

Radioecological monitoring of transboundary rivers of the Central Asian Region

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This paper presents the results of radioecological investigation of Central Asian rivers. This work was done as part of the Navruz Project, a cooperative, transboundary river monitoring project involving rivers and institutions in Kazakhstan, Kyrgyzstan, Tajikistan, and Uzbekistan, and facilitated by Sandia National Laboratories in the United States. The study of waterborne radionuclides and metals concentrations in Central Asia is of particular interest because of the history of nuclear materials mining, fabrication, transport, and storage there, when it was part of the Soviet Union. This development left a legacy of radionuclides and metals contamination in some Central Asian regions, which poses a clear health hazard to populations who rely heavily upon surface water for agricultural irrigation and direct domestic consumption.

Introduction

The Navruz Project, described in this report, monitors basic water quality parameters, radionuclides, and metals in the Syrdarya and Amudarya rivers and their major tributaries. Navruz is a Central Asian term that refers to the spring season. It means “new beginning.”

The total stock of water in Central Asia is 136.5 km³ per year (87 km³/year in the Amudarya watershed and 49.3 km³/year in the Syrdarya). The transboundary nature of water resources demands a transboundary approach to their monitoring and management.

The Navruz Project addresses three main goals: (1) to help increase capabilities in Central Asian nations for sustainable water resources management; (2) to provide a scientific basis for supporting nuclear transparency and non-proliferation in the region, and (3) to help reduce the threat of conflict in Central Asia over water resources, proliferation concerns, or other factors.

The following organizations took part in the project: Kazakhstan Institute of Nuclear Physics, Kyrgyzstan Institute of Physics, Tajik Atomic Energy Agency, Uzbekistan Institute of Nuclear Physics, and Sandia National Laboratories in the United States.

Data obtained in the project are shared among all participating countries and the public through an Internet web site⁴ and are available for use in further studies and in regional transboundary water resource management efforts. All the data from the investigation – in provisional form – are located on the website, along with various reports on and analysis of the data, maps, and other materials.

Experimental

The Central Asia Cooperative Monitoring Experiment (CME) began with the development of a sampling and analysis plan.¹ This plan was referred to as an instruction manual for participants on sample collection activities, sample equipment operations and procedures, sample handling, laboratory analysis, detection limits, sample preparation, and instrument procedures.

Participating countries – the Central Asian states of Kazakhstan, Uzbekistan, Kyrgyzstan, and Tajikistan – collected water, bottom sediment, vegetation, and soil samples from a total of 60 stations along the rivers (15 stations for every country). Sampling occurred in the fall and the spring of each sampling year. These samples were sent to laboratories in Kazakhstan and Uzbekistan for analysis. Control samples from two stations in each country were also sent to Sandia National Laboratories for analysis.

The sampling stations are shown in Fig. 1, and are described in greater detail on the website.⁴

For the Syrdarya and its tributaries, there are fifteen sampling stations in Kyrgyzstan, three stations in Tajikistan, nine stations in Uzbekistan, and fifteen stations in Kazakhstan for a total of forty-two stations. On the Amudarya river system, there are twelve sampling stations in Tajikistan and six stations in Uzbekistan for a total of eighteen stations.

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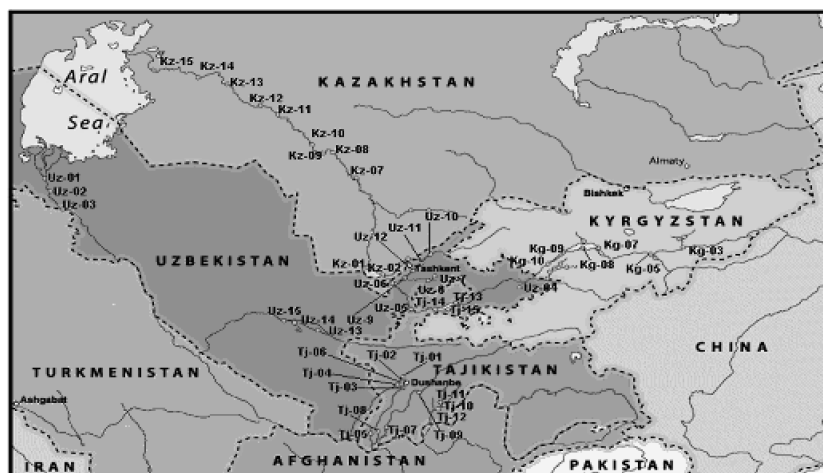


