# Redistribution of U, Ra, Th and K-40 over Landscapes of the Priobskoye Plateau and East Kulunda

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#### **Abstract**

The distribution of natural radioactive elements (uranium, radium, thorium and potassium) in the soils of the Priobskoye plateau and the eastern part of the Kulunda plain depicting different physical geographical zones is presented. The most evident radiogeochemical zoning is revealed for uranium and potassium in the autonomous landscapes. The concentrations of radium and thorium remain at the same level without any noticeable redistribution. A method is proposed to estimate uranium carry-over during Holocene (12–10 thousand years) from the soil eluvial profiles of the zone of hypergenesis. With the help of this method, the amount of uranium carry-over from the first meter of the soil cover is estimated.

#### INTRODUCTION

Compilation of reliable balances of matter transfer between continents and the ocean is still at the initial stage. Estimations made by researchers are based mainly on calculations of the content of elements in the mineral and dissolved forms, in rivers and in the volume of river run-off. It is extremely difficult to carry out such estimations correctly; what is more, run-off gives only an estimation for the current moment, because one of the main parameters governing carry-over is climate, which causes permanent changes in the volume of the matter to be transferred. Meanwhile, for radioactive elements, there is another way to estimate the intensity of carry-over. Heavy radioactive elements form long decay chains; separate terms of these chains are in secular equilibrium in closed systems. In open systems the elements of these systems are carried with water flows at different rates since they possess different chemical properties. Researchers of geochemistry of uranium established that the oceanic water is enriched with uranium (with respect to radium) while the soil eluvial

profiles are depleted of uranium (with respect to radium) [1–3], which is the evidence of more rapid uranium transfer in comparison with radium. Taking into account low mobility of radium in comparison with the parent uranium, one may perform quantitative estimations of its carry-over within the geological time interval, which seems impossible to be done for other elements.

We carried out investigation of uranium carry-over in the zone of active water exchange from the soil eluvial profiles of the southern part of Zapadno Sibirskaya Ravnina plain which were formed during Holocene (10 000 years ago – present time). In addition to uranium, we considered other natural radioactive elements (NRAE)\*.

<sup>\*</sup> Natural radioactive elements mean radioisotopes making the major contribution into the land constituent of the natural radiation background. These include uranium isotopes  $U^{238}$  and  $U^{235}$  which are mobile in the maximal oxidation degree, their initial content in natural uranium is 99.275 and 0.720 %, respectively; low-mobile  $Ra^{226}$  and  $Th^{232}$  accounting for almost 100 % of natural radium and thorium in soil profiles; and  $K^{40}$  radioisotope making 0.0119 % of natural potassium macroelement [4].

#### EXPERIMENTAL PROCEDURE

In order to reveal the features of NRAE behaviour (mobilization, transport and accumulation) in natural flow reactors of the Priobskoye plateau and the eastern part of the Kulunda plain, a series of grounds belonging to different landscape-climatic zones was chosen. The following criteria were used: 1) membership in the head reaches of the Holocene hydronet; 2) typical nature of the chosen ground for the corresponding landscape-climatic zone; 3) prevalence of pre-Holocene cover loess in the soil-forming substrates; 4) the presence of all the types of catenae-bound elementary landscapes at the ground; 5) minimal disturbance of the landscape by anthropogenic action.

Six regions were chosen in three landscape zones (from the southern taiga to the dry steppe) with comparative development of hydronet at different grounds, so that drainability of the landscapes was approximately the same. This requirement is met by the head reaches of the modern hydronet. The dimensions of different grounds were 35 to 80 km<sup>2</sup>.

According to the approach proposed by B. B. Polynov and developed by A. I. Perelman [5, 6], the following elementary landscapes were distinguished at the grounds: 1) autonomous ones, represented by ridge heights and flat watershed spaces; 2) transition ones, corresponding to the slopes of different steepness; 3) subordinate ones: thalwegs of valleys with temporary and permanent water flows.

The turf-humus horizon and the layer of subsoil substrate were examined in the elementary landscapes. Complete soil profiles were also sampled in autonomous and subordinate landscapes; bottom sediments, samples of the surface and underground water were taken. Soil was sampled at the depth of horizon and placed in cloth bags. The amount of sampled soil was 600–700 g. Bottom sediments were sampled in plastic wide-neck bottles (1000–1200 g per bottle), water samples were placed in glass bottles (the volume of a sample was 500 ml).

The NRAE content was determined in the samples. The samples of soil-forming rock represented by loess-like loam were analyzed for mechanical and macroelemental composition.

Radium, thorium and potassium-40 were determined at the analytical centre of the UIGGM, SB RAS with gamma-spectrometric devices with the well scintillation NaI (Tl) crystals. Metrological characteristics of the devices were obtained on the basis of the analysis of standard rock samples [7]. Uranium content was determined by laser luminescence procedure examining luminescence of water extracts with the Angara laser analyzer in Berezovgeologiya PGO.

