Anomalies of Radioactivity on the Southern Bank of the Ysyk-Köl Lake (Kyrgyzstan)

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Abstract

On the basis of simultaneous gamma spectroscopic determination of radionuclides of the uranium series (^{238}U, ^{226}Ra, ^{222}Rn, ^{210}Pb) and isotopes arriving from the atmosphere ($^{210}Pb_{atm}$ and ^{137}Cs) in rocks and bottom sediments, radiogeochemical background of land sediments represented mainly by granitoid weathering products was obtained (35–55 Bq/kg), along with evidences of the hydrodynamic mode of sediment formation within the recent century. Three types of radioactivity anomalies exceeding the background by one-two orders of magnitude were revealed on the southern bank of the lake: natural uranium and radium anomalies; industry-related radium anomalies (ash obtained from burning uranium-bearing coal); industry-related uranium anomalies. Sedimentation rate was estimated in the deep water region using the decay of $^{210}Pb_{atm}$ activity to be 0.2–0.4 mm/year during the recent century. On the basis of uranium to radium ratio, it was established that the fraction of uranium in deep water sediments arriving from the lake water is 1.5–2 times higher than the fraction arriving with the suspension. One of the sources of uranium in the lake water was erosion of uranium-bearing coal within the geological time scale. A layer enriched with radium was discovered near the bank at the depth of 5 to 20 cm. The presence of high-temperature mineral mullite in the sediment is an evidence of the penetration of industry-related radioactive ash into the lake in amounts that can hardly be dangerous for the biogeosystem of the lake Ysyk-Köl.

INTRODUCTION

As V. V. Koval'sky demonstrated in 1974 [1], the Ysyk-Köl intermountain trough is a biogeochemical province in which all the components of the environment are enriched with uranium: rocks, soil, lake, river and underground water, lake sediments and living organisms [2]. He assumed that the source of uranium is granite and carbonaceous-siliceous shale of the surrounding height. Only in 1996 information appeared on the Internet concerning the existence of radioactive wastes accumulated at the southern bank of the lake during exploitation of the uranium-coal deposit later abandoned [3]. Since there are sanatoriums, famous resort Cholpon-Ata, holiday homes and tourist bases on the banks of the lake, which are popular far off Kyrgyzstan, anxiety arose in the international community concerning the future and in particular the possibility of radioactive pollution of the lake.

In order to estimate modern status of the lake Ysyk-Köl and outlooks of changes in its water balance and chemical composition, including the level of radioactivity, international project APELIK "Assessment and Prognosis of Environmental Changes in Lake Ysyk-Köl (Kyrgyzstan)" was developed and supported by the European Commission within the program Copernicus-2. Investigations of radioactivity outlined in the project started at the Analytical Centre of the UIGGM, SB RAS (Novosibirsk) in September 2001 and continued in 2002.

INSTRUMENTATION AND PROCEDURES

Determination of ^{210}Pb, ^{226}Ra, ^{238}U and ^{137}Cs isotopes in the samples under investigation was carried out by means of direct high-resolution semiconductor gamma spectrometry. The method was adapted to the well-type semiconductor detector (SD). In order to check the operation capacity of the developed procedure, we used the results obtained by means of scintillation gamma spectroscopy (^{226}Ra and ^{137}Cs) and XPA (^{238}U).

Traditionally, the content of ^{238}U is estimated basing on daughter products ^{214}Pb and ^{214}Bi. These isotopes are the first in the ^{238}U series the decay of which gives rather intensive gamma lines: 242.0 keV (the yield of the line is 7.5 %), 295.2 keV (18.5 %), 351.9 keV (35.8 %) for ^{214}Pb and 609.3 keV (44.8 %), 1120.3 keV (14.8 %) 1764.5 keV for ^{214}Bi. If we admit that ^{238}U and products of its decay along the whole uranium series are in the state of radioactive equilibrium (which is achieved in reality within not more than 1 mln years), then, determining the activity of ^{214}Pb and ^{214}Bi isotopes we actually obtain also the information about ^{238}U through ^{222}Rn and ^{226}Ra. Such an approach to measurements, though relative, provides estimation of ^{238}U content with high reliability for the major part of rock types. Distortion of radioactive equilibrium in the chain cannot be excluded during various geological and geochemical processes. These distortions can be connected both with possible ejection and with the arrival of long-lived elements: ^{238}U ($T_{1/2}$ = 4.47 10^9 years), ^{234}U (2.455 10^5 years), ^{230}Th (7.54 10^4 years), ^{226}Ra (1600 years) and also relatively short-lived ^{222}Rn (3.82 days). If one does not take into account these distortions, he may disfigure the results obtained not only for ^{238}U but even for ^{226}Ra, on the basis of which uranium is in fact estimated.

With the direct gamma spectrometric method, determination of the content of radioisotopes is carried out directly using the lines of their own gamma radiation. An example of this approach is determination of ^{137}Cs on the basis of the line 661.7 keV (85.1 %). According to their nature, ^{238}U and ^{226}Ra are purely alpha radiating elements, while ^{210}Pb is beta radiating element. The spectra of their radiation contain no strong gamma lines. Nevertheless, low-energy gamma quanta are formed during their decay, along with alpha particles for ^{238}U and ^{226}Ra and beta particles for ^{210}Pb. For instance, the spectrum of ^{226}Ra contains eigen line 186.1 keV (3.5 %), ^{210}Pb – the line 46.5 keV (4.25 %). The spectrum of ^{238}U contains no eigen gamma lines having essential yield. However, ^{234}Th isotope, being a direct product of the decay of ^{238}U, has such a line: 63.3 keV (4.8 %). Due to short half-life of ^{234}Th (24.1 days), it will always be in radioactive equilibrium with ^{238}U. Because of this, in the absence of actually proceeding separation of uranium from thorium, it is quite reasonable to estimate the content of ^{238}U on the basis of ^{234}Th content. The yields of analytical gamma lines which can serve to determine ^{238}U, ^{226}Ra and ^{210}Pb directly are small; this fact, along with substantial absorption coefficients of these lines in the volume of samples under analysis and detector material, requires the use of recording systems possessing rather high efficiency and energy resolution in the low-energy region of gamma spectrum.