Fig. 1. Sampling stations

Table 1. Number of samples

Country	Total sampling points	Number of samples					Total
		Water	Bottom sediment	Soil	Vegetation		
Kazakhstan	15	45	45	43	27	160	
Kyrgyzstan	15	45	5	45	No	95	
Tajikistan	13	28	26	27	5	86	
Uzbekistan	12	42	42	42	42	168	
Total:	60	160	118	157	74	509	

At the same time as sampling was done, a series of four photographs was taken at each sampling location looking upstream, downstream, to the left, and to the right. GPS was used to determine the latitude and longitude of each sampling location. A sensor and data logging system manufactured by Hydrolab, Inc., was used during sampling to measure the following field parameters: water temperature, specific conductivity, oxidation/reduction potential, pH, dissolved oxygen, salinity, and total dissolved solids. The total number of samples taken during all sampling periods (Fall 2000, Spring 2001, Fall 2001) is shown in Table 1. In all, 509 samples were taken, 160 water, 118 bottom sediments, 157 soil, and 74 vegetation.

An automatic alpha- beta-counting system was used to measure total radioactivity of samples. This system was calibrated with the help of standard sources of known activity. The detection limit of this system is 10^{-2} Bq. The measurement error for beta-activity is 5–10% and for alpha-activity, 10–15%. All samples were measured no fewer than three times each. The determination of radionuclides was made by gamma-spectrometry. The measurements were conducted using a type GGPC 25-185-R detector with the following characteristics: external diameter 15.7 mm, efficiency

26.6%, measured energy resolution 0.66 keV at 122 keV and 1.75 keV at 1332 keV.

Elemental composition of the samples, was determined using instrumental neutron activation analysis (INAA). Samples and standards were sealed in bags made of pure polyethylene film and irradiated with neutrons. The neutron flux was $5 \cdot 10^{13}$ n·cm⁻²·s⁻¹ and irradiation time was 15 seconds. Gamma-spectra were measured using an HPGe detector and a computed gamma-spectrometry system 10 minutes after irradiation and again after 2 hours. After one week, the samples were repacked and irradiated at the same flux for 15 hours. Gamma-spectra were measured one week after the second irradiation. One month after irradiation, gamma spectra were measured again for determination of long-lived isotopes.

Standards were made by placing a micro-volume of a solution of chloride or nitrate of each element on a strip of ashless filter paper and drying it. At the same time, the comparator method was used with Zn chosen as the comparator. The laboratory reference material was homogenized human blood. Homogeneity was 3–7%

The analytical quality was tested using the standard procedures of research material IAEA-336 and AQCS cabbage material IAEA-359.

Table 2. Hydrolab field data collection log

Location	DO, mg/l		EC, $\mu\text{S}/\text{cm}$		Salinity, g/l		TDS, mg/l		pH		ORP, mV							
	Fall	Spring	Fall	Spring	Fall	Spring	Fall	Spring	Fall	Spring	Fall	Spring						
1	11.7	1.56	11.7	1941	3199	2041	1.04	1.73	1.09	1.24	2.05	1.31	8.22	7.85	7.08	321	255	
2	12.4	1.68	11.7	1799	3146	2932	0.96	1.70	1.03	1.15	2.01	1.24	8.19	7.85	7.23	329	245	
3	11.1	1.52	11.5	1790	5714	1781	0.95	1.46	0.95	1.15	1.74	1.14	8.15	7.75	7.09	342	267	
4	9.2	0.92	8.8	799	1559	1227	0.41	0.83	0.65	0.51	0.99	0.79	8.05	7.78	7.20	374	195	
5	10.5	0.91	10.4	222	1747	1766	1.19	0.93	0.94	1.42	1.12	1.13	7.81	7.45	7.46	688	278	
6	9.90	0.86	8.6	1798	1934	1770	0.96	1.03	0.94	1.15	1.24	1.13	8.05	7.89	7.25	344	220	
7	9.8	0.62	9.3	189	131	533	0.09	0.05	0.11	0.12	0.08	0.15	8.00	8.10	7.52	365	254	
8	9.5	0.81	9.1	218	160	333	0.10	0.07	0.16	0.14	0.10	0.21	7.97	8.53	7.66	363	282	
9	9.3	0.34	9.0	687	701	738	0.35	0.36	0.38	0.44	0.45	0.47	8.31	7.38	7.22	359	329	
10	9.4	1.70	9.0	220	217	498	0.10	0.1	0.25	0.14	0.14	0.30	7.91	7.76	7.635	416	260	
11	11.3	2.03	11.1	385	304	345	0.19	0.15	0.17	0.29	0.19	0.22	8.54	8.14	8.00	355	235	
12	12.9	0.75	10.6	492	455	500	0.25	0.23	0.25	0.32	0.29	0.32	8.79	8.23	7.70	336	282	
13	-	1.14	10.1	-	282	455	-	0.14	0.23	-	0.18	0.29	-	1.93	7.36	-	278	218
14	-	1.23	9.9	-	725	745	-	0.37	0.38	-	0.46	0.48	-	7.96	7.47	-	278	223
15	-	1.14	6.0	-	3198	2990	-	1.73	1.61	-	2.05	1.91	-	6.65	6.31	-	298	272