Mechanical composition was examined using Kachinsky's procedure at the SSI, SB RAS, macroelemental composition was studied by means of X-ray fluorescence analysis at the Analytical Centre of the UIGGM.

In addition to absolute content of elements, geochemical markers were analyzed: the difference between uranium content calculated from radium content (U(Ra)) and uranium content determined by laser luminescence procedure (U<sub>lum</sub>), the thorium to uranium ratio. The former marker (the difference U(Ra) – U<sub>lum</sub>) allows estimating uranium carry-over degree because radium, being the daughter element of uranium, is present at the ratio of Ra/U =  $3.36\ 10^{-7}$  in the absence of carry-over. This marker is equal to zero in closed systems.

The hypergenesis zone is an open system in which permanent redistribution of the matter occurs under the action of atmospheric water and biota. Since radium and uranium are geochemically contrasting elements, they strongly differ by their behaviour in landscapes. The former element is well retained in geological media, while the latter one is readily leached (in the absence of geochemical barriers). Because of this, determination of uranium concentration in rocks, ground and soil by means of gamma spectrometry actually only depicts trace concentrations of radium, while uranium can already be carried out of these layers (or brought into them) [8, 9]. The ratio  $U_{lum}/U(Ra)$ was used as a version of this marker, indicating the difference between actual uranium concentration and the concentration in equilibrium with radium. The stronger this ratio differs from unity, the more intensive uranium carry-over is. The second geochemical marker is Th/U ratio. Chemical properties of uranium and thorium strongly differ from each other under the weathering conditions. Thorium, which does not change its valence, is a typical hydrolyzate element; A. I. Perelman placed it among the elements with weak migration. Transference of a part of it into the pore solutions of the soil eluvial profiles results in almost all the cases in precipitation at the place in the form of hydroxide  $Th(OH)_4$ . In river water, thorium migrates is solid suspensions; only 1-2% is carried in truly dissolved form [4].

We accept, though with a proviso, that by the beginning of Holocene the members of the uranium decay row were in radioactive equilibrium with each other. As a result of homogenization during the transfer of the ash terrigenous material, the difference in the ratios between isotopes arising in the weathering products of the desert landscapes got leveled. Loess rocks were forming during the stage of maximal glaciation, when the activity of water exchange along the vertical direction was sharply decreased. The model did not take into account the possibility of distortion of the equilibrium between other elements of this chain. About 7–10 radium half-life periods have passed since the formation of the loess mantle. So, 0.78-0.097 % of the amount of radium which was present at the moment of mantle formation remains till present; radium which we detect has been formed already in Holocene.

Statistical estimations of radionuclide distribution are represented by arithmetic mean values with confidence intervals corresponding to the significance level 0.05, as well as by the value ranges. Calculation of confidence intervals is used for the cases of normal distribution; however, as demonstrated in [10], deviation from the normal distribution in geochemical practice has a small effect on the confidence interval value.

## **OBJECT OF INVESTIGATION**

The Priobskoye plateau and the eastern part of the Kulunda plain were chosen as the objects of investigation. The basis for the choice of this region was substantial sub-meridional stretch of the territory, which is relatively uniform in the geologic and geomorphological structure, and on which different landscape-climatic zones had superimposed during Holocene.

The transecting line crosses several landscape-climatic zones from the southern taiga to the steppe zone; each of them is characterized by its own climatic conditions and geochemical barriers (Fig. 1). This allows one to estimate the difference in mobilization, transportation and deposition of the elements of interest depending on climate (Table 1). Radiation balance and average annual temperatures increase gradually from north to south, while the annual amount of precipitation and HTC, vice versa, decrease along the same direction. Salt content of natural waters decrease inversely proportionally to the amount of precipitation. The salt content of water is ultralow in the regions of the taiga zone, moderate and low in forest steppe, while water of the steppe zone is weakly salty (Table 2).

At the major part of the territory under investigation, the soil-forming substrate is pre-Holocene loess, as well as loess-like loam and clay sand (Fig. 2) [11, 12]. Mechanical composition of the soil-forming rocks reveals essential similarity with the substrates of autonomous landscapes of separate pieces of land. Coarse dust (particle size 0.05-0.01 mm) and silt (<0.001 mm) are prevailing in them. The Aleus region is distinguished by an increase in the fraction of dusty sand (0.25-0.05 mm) to 26.2 %. The reason seems to be neighbourhood with the ancient depression of drain in the thalweg of which sands are predominant.

Stable high amount of the fractions of coarse dust and silt (varying within the range 36-54 and 20-33 %, respectively) confirms loess-like appearance of these rocks and points to the similarity of their genesis. At the same time, these rocks differ in losses from the action of HCl, that is, in the content of calcium and magnesium carbonates. Substrates of the subordinate landscapes at different regions are similar to each other, too. In addition to coarse sand and silt, dusty sand appears in them in noticeable amount (8-30 %). At separate regions when passing from autonomous landscapes to subordinate ones, the fraction of dusty sand increases (by 14 % as a mean), while the fraction of coarse dust decreases (by 13 % as a mean) (Fig. 3).