To record analytical gamma lines, in the present work we used gamma spectrometric system composed on the basis of the well coaxial HPGe SD with low-background cryostat EGPC 192-P21/SHF 00-30A-CLF-FA of EURISYS MEASURES Co. (France). Active volume of the detector was 200 cm^3, absolute efficiency of recording 15 %, resolution with respect to line 1332 keV was 2.2 keV, and with respect to line 122 it was 1.4 keV, well dimensions were 21×60 mm. Due to large working volume and the presence of a well inside the active region, allowing one to carry out measurements in the geometry approaching 4π, the detector is characterized by high efficiency of recording gamma quanta within a broad energy range from 20 to 2000 keV. Judging from the design, this detector relates to semiconductor detectors of P type. Such detectors are characterized by a maximum on the curve of recording efficiency for gamma quanta with energy within the range 20 to 150 keV. When a sample with the mass 1 to 15 g is placed inside the well, the efficiency of recording gamma quanta with the energies 46.5, 63.3 and

186.1 keV achieves 50–60 %, for the line 661.7 keV it was 7–8 %. The application of usual coaxial and planar SD for solving the same tasks in our previous experiments showed that the efficiency of recording with small weighed portions was an order of magnitude lower. The use of small weighed portions of 1–15 g to determine the activity of radioisotopes at a level of 0.05–0.5 Bq, corresponding to uranium content of the sample 0.3–3 mg/kg, imposes rigid requirements on the background of the measuring installation. In order to decrease the background of the spectrometer, which was due to the action of external background gamma radiation, we used a combined protection consisting of the external and internal circuits. The external circuit is a standard 10 cm thick protective layer made of radiationally pure lead, reinforced by a layer of tungsten 12 mm thick. The internal circuit, which is intended for additional decrease in background in the low-energy part of the gamma spectrum ($E\gamma < 100$ keV) and is a special case made of cadmium and copper, is located directly around the measuring unit of the detector. In order to decrease the contribution from the background component brought about by the gamma-radiating products of radon decay, the detector was placed in a special chamber with permanent flow of the atmospheric air taken at the height of 6 m from the surface. Such a design of the protection allowed us to decrease the background of the spectrometer more than 200 times in comparison with the geometry in the case of the open detector.

For direct determination of the content of ^{210}Pb, ^{226}Ra, ^{238}U and ^{137}Cs isotopes in the samples under investigation by means of direct gamma spectrometry, the procedure of the analysis of small weighed portions with a mass 1 to 15 g and volume 1 to 10 cm^3 was developed. The sample under investigation was placed in a special cell and put into the detector well for measurement. Time of measurement of a single sample was from 1–2 to 24 h, depending on the activity (content) of the isotopes under determination.

The key stage in the procedure is determination of the coefficient of recording efficiency for analytical lines which are used to calculate the content of isotopes. The algorithm of calculation of these coefficients takes into account the effect of many factors and includes the following procedures: estimation of the contribution from background and the background lines; calibration over the geometry of measurement, connected with the necessity to work with the samples differing in volume; accounting for the density of the sample or its effective atomic number in order to compensate for the losses arising during absorption and scattering of gamma quanta, especially low-energy ones, in the material of a bulk sample; subtraction of the contribution from interfering lines. The latter is urgent for the determination of ^{226}Ra using the line 186.1 keV, since the radiation from the line of ^{235}U 185.7 keV (57.2) is imposed on this line. The energy resolution of the direct spectrometric tract was insufficient for direct separation of these lines, so a special procedure of separation of the fraction related to line ^{226}Ra from the total peak was necessary. This procedure is based on constancy of ^{235}U : ^{238}U ratio in natural objects. Knowing half-lives of ^{238}U, ^{235}U and ^{226}Ra, yields of their gamma lines, and accepting that the coefficients of recording efficiency for lines 186.1 and 185.7 keV differ only slightly, one may calculate the contributions from ^{226}Ra and ^{235}U into the overall line. For the system in radioactive equilibrium, these contributions are 56.76 and 43.24 %, respectively. Taking into account this relation, when measuring the equilibrium uranium reference samples described below, we build up an empirical dependence between the intensity of lines 63.3 keV (^{234}Th) and 185.7 keV (^{235}U), which is further used to calculate the contribution of the latter into the overall line. The algorithm of determination of recording efficiency coefficients was adjusted using a set of uranium reference samples which were mixtures of standard sample of the uranium equilibrium silicate ore UR-47C with specially pure quartz, activated carbon and wood cuts. The content of ^{238}U in these reference samples was about 50 mg/kg.

Estimation of the main metrological characteristics of the developed procedure demonstrated that the reproducibility of the method for the analysis of samples with the vol-

ume of 10 cm^3, mass 10 to 15 g, measurement time 12 h and activity of the isotopes under determination 100–120 Bq/kg according to the 2 sigma criterion was ±5 % for ^{137}Cs, ±8 % for ^{210} Pb and ^{238}U, ±10 % for ^{226}Ra. These analysis reproducibility values do not take into account the possibility of non-uniformity of the distribution of isotopes under determination within the sample volume. The correctness of the method was estimated using both the results of measurements of the reference rock samples BIL-1, BIL-2, ZUK-1, SDO-1, SG-1A, DVG, DVT, DVR, reference sample of the International Atomic Energy Agency IAEA-135 and comparison with the results obtained using other methods (scintillation gamma spectrometry and XPA). For comparisons, mass estimations of uranium obtained on the basis of XPA (SR) were recalculated into activity units using a known equation: $C = 2.8\ 10^{-6}\ ATQ$ where C is the mass of uranium; A is mass number; T is half-life, years; Q is the mass of uranium in the sample, calculated according to the data of XPA (SR). Good convergence of the results with the certificate and previously obtained values was demonstrated, as well as the absence of essential systematic error of analysis. Detection limits under the conditions indicated above were not more than 0.03 Bq for the isotopes of uranium series, which corresponds to the uranium content in the sample at a level of 0.25 mg/kg, and for ^{137}Cs – 0.01 Bq (1 Bq/kg).