DO: Dissolved oxygen.
 EC: Electrical conductivity.
 ORP: Oxidation reduction potential.

Table 3. Flow meter discharge

Collection point	Fall 2000		Spring 2001		Fall 2001	
	Velocity, m/s	Discharge, m ³ /s	Velocity, m/s	Discharge, m ³ /s	Velocity, m/s	Discharge, m ³ /s
1	2	3	4	5	6	7
UZ-01 – Amud.	0.15	4.29	0.06	3.06	0.13	3.37
UZ-02 – Amud.	0.26	79.0	0.47	119	0.42	174
UZ-03 – Amud.	0.25	89.0	0.44	116	0.43	175
UZ-04 – Syrd.	0.32	248.0	0.55	195	0.58	186
UZ-05 – Syrd.	0.13	51.3	0.05	19.5	0.20	86.9
UZ-06 – Syrd.	0.27	114.0	0.27	286	0.60	334
UZ-07 – Achang.	0.63	3.21	1.8	0.87	0.59	0.57
UZ-08 – Achang.	0.68	30.2	0.43	17.5	0.38	12.2
UZ-09 – Achang.	0.20	2.66	1.03	10.4	0.33	9.44
UZ-10 – Chirch.	1.06	113.0	1.42	21.4	1.68	170
UZ-11 – Chirch.	0.21	2.48	0.49	24.9	0.46	18.8
UZ-12 – Chirch.	0.03	0.33	0.43	22.9	0.44	18.2
UZ-13 – Zaravch	–	–	1.87	23.5	0.57	17.04
UZ-14 – Zaravch	–	–	0.87	18.0	0.48	20.5
UZ-15 – Zaravch	–	–	0.18	10.4	0.14	2.06

Table 4. INAA results for elements determined in RM-IAEA-336 and AQCSM-IAEA-359 (in mg·kg⁻¹)

Element	RM-336		AQCSM-359	
	This analysis	Certified value	This analysis	Certified value
Br	14 ± 0.85	13.04 ± 0.52	7.0 ± 0.058	6.7 ± 0.26
Co	0.22 ± 0.018	0.29 ± 0.0014	0.1 ± 0.001	0.12 ± 0.008
Cr	0.95 ± 0.1	1.07 ± 0.067	1.1 ± 0.08	1.3 ± 0.05
Fe	461 ± 35	430 ± 10	174 ± 10	154.7 ± 3.4
Na	382 ± 31	323 ± 2.4	594 ± 16	577 ± 14.5
Sc	0.176 ± 0.012	0.17 ± 0.01	0.028 ± 0.0012	0.025 ± 0.0014
Zn	28.6 ± 1.4	30.2 ± 0.72	37 ± 0.72	34.9 ± 0.46

Table 5. Element concentrations in liquid phase of river waters (in µg·l⁻¹)

Element	Average	<i>m</i>	Minimum	Maximum	World average in surface waters
Ag	2.2		<0.1 (most)	26	0.13–0.20
Au	0.80		<0.001 (most)	16	0.0020–0.060
Ba	134	10	15	310	20–54
Br	42	6.0	<1.0 (some)	170	21
Ca	123000	15400	5000	180000	15000
Co	1.90	0.50	0.36	350	0.30–0.90
Cr	8.3	5.7	<0.360	2800	0.18–1.0
Cu	1000	95	70	660	7.0
Cs	0.85	0.45	<0.03 (many)	13	0.03–0.2
Fe	170	23	18	800	410–870
Hg	0.11	0.014	<0.01 (many)	0.40	0.08
La	3.2	3.45	1.10	0.12	0.2
Mg	37000	4600	5800	120000	4100
Mn	45	6.7	3.2	220	10–12
Ni	77		<30 (most)	4.1	2.5–10
Rb	2.1		<1.0 (most)	4.1	1.5–2.0
Sc	0.074	0.017	0.012	0.87	0.010
Se	1.4	0.16	<0.1 (many)	5.8	3.2
Zn	41	15	6.0	970	10–20
U	8.7	1.1	0.40	41	0.5–1.0