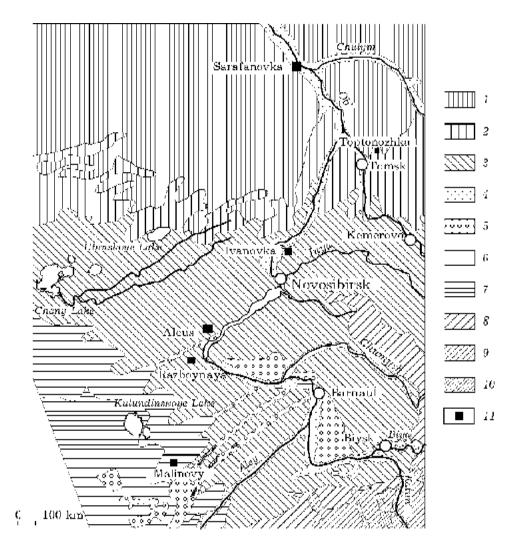


Fig. 1. Landscape-climatic zones of the south of West Siberia: 1 – southern taiga, 2 – sub-taiga, 3 – forest-steppe, 4 – flood-plain landscapes, 5 – pinery terraces, 6 – steppe, 7 – dry steppe, 8–10 – mountain landscapes (forest-steppe, forest, inter-zonal), 11 – type regions.

TABLE 1 Climatic characteristics of the grounds of transect

Region	Radiation balance, kcal/cm <sup>2</sup>	Annual amount of precipitation, mm	Number of days with ten higher than 10 $^{\circ}\mathrm{C}$	nperature HTC*
Sarafanovka	85.3	510	110	1.20
Toptonozhka	87.4	495	115	1.15
Ivanovka	89.4	409	123	1.00
Aleus	93.3	400	130	0.95
Razboynaya	95.6	390	135	0.85
Malinovy	102.6	315	140	0.80

<sup>\*</sup>Hydrothermal coefficient is equal to the ratio of the sum of precipitation within a stable period with temperature above  $10~^{\circ}$ C to the sum of positive temperatures within the same period, decreased by a factor of 10.

Cation and anion composition, pH and total salt content of surface water on type regions

CABLE 2

Sarafanovka         7.0         7.0         12.6         <1	Region	Hd	Anions				Cations				
vka         7.0         7.0         12.6           chka         7.6         62         1.5           r         7.0         8.9         5.0           rya         8.0         21.9         44.0           rya         8.0         53.9         120.0           r         7.8         123.8         357.5           7.3         1700.0         900.0           Zarovoye         6.8         1950.0         1600.0			CI_	$\mathrm{SO}_4^{2-}$	$NO_3^-$	$\mathrm{HCO}_{3}^{-}$	Fe <sup>3+</sup>	Na+	K+	${ m Mg}^{2+}$	Ca <sup>2+</sup>
hka 7.6 6.2 1.5 7.0 8.9 5.0 8.0 21.9 44.0 ya 8.0 53.9 120.0 7.8 123.8 357.5 7.8 1700.0 900.0 Aarovoye 6.8 1950.0 1600.0	rafanovka	7.0	7.0	12.6	\ \	72.2	99.0	10.7	3.4	80	51.0
tya     7.0     8.9     5.0       sya     8.0     21.9     44.0       rya     8.0     53.9     120.0       rya     7.8     123.8     357.5       rya     7.3     1700.0     900.0       rya     6.8     1950.0     1600.0	optonozhka	9.7	6.2	1.5	7	78.0	3.70	6.1	9.0	9.1	71.0
ya     8.0     21.9     44.0       .ya     8.0     53.9     120.0       .     7.8     123.8     357.5       7.3     1700.0     900.0       Aarovoye     6.8     1950.0     1600.0	anovka	7.0	8.9	5.0	< 1	94.6	2.63	5.9	1.1	9.8	90.5
.ya 8.0 53.9 120.0 7.8 123.8 357.5 7.3 1700.0 900.0 Aarovoye 6.8 1950.0 1600.0	leus	8.0	21.9	44.0	< 1	131.8	0.22	111.0	45	31.0	48.5
7.8 123.8 357.5 7.3 1700.0 900.0 Parovoye 6.8 1950.0 1600.0	ızboinaya	8.0	53.9	120.0	~	9.7.6	0.20	139.0	48	41.0	43.0
7.3 1700.0 900.0 Farovoye 6.8 1950.0 1600.0	alinovy	7.8	123.8	357.5	~	230.2	0.20	567.5	7.1	92.5	49.0
6.8 1950.0 1600.0	uchuk	7.3	1700.0	0.006	~	392.8	0.28	0.069	550.0	2000.0	252.5
	aloye Yarovoye	8.9	1950.0	1600.0	\ \	500.0	0.23	773.0	610.0	2900.0	250.0
Petukhovo 9.6 675.0 33.5 <1	stukhovo	9.6	675.0	33.5	~	1300.0	0.16	1091.5	1106.8	13.0	40.3

The macroelemental composition of the loess cover of the stratum is consistent along the lateral and vertical lines. Macroelemental compositions of the substrates from the grounds of the transect are shown in Table 3.