RADIOGEOCHEMICAL BACKGROUND AND INDUSTRY-RELATED ANOMALIES OF RADIOACTIVITY

The southern bank of the Ysyk-Köl lake is cleaved by branchy valleys of temporary water flows (sais). The distance along the bank between the mouths of two neighbouring says (Jilubulak Say and Ak Say) does not exceed 3 km. At the distance of 2.5 km from the lake, the summits of their side tributaries approach each other; there, gypsometrically at the height of 180 m above the lake surface, the layers of radioactive brown coal of Jurassic formation emerge on the surface. The bottoms of the valleys are composed mainly of coarse detrital sediments – sand, gravel, gruss, almost devoid of soil cover. Finer grained aleuric clayish sediments coated with a thin layer of humus-containing soil occur only on rare small

TABLE 1

Radioactive element content accepted as radiogeochemical background (the data of scintillation gamma spectrometry)

Rocks, sediments	*n*	U (with respect to Ra)*		Th, μg/g	*K*, %	Th/eU	Emanation coefficient, %**
		μg/g	Bq/kg				
Assumed sources of the sediment material							
Granites	4	3.7 ± 2.2	46.5	15.2 ± 4.9	3.33 ± 0.57	4.1	
Soil profile (Barskaun pass)	8	5.1 ± 0.6	64.2	13.6 ± 0.9	2.46 ± 0.14	2.7	19.9 ± 6.8
Bottom sediments of Jilubulak Say							
Coarse-grained quartz-feldspar sand (granite weathering products)	23	2.9 ± 0.2	36.5	8.7 ± 0.8	3.30 ± 0.10	3.8	10.4 ± 3.2
Bottom sediments of Ak Say							
The same	45	4.4 ± 0.4	55.3	10.3 ± 0.9	3.15 ± 0.06	2.3	8.6 ± 2.3

Note. n is the number of samples.
*Estimations of uranium content and activity on the basis of radium activity, assuming radioactive equilibrium.
**$Ra_2 - Ra_1$ in % of Ra_2, where Ra_1 and Ra_2 is activity in an open and tight sample, respectively.

TABLE 2

Activities (Bq/kg) and quantitative relations between radionuclides in bottom sediments of the valleys of temporary water flows (the data of semiconductor gamma spectrometry)

Sample	^{238}U	^{226}Ra	^{210}Pb	^{232}Th	^{137}Cs	^{226}Ra / ^{238}U	^{210}Pb /^{226}Ra	^{210}Pb /^{137}Cs
				Jilubulak Say				
IK_{00}–1/3	34.3	33.4	28.4	24.2	5.7	0.97	0.85	5.0
IK_{00}–4/2	29.1	30.5	36.1	39.0	5.9	1.05	1.18	6.1
IK_{00}–5/1	31.3	31.1	28.9	39.9	2.8	0.99	0.93	10.3
IK_{00}–7/1	19.9	22.4	15.3	29.6	3.1	1.12	0.68	4.9
				Ak Say				
IK_{01}–44/3	35.2	43.2	44.8	43.9	4.5	1.23	1.04	9.9
IK_{01}–48/1	35.4	48.8	34.0	38.7	1.6	1.38	0.70	21
IK_{01}–49/2	32.8	51.5	37.2	41.1	5.0	1.57	0.72	7.4
IK_{01}–50/3	38.2	40.0	30.8	44.5	2.4	1.05	0.77	12.8
IK_{01}–51/2	42.5	40.5	36.0	44.1	3.3	0.95	0.89	10.9
IK_{01}–56/1	41.8	69.1	77.0	47.6	4.1	1.65	1.11	18.8
IK_{01}–64/3	73.5	110.4	84.6	60.0	11.9	1.50	0.77	7.1
Mean	37.6	47.3	41.2	41.1	4.7	1.22	0.88	10.4
Background values* (Table 1)		36.5 55.3		35.4 41.9				

*Upper value is for the Jilubulak Say, lower for the Ak Say.

terraces. Prevailing matter of the coarse sediments consists of weathering products of granites; their mean radioactivity is accepted as the local radiogeochemical background (Table 1).

Estimation of uranium content on the basis of radium is usually made using scintillation gamma spectrometry. However, comparison with the data of semiconductor technique indicates that the uranium to radium ratios substantially deviate from the equilibrium in soil and in bottom sediments (Table 2).

So, we accept the background value of uranium-radium activity to be within the range 35–55 Bq/kg.

Anomalies of three types are distinguished above the level of the radioactive background.

1. Rare outcropping of radioactive coal from under loose deposits. Coal beds are strongly deformed and lie nearly upright.

2. Friable fine-grained gray-coloured mass limited from all the sides with a concrete wall.

3. Aleuric-clayish sediments in a chain of six artificial settling reservoirs connected with pipes (Fig. 1). Flat surfaces of the reservoirs are almost devoid of vegetation.

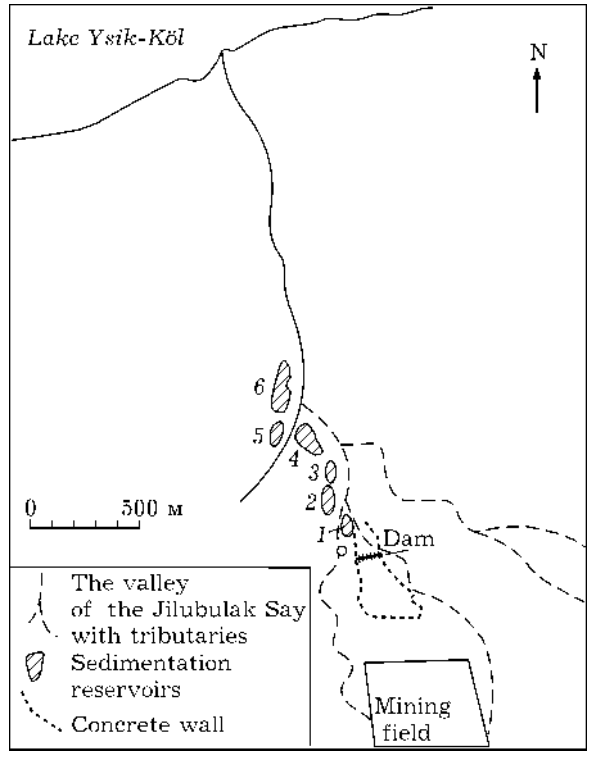

Fig. 1. Map of the sites investigated in Jilubulak Say: *1–6* – sedimentation reservoirs.

