average for the region	
	Dry matter	Peat ash*	Dry matter	Peat ash*	Dry matter	Peat ash*
Th	0.87	6.5	0.45	14.0	0.62 ± 0.1	8.5
U	0.46	3.4	0.31	9.7	0.37 ± 0.08	5.1
A^d(%)	13.4	100	3.2	100	7.3 ± 0.9	100
Number of samples	702	702	1225	1225	1927	1927

* Calculated over the ash; A^d — ash yield, %.

substances, including radioactive elements, in surface and ground waters. In certain peat deposits the U and Th contents are comparable with their contents in coal deposits (Table 5).

According to the study of three Canadian deposits (Shotyk et al., 1990), the U concentration in the deep horizons of the peat deposits is 7.3 ppm, 6.5 ppm (26 ppm in the ash, ash yield is 6.4%), and 1.5 ppm (42 ppm in the ash, ash yield is 3.6%). There are numerous examples of anomalous radioactive peats. In Filippi (Greece), the thickest peat deposit known at present, the U content in the ash ranges from 11 to 208 ppm. For Th, the range is 4.6 to 14.8 ppm (Kalaitzidis et al., 2002). Among 167 studied peat deposits in the forest-steppe zone of the southern Siberia, there are 35 deposits with the uranium content of higher than 0.01% (Roslyakov et al., 2004).

A buried peat deposit, being a water-bearing stratum, sorbs additional mobile U. Elevated content of U in the peat of the deposits Kizhirovo and Obskoe and low Th–U ratio indicate this (Table 5).

Another constituent of the hydrological factor is the composition of surface and ground waters. Despite the evident role of this factor in coal deposits, it is difficult to single it out because of the lack of reliable data on the paleo-hydrochemistry of the region. The published data of Lisitsyn et al. (1967) indicate that, under the conditions of climate aridization, waters get enriched with U and the U-bearing of peat deposits increases. According to some authors (Dai et al., 2008; Gayer et al., 1999; Shao et al., 2003; Van Der Flier and Fyfe, 1985) the U concentration in coals increases and the Th–U ratio decreases under the influence of sea waters. This indicates the U sorption from the sea water.

4.2.6. Factor of coal metamorphism

At present there are no convincing facts which would prove a loss or input of radioactive elements under the coal metamorphism (Affolter and Hatch, 1984; Goodarzi and Cameron, 1987). Nevertheless, according to the estimations of the mean U content in brown and bituminous coals (Ketris and Yudovich, 2009) and comparion of the contents of U and Th in coals and anthracites of certain basins (Arbuzov and Ershov, 2007), it may be assumed that the coal metamorphism processes lead to the loss of radioactive elements in the organic matter of coal.

The comparison of the average contents of U and Th in coals with a different rank can be correct only if the other conditions of their formation are equal. It is evident that the search of such objects is very difficult due to the lack of knowledge about the conditions of the coal formation even for the well studied coal basins. Thus, the Jurassic brown coals and Pennsylvanian-Permian coals of Siberia are unsuitable for the comparison due to the significant influence of the syndepositional volcanism on the formation of the geochemical background of the latter. Such comparison in Siberia is only possible with certain limitations for single-age coals of Kuznetskiy basin, because the coals there have almost the whole range of grade composition. The direct comparison of the contents of U and Th in coals of different grades does not allow for correct deductions because of interferences connected with the influence of heterogeneity of the source area composition, facial differences, varied influence of syndepositional volcanism, and other factors. In addition, the statistical analysis of the materials with the calculation of regression equation indicates the decrease of the U content during the coal metamorphism process.

According to these data, no more than 50% of U is lost, even comparing last members of the studied metamorphic range. Thorium does not have distinct tendencies to loss upon coal metamorphism. This is confirmed, for example, by the increase of thorium–uranium ratio from brown to bituminous coals.

4.2.7. Hypergene oxidation of coals

In some cases, hypergene oxidation causes an accumulation of U in coal. The high sorption capacity of the organic matter of coal in relation to U enhances its concentration from ground waters even with the low content of the metal. Arid climates with high contents of U and oxygen in water are especially favorable for this process. The size of these zones of U accumulation is generally not too large in comparison with the scale of coal basins and cannot significantly influence the estimation of the geochemical background of coal deposits. However, within certain deposits this influence can be significant. For instance, the high content of U in the Aduunchuluun deposit (Mongolia) is determined by its epigenetic accumulation up to commercial concentrations. The oxidized coals of certain deposits of Russian Far East and Siberia are essentially enriched with U. In the Kansko-Achinskiy basin, the coal resources of up to 5% are oxidized, which contain, in some cases, up to 0.2% U (Gavrilin and Ozerskiy, 1996).

The top part of a coalbed is usually enriched with U. The maximum concentration is related to the front of the zone of oxidation (Fig. 4). Thereby, the whole zone of oxidation is defined by the anomalous content of U. This fact indicates a consistent role of the sorption and oxidation-reduction geochemical barriers in the accumulation of U in the oxidized coals.

5. Conclusions

In general, the coals of northern Asia have low levels of U and Th accumulation. The average content of Th is lower than the mean content of Th in the coals of the world and that for U is comparable with the mean content of U in the coals of the world.

The distribution of U and Th in the coal basins is irregular. This is influenced by a combination of factors. These factors are: heterogeneity of rocks composition of folded boundaries of the basins, difference of the facies conditions of the coal accumulation, influence of volcanism, climatic conditions, and degree of the coal metamorphism.