Experts have not arrived to consensus of opinions as regards the genesis of loess-like sediments. Some researchers believe that these sediments were formed mainly as a result of aeolian factor; others think that these are polygenetic sediments formed by aeolian, fluvial, soil, cryogenic and other processes [12, 14, 15-17]. In general, we accept the aeolian concept of the formation of sediment, which is confirmed by our data on the mechanical composition of the substrate; however, since all the substrates of the subordinate landscapes are enriched with respect to the autonomous ones by the fraction of medium-sized and fine sand (0.25-0.05), the formation of these substrates in diagenesis was participated by fluvial, slope and other processes. The age of the upper part of the loess cover varies from 21 to 10 thousand years, according to the data of different researchers. The major part of the matter in the upper part of the loess sediments was accumulated within the period from 19-18 till 14-15 thousand years (Yeltsovskiy loess horizon) and 12-10.2 thousand years (Baganskiy loess horizon) [12, 13].

The matter of this stratum has homogenized during aeolian transport and post-sediment resedimentation. The homogeneity of this stratum was noted by many researchers [11, 12, 14, 15]. This is confirmed also by the similarity of NRAE concentrations in loess-like loamy soils at different grounds. Sediments at these depths were less affected by the processes of Holocene hypergenesis and can be considered as the samples of the initial composition of the soilforming substrate within the limits of each ground. The NRAE content of loess is weakly varied in the substrates of the grounds of the transect (Table 4). At present, due to uranium carry-over, the equilibrium between Ra and U is shifted towards radium. We accept (with a proviso) that this carry-over occurred within the Holocene stage, since the sediments were mixed many times during transportation and accumulation of the sediments with deficiency and excess of Ra. The concentrations of Ra

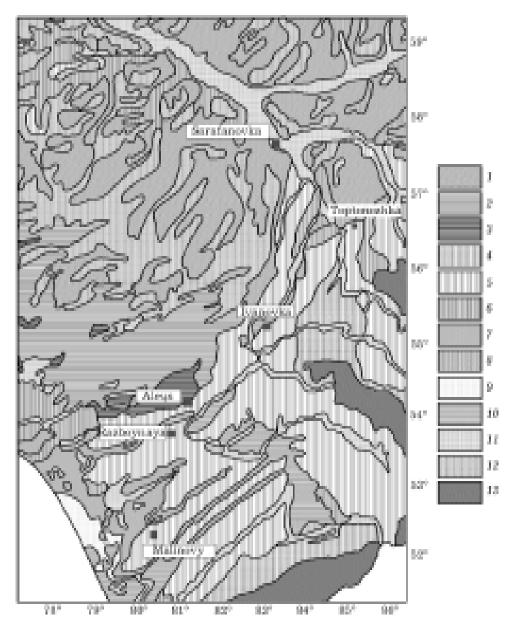


Fig. 2. Map of mechanical composition of the soils of West Siberia (layer 0-150 cm [2]: 1 – heavy loam and muddy-dusty clay; 2 – heavy loam and clays: dusty-muddy and muddy-sandy; 3 – heavy and medium loam, more rarely clays: sandy-muddy and muddy-sandy; 4 – heavy loam: sandy and muddy-sandy; 5 – medium, more rarely heavy muddy-dusty loam; 6 – medium and light loess and loess-like loam: muddy-dusty and dusty-sandy; 7 – light loam, more rarely clay sand and muddy-sandy; 8 – sands and clay sand on loam and clay; 9 – clay sand; 10 – sands; 11 – layered clayish-loamy-clay sand sediments; 12 – organogenic sediments on loam and clay, more rarely on clay sand and sand; 13 – rock outcrop and loamy-rubbly sediments in mountain depressions and river valleys.

and U were leveled and approached the equilibrium values.