TABLE 3

Relations between ^{238}U and ^{226}Ra in coal and in waste material

Sample	Ra^1		Ra^2		U^2		$\frac{A(Ra^2)}{A(U^2)}$	U^3	
	pg/g	*A*, kBq/kg	pg/g	*A*, kBq/kg	μg/g	*A*, kBq/kg		μg/g	*A*, kBq/kg
Anomalies of the first type (coal and waste with coal particles), A(Ra) ~ A(U)									
Ikoo-31/1	46	1.7	63	2.3	128	1.6	1.4	134	1.7
Ikoo-31/2	115.6	4.3	102	3.8	190	2.4	1.6	254	3.2
Ikoo-33/1	442	16.3	340	12.6	1200	15.1	0.8	1105	13.9
Ikoo-34/1	34	1.2	32	1.4	110	1.4	1.4	63	0.79
Ikoo-34/2	26	0.95	22	0.78	58	0.73	1.07	49	0.62
Ikoo-35/1	30	1.12	30	1.12	87	1.09	1.03	75	0.94
Ikoo-58/1	574	21.2	564	20.9	1090	13.7	1.5	1117	14.0
Anomalies of he second type (hot waste – ash), A(Ra) >> A(U)									
Ikoo-39/1	144.8	5.4	136	5.0	65	0.82	6.1	48	0.60
Ikoo-40/1	372	13.8	306	11.3	1120	14.1	0.8	978	12.3
Ikoo-41/1	1316	48.7	1003	37.1	640	8.0	4.6	726	9.1
Ikoo-42/1	801	29.6	765	28.3	420	5.3	5.3	543	6.8
Ikoo-43/1	814	30.1	653	24.1	500	6.3	3.8	563	7.1
Ikoo-44/1	809	29.9	714	26.4	420	5.3	5.0	550	6.9
Ikoo-46/1	749	27.7	1003	37.1	450	5.7	6.5	494	6.2
Ikoo-47/1	286	10.6	279	10.3	130	1.6	6.4	140	1.8
Ikoo-47/2	138	5.1	139	5.2	106	1.3	4.0	109	1.4
Anomalies of the third type (clay and aleurite from the upper sedimentation reservoirs), A(Ra) << A(U)									
Ikoo-29/1	3.0	0.11	Sedimentation reservoir No. 1, depth: 0–120 cm					9.6	0.12
Ikoo-29/2	2.5	0.09						151	1.9
Ikoo-29/3	2.1	0.08						121	1.5
Ikoo-29/4	2.4	0.09						41	0.51
Ikoo-27/1	3.0	0.11	Sedimentation reservoir No. 2, depth: to 40 cm					70	0.88
Ikoo-27/2	2.1	0.08						203	2.5
Ikoo-27/3	2.7	0.10						141	1.8
Ikoo-27/4	2.2	0.08						192	2.4
Ikoo-28/1	3.0	0.11						68	0.88
Ikoo-24/1	1.4	0.05	Sedimentation reservoir No. 3, depth: 0 to 15 cm					63.8	0.80
Ikoo-25/1	3.2	0.12						10.6	0.13
Ikoo-25/2	2.0	0.07						45.3	0.57
Ikoo-26/1	2.7	0.10						72	0.90
Ikoo-22/1	2.7	0.10	Sedimentation reservoir No. 4					22.9	0.29
Ikoo-22/2	3.1	0.12						19.4	0.24
Ikoo-22/3	1.9	0.07						53.2	0.67
Ikoo-22/4	2.1	0.08						11.2	0.14
Ikoo-22/5	3.2	0.12						6.9	0.09
Ikoo-23/1	1.7	0.06							
Background radioactivity (clay and aleurite from two lower sedimentation reservoirs), A(Ra) ~ A(U)									
Ikoo-18/1	2.3	0.09	Sedimentation reservoir No. 5, depth: to 50 cm					5.5	0.07
Ikoo-18/2	1.6	0.06						3.8	0.05
Ikoo-19/1	2.5	0.09						5.8	0.07
Ikoo-20/1	2.4	0.09						6.0	0.07
Ikoo-15/1	2.8	0.10	Sedimentation reservoir No. 6, depth: 0 to 45 cm					6.7	0.08
Ikoo-15/2	2.4	0.09							
Ikoo-15/3	3.0	0.11						4.7	0.06
Ikoo-15/4	1.3	0.05						7.7	0.10

Notes. 1. *A* is activity, kBq/kg. 2. Analytical methods: Ra^1 – gamma spectrometry, NaI(Tl) detector, sample mass 100–450 g; Ra^2 – gamma spectrometry, Ge–Li coaxial and Ge planar detectors, sample mass 100 g; U^2 – the same analysis conditions; U^2 – XPA with synchrotron radiation, sample mass 30 mg.

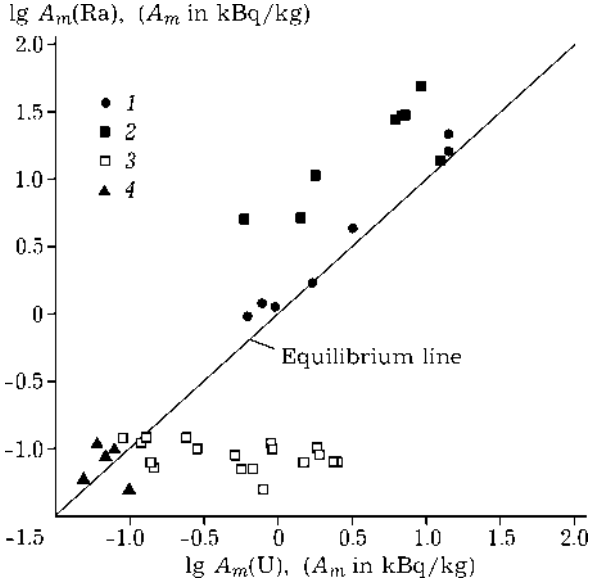

Fig. 2. Relations between uranium and radium in the investigated objects (A_m is specific activity of the source): *1* – coal and sand with coal particles; *2* – ash depleted of uranium; *3* – sedimentation reservoirs enriched with uranium; *4* – two lower sedimentation reservoirs (background).