The high concentrations of U and Th in coal-bearing deposits are related to the blocks of rocks enriched with U and Th at the peripheries of the basins or connected with the occurrence of a volcanism in the period of the coal accumulation. The combination of conditions was defined by the occurrence of radio-geochemical complexes at the peripheries of coal-bearing structures, which were subjected to the weathering at humid climate and to volcanic activity in the period of the coal formation. This caused the formation of geochemical anomalies of U and Th in some coalbeds and deposits of the region.

Table 5
Average contents of U and Th in some peat deposits of Western-Siberian Plate (ppm).

Elements	Ombrotrophic		Minerotrophic and transitional			
	Vasyuganskoe	Vodorazdelnoe	Modern		Buried	
			Berezovaya Griva	Pukhovskoe	Kizhirovo	Obskoe
Th	0.22	0.34	1.3	2.9	1.7	1.9
U	0.13	0.15	1.2	0.87	2.7	3.9
Th/U	1.7	2.3	1.1	3.3	0.6	0.5
A^d	4.0	3.9	7.5	17.8	27.7	40.9

A^d — ash yield, %.

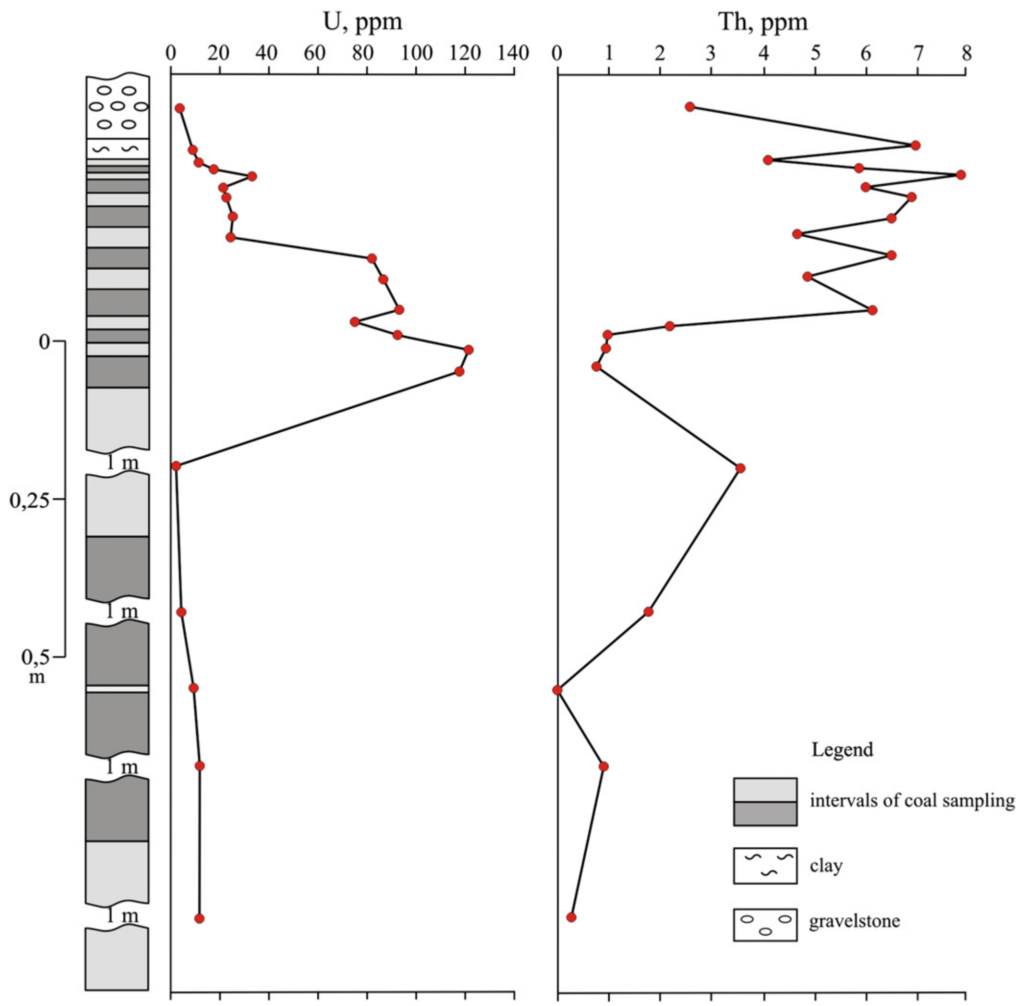

Fig. 4. Distribution of U and Th contents in a section of the coalbed "Itatskiy" of Itatskoe deposit of Kansko-Achinskiy basin.

Various periods of coal accumulation in different regions of northern Asia differ by the contents of the radioactive elements in the coals. From west to east the role of volcanic material in the concentration of U and Th changes. Volcanism in Siberia, Zabaikalie and Yakutia, and eastern Eurasia influenced Paleozoic, Mezosoic, and Cenozoic coals, respectively. The occurrence of the volcanic activity is determined by the high Th–U ratio for ordinary coals and their accumulation of both U and Th.

The anomalous contents of U generally have the epigenetic nature and are connected with of oxygenenous U-bearing waters in coalbeds. They cause the coal oxidation and accumulation of high U concentrations. The regions of high radio-geochemical background and arid climate are more favorable for the formation of oxygenous waters, contributing to the accumulation of anomalous U concentrations in organic matter.

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