The formation of modern soil cover at watersheds is connected with the Holocene time (10-0 thousand years). The accumulation of humus occurred during wetting after degradation of the last cover glaciation at the north of the Zapadno Sibirskaya Ravnina. After

the formation of loess-like cover under the changed hydrothermal conditions of Holocene an active redistribution of chemical elements started under the action of soil forming processes. A united soil horizon was formed on the flat countries between rivers during the Holocene time; several soil profiles were formed in lower sites of the relief under the action

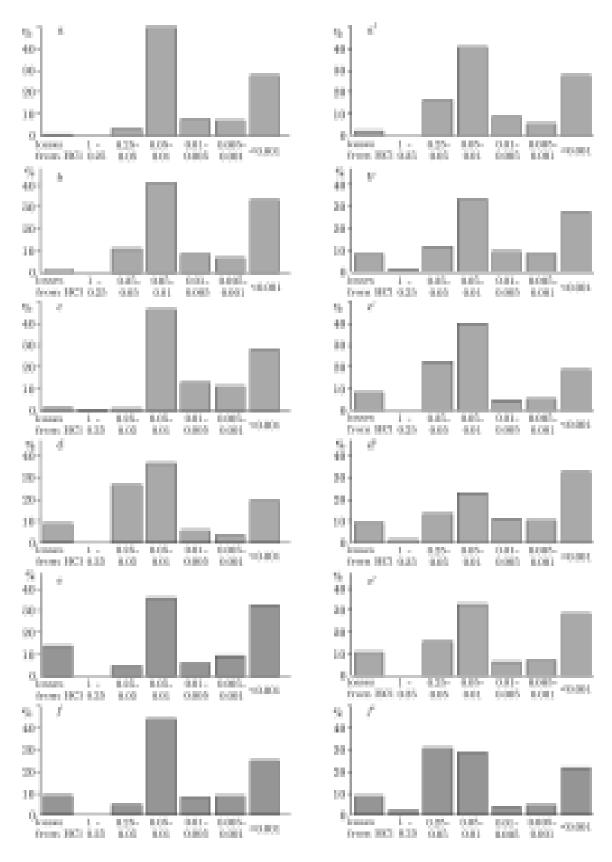


Fig. 3. Distribution of granulometric fractions in autonomous (a-f), subordinate (a'-c',e',f') and transition (d') substrates of the grounds of transect: a, a' – Malinovka; b, b' – Toptonozhka; c, c' – Ivanovka; d, d' – Aleus; e, e' – Razboynaya; f, f' – Malinovy.

Macroelemental composition of soil-forming rocks at the grounds of transect,

TABLE 3

Region	$\mathrm{SiO}_2$	${ m TiO}_2$	$\mathrm{Al}_2\mathrm{O}_3$	${ m Fe}_2{ m O}_3$	MnO	MgO	CaO	$\mathrm{Na_2O}$	$ m K_2O$	${ m P_2O_5}$	Ba <sup>2+</sup> C <sub>3</sub>	Calcination Total loss	Total
Sarafa-													
novka	64.28	0.81	12.91	4.95	960.0	1.96	4.48	99.0	1.87	0.158	0.040	7.23	99.45
Topto-													
nozhka	66.46	0.82	13.67	5.53	0.097	1.60	1.47	99.0	2.04	0.162	0.043	6.40	98.95
Ivanovka	63.72	0.73	11.51	4.64	0.090	2.34	2.67	1.46	1.91	0.147	0.033	7.36	99.61
Aleus	72.91	9.0	11.62	4.26	0.078	1.15	1.30	99.0	2.01	0.126	0.042	4.70	99.51
Razboy-													
naya	61.05	89.0	12.79	4.60	0.091	2.56	5.77	1.46	2.02	0.140	0.036	8.44	89.63
Kuchuk	02.99	0.72	10.92	4.03	0.083	1.96	5.34	1.14	1.85	0.146	0.038	6.65	99.57
Mean													
7 26 ±													
conf. int.	$65.9 \pm 4.2$	$65.9{\pm}4.2 \qquad 0.74{\pm}0.07  12.2{\pm}1.1  4.7{\pm}0.56$	$12.2\pm1.1$	$4.7 \pm 0.56$	$0.09\pm0.007$	$1.9\pm0.53$	$4.0\pm 2.18$	$1.0 \pm 0.42$	$1.95\pm0.087$	$0.15\pm0.013$	$0.035\pm0.004$	$6.8 \pm 1.3$	

of sliding, erosive drift and other exogenous processes.

#### **DISTRIBUTION OF RADIOELEMENTS**

The distributions of NRAE in autonomous landscapes turned out to be most clearly connected with hydrothermal zoning. It is more difficult to distinguish zonal trends in transeluvial landscapes and especially in subordinate ones. This is connected with the action of the surface water which carries the matter from all the horizons of different types of landscape thus masking non-uniform action of the climatic factors. Changes in mean concentrations in the humus horizon and in the subsoil substrate of the autonomous landscapes are shown in Fig. 4 and in Tables 5 and 6.

#### Uranium

Changes in uranium concentration in the soils of the typical grounds of the transect are shown in Fig. 4, a, a'. A gradual decrease in the mean uranium content from north to south is observed in the soil-forming substrate, in spite of overlapping confidence intervals for different grounds. Minimal values were observed at the Aleus region. The reason is dilution of the clayish substrate of this ground by quartz-feldspar sand brought from the neighbouring shallow gully.