As investigations indicated, three types of radioactive anomalies are sharply different from each other in radionuclide composition (Table 3).

Anomalies of the first type. They are of natural origin. Uranium-bearing coal of Jurassic age was traced in the past to the depth by wells and extracted through mines. Uranium is distributed in coal extremely non-uniformly: along with high content (up to 0.1–0.4 %), layers weakly enriched with uranium occur (100 g/t and lower). Waste material containing the particles of this coal remained around the mine. These natural anomalies are characterized by comparatively insignificant (not more than by a factor of 1.6) deviations to one side or another from the equilibrium uranium to radium ratio (see Table 3).

Anomalies of the second type. These are mainly radium anomalies in which the activity of radium is 4–6 times higher than that of uranium. Clayish minerals are not detected in the fine-grained gray matter by means of X-ray diffraction, but mullite is observed; this high-temperature mineral provides evidence of roasting of the aluminosilicate matter. This is likely to be ash remaining after burning uranium-bearing coal, from which uranium was extracted; high specific activity of radium can allow considering high initial concentration of uranium in the burnt coal (see Table 3, Fig. 2).

Anomalies of the third type. These anomalies are discovered in the chain of settling reservoirs with an even flat surface. Samples from these settlement reservoirs were taken with a steel ring from the depth of 20–40 cm and more, with 5 cm intervals. In fine-grained aleuric clayish substance of the settling reservoirs, prevailing are quartz, feldspar, calcite and clayish minerals. As opposed to the anomaly of the second type, the activity of uranium in settling reservoirs is higher than that of radium; it decreases from the upper reservoir to the lower ones, the activity of radium remaining the same at a level corresponding to the background values (Fig. 3).

On the route from radioactive anomalies to the lake, in the sediments of Jilubulak Say and Ak Say, any clear traces of migration of radioactive elements are absent. Only microscopic fragments of brown coal with uranium content 13–63 g/t occurred rarely in sand.

QUANTITATIVE RELATIONS BETWEEN ^{238}U, ^{226}Ra AND ^{210}Pb AS THE TRACES OF INDUSTRY-CAUSED PROCESSES IN THE LAKE SEDIMENTS NEAR THE BANKS

Though no evidence of migration of radioactive wastes was discovered on the lake bank,

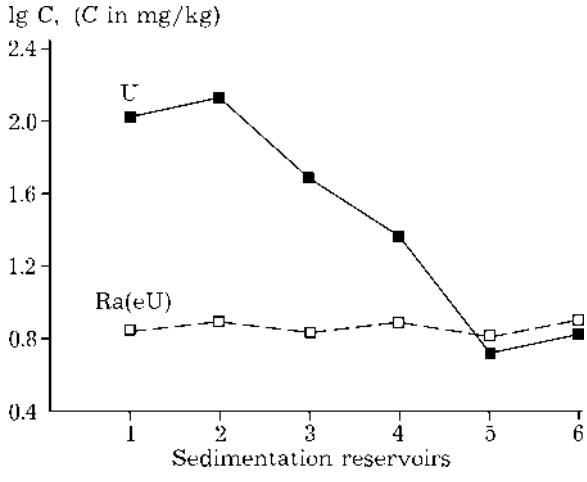

Fig. 3. Relations between uranium and radium in the cascade of sedimentation reservoirs from the upper one (1) to lower one (6).

their undoubted presence was established in shallow-water sediments. Sediments of two types were found in several columns which were examined.

1. To the west from the mouth of Jilubulak Say, sand sediments are covered with clayish ones, in which the concentrations of the radionuclides of atmospheric origin (^{137}Cs and ^{210}Pb) subexponentially decrease top-down, which points to calm continuous sedimentation during at least 50 years (Fig. 4, stations IK_{01}–12 and IK_{01}–3). A similar distribution is characteristic of deep-water sediments in the northern part of the lake Ysyk-Köl (see Figs. 4, 6, stations IK_{01}–1 and IK_{01}–3); of course, they contain no traces of industry-related material.

2. To the east of the mouth of Jilubulak Say, where the suspensions are shifted by currents, sedimentation regime was distorted. Here ^{137}Cs and $^{210}Pb_{atm}$ were not accumulated on the surface of the sediment, but got distributed uniformly during mixing. In addition, at a depth 5 to 20 cm from the sediment surface, a layer is detected, which has no clear visible characteristics but is sharply enriched with radium and ^{210}Pb, a product of its decay, to a higher extent than with uranium (Figs. 5, 6, stations IK_{01}–13, 14, 16, 17). The presence of mullite in this layer makes one sure that this is ash enriched with radium. It was likely to be brought into the lake before the concrete wall limiting ash storage area was built.

ESTIMATIONS OF AUTHIGENIC CONSTITUENT OF TOTAL URANIUM CONTENT ON THE BASIS OF NON-EQUILIBRIUM RELATIONS BETWEEN ^{238}U AND ^{226}Ra IN DEEP-WATER SEDIMENTS

With the example of the column excavated in 2001 at the station IK_{01}–1 in the northern part of the lake from the depth of 336 m, on the basis of radionuclide ratio, it is possible to separate the part of uranium, which arrived into the sediment from lake water, from the part brought with terrigenous material deposited from the suspension. Calculating using the ratio

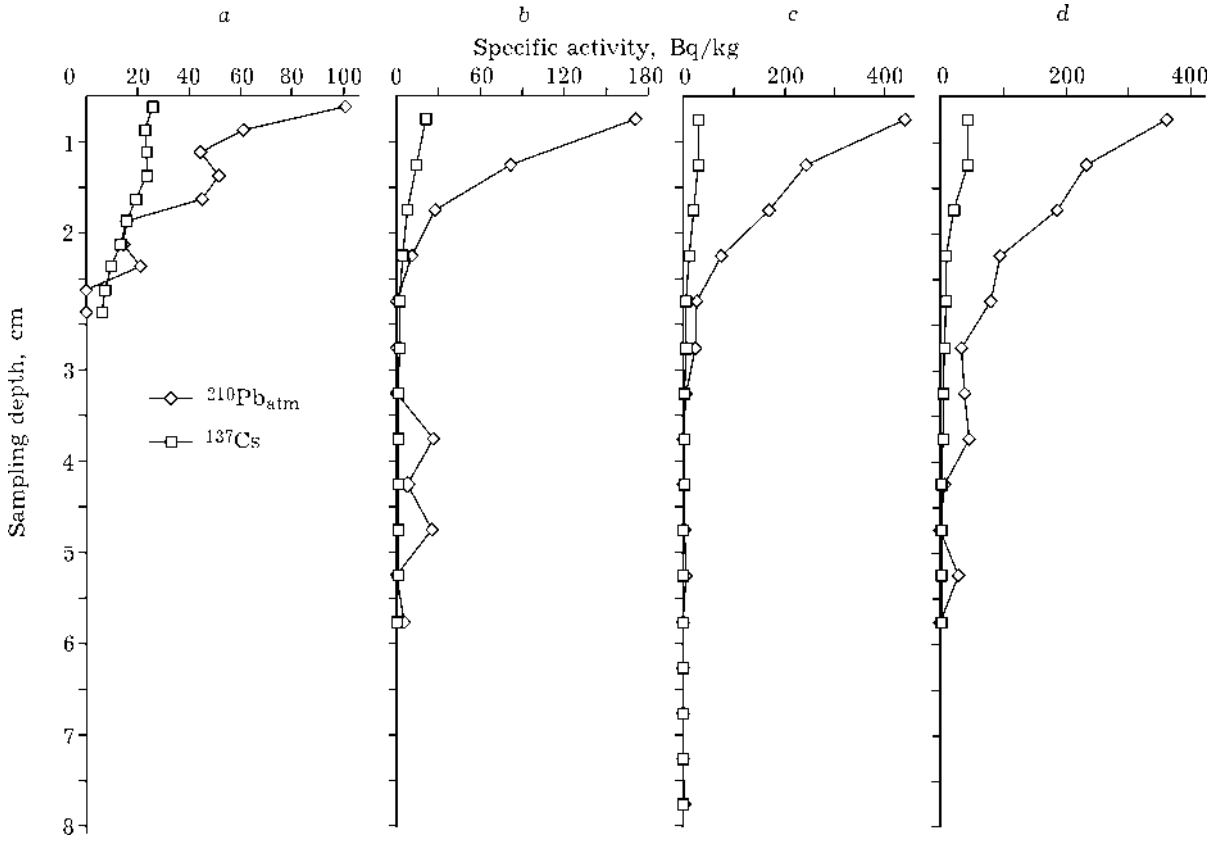

Fig. 4. Distribution of ^{210}Pb and ^{137}Cs over columns of bottom sediments in the regions of calm sedimentation (columns IK_{01}–12 and IK_{01}–22 in the littoral zone, IK_{01}–1 and IK_{01}–3 in deep water): *a* – IK_{01}–12, *b* – IK_{01}–22, *c* – IK_{01}–1, *d* – IK_{01}–3.