A trend of decreasing the mean uranium content in soil and soil-forming substrate from north to south arouse after the formation of the loess mantle under the action of hypergenous processes. The same trend is exhibited even more clearly in the humus horizon; the confidence intervals at the most contrasting regions, Sarafanovka and Malinovy, do not overlap any more. We think that an increase in uranium carry-over at the southern regions is connected with an increase in the concentration of hydrocarbonate ion in atmospheric and soil water. In the presence of hydrocarbonate ion, uranium easily forms polyuranate complexes which are able to migrate over long distances [2, 9] This increase in well observed in the chemical composition of the surface water flows (see Table 2).

 $\begin{tabular}{ll} TABLE~4\\ Concentration~of~radionuclides~in~the~subsoil~substrate~of~autonomous~landscapes \end{tabular}$ 

Region	Sampling depth, cm	$U_{lum}$ , $g/t$	U(Ra), g/t	Th, g/t	K, %
Sarafanovka	200-210	1.7	2.2	7.1	1.48
Toptonozhka	190-200	1.4	2.1	7.6	1.41
Ivanovka	180-190	1.6	2.4	8.0	1.54
Aleus	150-160	1.8	2.2	6.1	1.25
Razboynaya	150-160	1.8	2.3	6.5	1.43
Malinovy	175-185	1.5	2.2	6.5	1.34

TABLE 5
Concentrations of radionuclides in the humus horizon of autonomous landscapes

Region	Number of sites	$U_{gum}$ , $g/t$	U(Ra), g/t	Th, g/t	K, %
		Humus ho	rizon		
Sarafanovka	14	$1.7 \pm 0.16$	$2.0 \pm 0.13$	$6.6 \pm 0.50$	$1.3~\pm~0.10$
Toptonozhka	10	$1.7~\pm~0.20$	$2.1~\pm~0.24$	$7.0~\pm~0.56$	$1.32 \pm 0.09$
Ivanovka	11	$1.7~\pm~0.18$	$2.0~\pm~0.17$	$6.8~\pm~0.59$	$1.34 \pm 0.09$
Aleus	13	$1.5~\pm~0.12$	$1.7 \pm 0.17$	$6.8 \pm 0.61$	$1.59 \pm 0.09$
Razboynaya	9	$1.5~\pm~0.11$	$2.0~\pm~0.12$	$8.2 \pm 0.39$	$1.67 \pm 0.08$
Malinovy	12	$1.37 \pm 0.09$	$2.2~\pm~0.13$	$7.9~\pm~0.52$	$1.79 \pm 0.08$
		$Subsoil \ sub$	strate		
Sarafanovka	14	$1.8 \pm 0.35$	$2.1~\pm~0.12$	$7.5~\pm~0.5$	$1.48 \pm 0.07$
Toptonozhka	10	$1.64 \pm 0.12$	$2.1~\pm~0.12$	$8.3 \pm 0.50$	$1.5 \pm 0.09$
Ivanovka	11	$1.7~\pm~0.24$	$2.1~\pm~0.19$	$7.8 \pm 0.85$	$1.54 \pm 0.07$
Aleus	13	$1.4 \pm 0.19$	$2.0~\pm~0.15$	$7.5 \pm 0.61$	$1.64 \pm 0.06$
Razboynaya	9	$1.6 \pm 0.13$	$2.0~\pm~0.15$	$8.0 \pm 0.70$	$1.6 \pm 0.15$
Malinovy	12	$1.5~\pm~0.09$	$2.2~\pm~0.1$	$8.0~\pm~0.56$	$1.7~\pm~0.11$

## Radium

Uranium content calculated on the basis of the measured amount of radium is characterized by a more consistent distribution. Changes in the concentration of U(Ra) in the soils of typical grounds of the transect are shown in Fig. 4 a, a'. The concentrations are more consistent in the soil-forming substrate; they are at the same level: 2.04-2.28 g/t.

In soil, the concentration of U(Ra) is at the same level. Only the Aleus region deviates from the general trend. The reason is the addition of sand from the shallow gully.

Deficiency of uranium with respect to radium is observed in all the regions for all the types of elementary landscapes; this is an evidence of uranium carry-over with the surface and underground water.

## **Thorium**

Changes in the concentration of thorium in the soils of the typical grounds of the transect are shown in Fig. 4, c, c'. The distribution of thorium in the soil-forming substrate is uniform, without noticeable deviations. Confidence intervals overlap.

This uniformity is somewhat disturbed in the humus horizon; thorium concentration at the southern regions (Razboynaya and

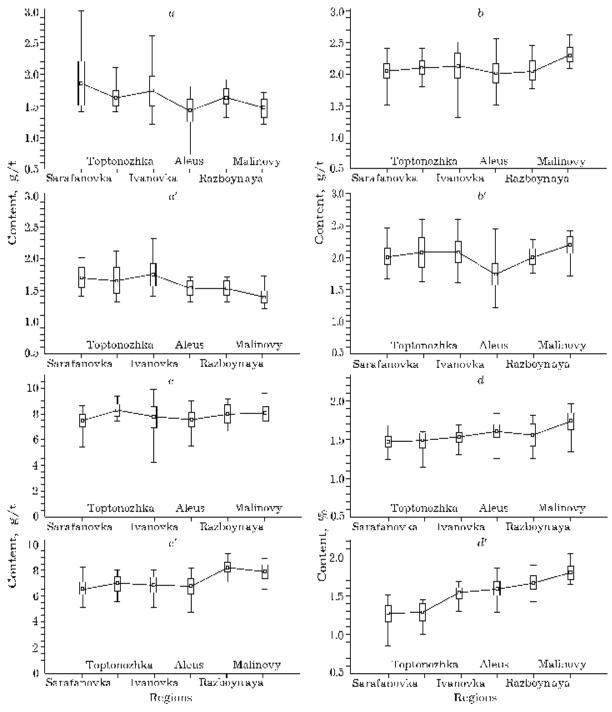


Fig. 4. Changes in the mean values, confidence intervals and ranges of NREA values in the substrate (a-d) and in humus horizon (a'-d') of autonomous landscapes: a, a' – uranium; b, b' – thorium; c, c' – uranium calculated from radium; d, d' – potassium.

Malinovy) increases. Passing from the subsoil substrate to the humus horizon at the Sarafanovka – Aleus grounds the dilution of the upper horizon with humus occurs; thorium concentration decreases by 10–15 %. At the grounds situated to the south (Razboynaya and Malinovyi) thorium content is at the same level.

### Potassium

Changes in potassium concentration in the soils of the typical grounds of the transect are shown in Fig. 4, *d*, *d*'. Our investigations show that potassium provides the strongest response to hydrothermal zoning. A clear trend of

increase in potassium concentration is directed from north to south. A reason of a decrease in potassium concentration is atmospheric precipitation. Potassium is a typical alkaline element which easily passes into aqueous solutions. Potassium content exhibits an inverse correlation with the amount of precipitation (correlation coefficient is 0.90 for the humus horizon and 0.91 for the subsoil substrate).

## Uranium carry-over

A decrease in uranium content is more clearly observed with geochemical markers. The Th/U ratio increases from 4.03 to 5.13 in the humus horizon and from 4.39 to 5.59 in the subsoil substrate. The U/U(Ra) ratio decreases from 0.84 to 0.62 in the humus horizon and from 0.90 to 0.66 in the subsoil substrate. The Th/U(Ra) ratios characterize the Th/U ratio in the initial moment of time (before redistribution of the matter under the action sol-forming processes). The trend of ratio in the humus horizon somewhat increases, while in the subsoil substrate it stays at the same level. This is one more evidence of initially uniform distribution of the NRAE in loess (Fig. 5).

Using geochemical markers we may estimate the amount of uranium carried over during the Holocene warming period from the regions under investigation. Estimating uranium carryover with the help of the first marker, the uranium concentration obtained by laser luminescence method (actual present concentration of uranium) is subtracted from the uranium concentration obtained by means of gamma spectrometry (the initial uranium concentration in the loess cover). The difference in uranium concentrations is calculated for a layer 50 cm thick and having the area of 1 m<sup>2</sup>. Since we tested only the first meter of the soil eluvial profiles, calculation of the carry-over was carried out for this meter. We accepted mean thickness of the humus horizon to be 50 cm, its density 1.6 g/cm<sup>3</sup>, and the same thickness of the subsoil substrate layer (50-100 cm), with the density of 2.6 g/cm<sup>3</sup>. So, we obtain the resulting amount of uranium carried over during Holocene from the humus horizon (0-50 cm) and the subsoil substrate (50-100 cm).

The second marker which is used to estimate uranium carry-over is the Th/U ratio. A quantitative estimation based on this marker was made as follows. For the substrate, U(Ra) and thorium values (2.10 and 7.85 g/t) were averaged and their ratio was calculated (3.74). These values were taken as the characteristics of the initial substrate. After that, using the deviation of Th/U we calculated changes in uranium concentration (thorium concentration was accepted to be constant). Further calculations were carried out in the same manner as for the first marker.

A plot of uranium carry-over in different landscape-climatic zones is shown in Fig. 6 (dots indicate mean values, lines and hatching show confidence intervals). One can see that estimations

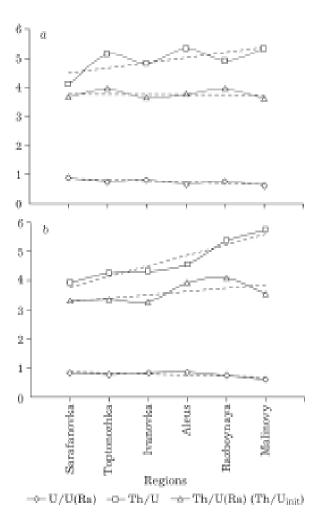


Fig. 5. Changes in the values of geochemical markers in autonomous landscapes of the «open» grounds of transect: a – soil-forming substrate, b – humus horizon; dash curves – trend lines.