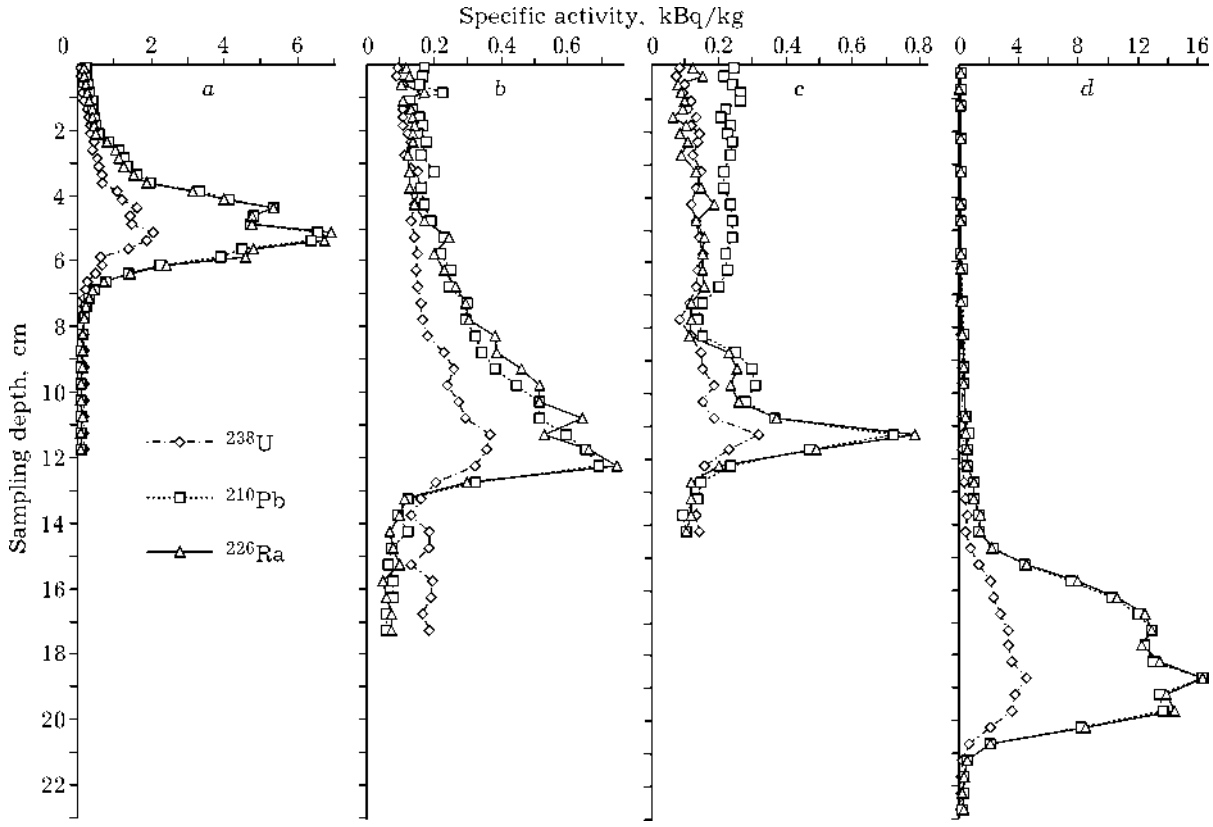

Fig. 5. Radium anomalies in the lake sediments (admixture of radioactive ash at different depth in the zone of distorted sedimentation): *a* – IK_{01}–13, *b* – IK_{01}–14, *c* – IK_{01}–16, *d* – IK_{01}–17.

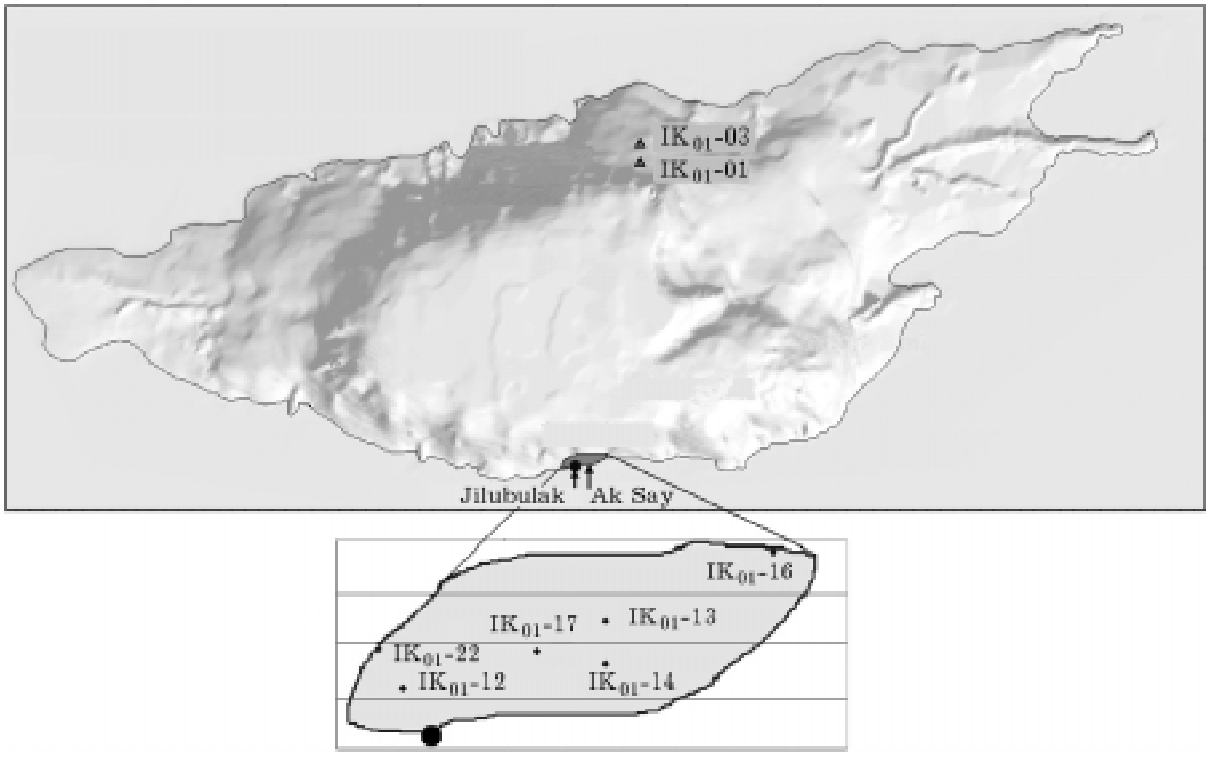

Fig. 6. Scheme of the location of investigated columns in the area of water of the lake Ysyk-Köl.

of radium to uranium mass, which is $3.4 \cdot 10^{-7}$, we obtain the amount of radium in equivalent units of uranium under the conditions of radioactive equilibrium (U_{eq}). Then, the difference between total amount of uranium (U_{tot}) and its part in equilibrium with radium (U_{eq}) can be accepted as an estimation of the excess (authigenic) uranium (U_{exc}) which entered the sediment from the lake water. It is indicative that the distribution of equilibrium uranium over the column is similar to the distribution of thorium (Fig. 7); this is quite understandable since both radionuclides arrive into the sediment mainly in the same accessory minerals.

DISTRIBUTION OF ^{210}Pb AND ^{137}Cs IN DEEP-WATER SEDIMENTS AS EVIDENCES OF HYDRODYNAMIC REGIME AND SEDIMENTATION RATE

It is known that both these radionuclides with close half-lives (22 and 30 years, respectively) get deposited from the atmosphere with suspension particles; further on, their activity decreases in agreement with the law of radioactive decay. However, since they originate from different sources, their distribution over the columns of sediments is not identical. Ideally, maximal ^{137}Cs should be at the stratigraphic level corresponding to 1963, when the agreement on atmospheric nuclear test ban was adopted; it must be absolutely absent from the sediments formed before 1949. In reality, it is usually accumulated closer to the surface as a consequence of its arrival from land, and spreads deeper than the zero level, most likely due to mechanical mixing or diffusion [4–8]. At the drainage area of Ysyk-Köl, density of radioactive cesium precipitation achieves the level comparable with the global background (100 nCi/m^2) only on rarely occurring terraces.

Quite different situation occurs with $^{210}Pb_{atm}$, which is formed as a result of radioactive decay of atmospheric radon. Under the conditions of calm sedimentation, its activity decreases exponentially from the maximum value observed near the surface of the sediment to the depth, thus providing the possibility to calculate sedimentation rate. If neither ^{137}Cs nor ^{210}Pb are detected on the surface or, quite contrary, both isotopes are distributed uniformly into the depth of the sediment, this is an indication of its mixing.