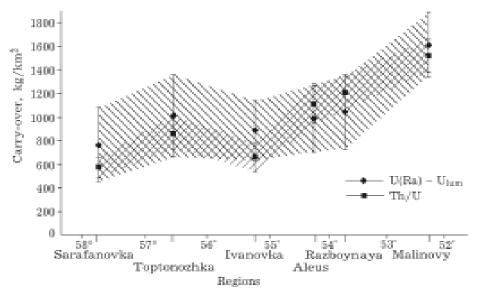


Fig. 6. Uranium carry-over from the outer layer 1 m thick during Holocene, estimated using the values of geochemical markers.

obtained with different markers correlate with each other. Maximal carry-over occurs in the steppe zone where it reaches 540-600 kg from the layer of subsoil substrate 1 km<sup>2</sup> in area, and 540-560 kg from the layer of the humus horizon 1 km<sup>2</sup> in area. Differences between the neighbouring sites are stronger expressed in the subsoil substrate, but generally the trend of an increase can be described by a straight line. The plot of uranium carry-over from the humus horizon can be divided into two parts: the first one goes from Sarafanovka to Ivanovka, the second one from Aleus to Malinovy. The reason is soil types: in the northern part there are meadow and forest soils, while in Aleus and further to the south the soils of autonomous landscapes are represented mainly by black earth.

At the concluding stage, we compared the obtained estimations with the modern uranium carry-over by the rivers. Carry-over from the eastern part of the south of West Siberia was estimated as follows. Water flow through the Ob' river near the Kolpashevo is  $4170 \text{ m}^3/\text{s}$  [18]; mean uranium concentration in the Ob' is accepted to be  $10^{-6}$  g/l [19]. Thus, the upper flow of the Ob' river carries 132 t of uranium per year. Since we are interested in the plain part, we are to subtract 17 t of uranium delivered from the Altay mountains and foothills (according to the data of the Fominskoye river post at the junction of the Biya and the Katun',

water flow is  $1090 \text{ m}^3/\text{s}$ , while the concentration is the same). Extrapolating these data for the whole Holocene we obtain  $9.8 \cdot 10^5$  t of uranium.

According to our data, mean carry-over from the upper meter of the soil of the taiga and forest-steppe zones is 800 kg/km² (the steppe part of the Altay belongs to drainage area of the Irtysh river and the lakes of the Kulunda and Barabinsk plains). Multiplying this value by the area of the drainage area of the steppe part of the Ob' river, which is 387 800 km² [18], we obtain an estimated value 3.1 10⁵ t of uranium. In our opinion, these estimations obtained using independent data are quite comparable because the thickness of the zone of active water exchange from which uranium is carried out several times exceeds the first meter of the soil.

# CONCLUSION

One might expect establishment of clear radiogeochemical zoning in the south of West Siberia during Holocene; however, in reality this is observed only for potassium and to a lesser extent for uranium. There are several reasons for this. First, the boundaries of the landscape-climatic zones shifted many times during the soil formation period both to the north and to the south from their modern

positions; more inert hypergenesis processes are too slow to bring the distributions of the elements under investigation into the agreement with the current climatic situation. Second, aeolian transport goes on leveling nonuniformities of the soil substrate. This process has been intensified especially during the recent decades due to plowing up the major part of the forest-steppe and steppe zones. Third, the surface flows wash the soil particles from the autonomous and transitional landscapes and bring them into the subordinate landscapes masking the effect of climatic factors. Because of this, the most clear difference in the behaviour of NRAE in different landscapeclimatic zones is observed in the autonomous landscapes.

Maximal uranium carry-over is observed in autonomous landscapes of the steppe, and especially dry steppe. This carry-over occurs both from the humus horizon and from the subsoil substrate; it is observed not only as a decrease in uranium concentration from the north to the south but also as uranium to thorium and uranium to radium ratios. The reason is an increase in the concentration of hydrocarbonate ion in the southern regions.

Potassium concentration increases from north to south in all the types of elementary landscapes by 20~% as a mea; in the subsoil substrate and in the humus horizon this happens together with an increase in the amount of precipitation.

The described estimation characterizes carryover from the first meter of the soil-eluvial profiles. Using the proposed approaches one may estimate uranium carry-over from the entire zone of hypergenesis and make reliable forecasts of uranium behaviour in industryrelated anomalies formed during mining and processing radioactive ores (including the tailings of the plants of nuclear fuel cycle); it is also possible to model the changes in the intensity of geochemical processes under the changing climatic conditions.

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