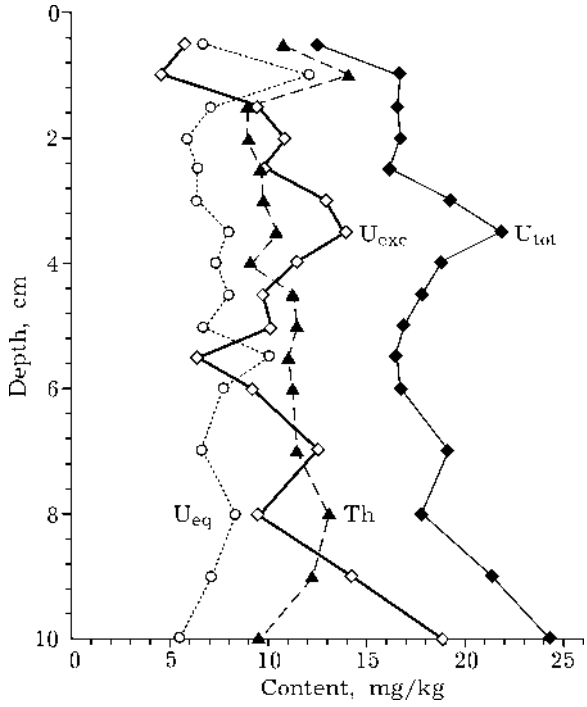

Fig. 7. Distribution of the components of total uranium (U_{tot}) over column IK_{01}–1 in the deep-water sediments: «equilibrium» (U_{eq}) and excessive over the equilibrium (U_{exc}).

Ideal possibilities for estimations of sedimentation rate are provided by the columns of deep-water sediments at the stations IK_{01}–1 (lake depth 336 m) and IK_{01}–3 (depth: 290 m) (see Fig. 6). Satisfactory estimations were obtained also for some littoral columns, for example IK_{01}–12, IK_{01}–22 in which there are no indices of distorted sedimentation.

With a constant rate of calm sedimentation, chronological estimations are obtained using the equation $t = -\lambda^{-1}\ln \Sigma A/\Sigma A_b$, where λ is the radioactive decay constant of ^{210}Pb; ΣA is the integral activity summed over the whole column; ΣA_b is the integral activity of the section subjacent to the layer to be dated [9–12]. The obtained dates lead to the following estimated rates of calm, undistorted sedimentation during the recent century, mm/y: 0.23 (IK_{01}–1), 0.39 (IK_{01}–3), 0.24 (IK_{98}–21bc), 0.42 (IK_{01}–22b), 0.21 (IK_{01}–12).

CONCLUSIONS

1. Procedures providing the possibility of simultaneous determination of radionuclides of uranium series (^{238}U, ^{226}Ra, ^{222}Rn, ^{210}Pb)

and isotopes arriving from the atmosphere ($^{210}Pb_{atm}$ and ^{137}Cs) in rocks and bottom sediments are developed.

2. For consistent revelation of anomalies of radioactivity, the value for radiogeochemical background (35–55 Bq/kg) was substantiated for the sediments on land, represented mainly by granitoid weathering products. Along with this, substantial distortions of radioactive equilibrium between uranium and radium over the background level were revealed; it was demonstrated that a widespread method of estimating the amount of uranium on the basis of radium gives results understated by 20 %.

3. Radioactive anomalies of three types exceeding the background level by one-two orders of magnitude were revealed:

– Natural uranium-radium anomalies. These include uranium-bearing coal of Jurassic age and waste material remaining after coal mining. For this type of anomalies, it was established that the ratio of uranium to radium is close to the equilibrium value. Attitude of coal bedding is strongly distorted; the beds are almost vertical; there is no doubt that their erosion during the geological time scale made substantial contribution into unusually high uranium content of the lake water.

– Industry-related radium anomalies were discovered in spacious masses of fine-grained substances paled with a concrete wall. Large (4–6 times) excess of radium in comparison with uranium, with respect to the equilibrium values, characterizes this type of anomalies. The presence of high-temperature mineral mullite in the fine-grained substance proves that it is ash from which uranium has been extracted after burning coal. Its radioactivity is 10–100 times higher than the level permissible in construction (370 Bq/kg).

– Industry-related uranium anomalies. They occur in the cascade of sedimentation reservoirs connected with each other, which had been constructed in order to prevent pollution of the lake with industry-related products. A moderate (an order of magnitude above the background) enrichment with uranium was detected in the clayish sediments of the upper sedimentation reservoirs. Uranium content decreases from upper reservoirs to lower ones, while radium content remains constant at the background level. These are evidently young anomalies which are due to uranium leakage from the technological process. At present, sedimentation reservoirs play their protective role, but they can be destroyed in a catastrophic event.

4. Subexponential decrease in the activity of radionuclides arriving from the atmosphere (^{137}Cs and ^{210}Pb) with depth as a result of radioactive decay was discovered in deep-water clayish sediments of the kale Ysyk-Köl, which provides evidence of calm, undistorted sedimentation during the recent century. Judging from the decay of $^{210}Pb_{atm}$ activity, the rate of deep-water sedimentation was estimated to be 0.2–0.4 mm per year (littoral clayish sediments were accumulated at the same rate in places where they were not mixed by the currents flowing into the lake). It was established on the basis of uranium to radium ratios that in deep-water sediments the fraction of uranium extracted from the lake water is 1.5–2 times larger than its fraction arrived with the suspension.

5. Radium anomalies were discovered in littoral lake sediments. In the places where sedimentation was distorted by currents, atmospheric radionuclides are buried under sand-clayish material brought from the bank. A layer enriched with radium was discovered in a number of littoral columns at a depth of 5 to 20 cm. The presence of mullite in the sediment is the evidence of the penetration of radioactive ash into the lake (perhaps before the concrete wall had been constructed). Since migration of dissolved radium was nowhere discovered, small amount of buried ash can hardly pose hazard for the biogeosystem of the lake Ysyk-Kцl.

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