Table 6. INAA results for bottom sediments (in mg·kg⁻¹)

Element	Average	<i>m</i>	Minimum	Maximum
Ag			<0.1 (most)	6.5
Au	0.0057		<0.001 (many)	0.10
Ba	296	22	<100 (some)	900
Ca	61000	3800	6800	196000
Co	9.3	0.59	0.3	40
Cr	35	3.5	<1.0 (some)	200
Cs	3.6	0.18	<0.10 (some)	11
Fe	18000	650	3000	41000
La	32	1.6	7.9	160
Mn	510	28	20	234
Rb	60	3.4	20	234
Sb	1.6	0.23	0.23	13
Sc	7.7	0.25	1.5	17
Se	0.34		<0.10 (most)	11
U	1.9	0.26	<0.10 (many)	10

Table 7. INAA results for plants (in mg·kg⁻¹)

Element	Average	<i>m</i>	Minimum	Maximum	World average in surface waters
Ag	0.43		0.10 (most)	15	0.080–0.10
Au	0.015		0.010 (most)	0.92	
Ba	240	15	100	1240	500
Ca	57000	2500	3500	170000	13700–15000
Co	9.0	0.33	1.1	26	8.0
Cr	35	2.7	6.2	240	100–200
Cs	3.8	0.17	0.68	14	5.0–6.0
Fe	18000	490	3000	37000	38000–40000
K	21000	820	1000	121000	14000–15000
La	35	1.8	7.6	230	30–40
Mn	550	13	590	1300	850
Na	12000	540	1800	80000	6000–6300
Rb	56	2.4	10	170	100
Sb	1.6	0.23	0.10 (some)	11	2.0–10.0
Sc	8.3	0.18	20	14	7.0
Th	10	3.2	20	500	5.0–6.0
U	1.6	0.14	<10 (some)	7.3	1.0

Table 8. INAA results for soil (in mg·kg⁻¹)

Element	Average	<i>m</i>	Minimum	Maximum	World average in soil
Ag	0.43		0.10 (most)	15	0.080–0.10
Au	0.015		0.010 (most)	0.92	
Ba	240	15	100	1240	500
Ca	57000	2500	3500	170000	13700–15000
Co	9.0	0.33	1.1	26	8.0
Cr	35	2.7	6.2	240	100–200
Cs	3.8	0.17	0.68	14	5.0–6.0
Fe	18000	490	3000	37000	38000–40000
K	21000	820	1000	121000	14000–15000
La	35	1.8	7.6	230	30–40
Mn	550	13	290	1300	850
Na	12000	540	1800	80000	6000–6300
Rb	56	2.4	10	170	100
Sb	1.6	0.23	0.10 (some)	11	2.0–10.0
Sc	5.3	0.18	20	14	7.0
Th	10	3.2	20	500	5.0–6.0
U	1.6	0.14	<10 (some)	7.3	1.0

Results and discussion

Chemical analysis data for the samples are shown in Table 2. As shown in the table, the pH of water at all sampling points is approximately 8.0. This value is within permissible levels (6.0–9.0). The specific conductivity (capacity of water to conduct electricity) ranges from 200 to 3000 pS/cm. This change is related to increasing water salinity, especially in the springtime. Oxidation-reduction potential, an important factor for chemical reactions involving metals in water, is between 300–400 mV for all samples.

Dissolved oxygen is an important factor for many biological and chemical reactions in surface water.

This parameter varied between 0.8–12 mg/l. Increasing dissolved oxygen is related to improvements in water quality.

A flow meter was used to measure velocity of flow and discharge of water for rivers of Uzbekistan. The dates and results of these measurements are in Table 3.

As shown in Table 3, the discharge of water in the spring period is generally higher than in the fall. For the lower Amudarya, however, the discharge of water decreases in the spring period. There is a very small water discharge from the Zaravshan River especially in the fall. Figure 2 shows the salinity and β - α activity of water samples collected in the Syrdarya during the three sampling expeditions (Fall 2000, Spring 2001, and Fall 2001).

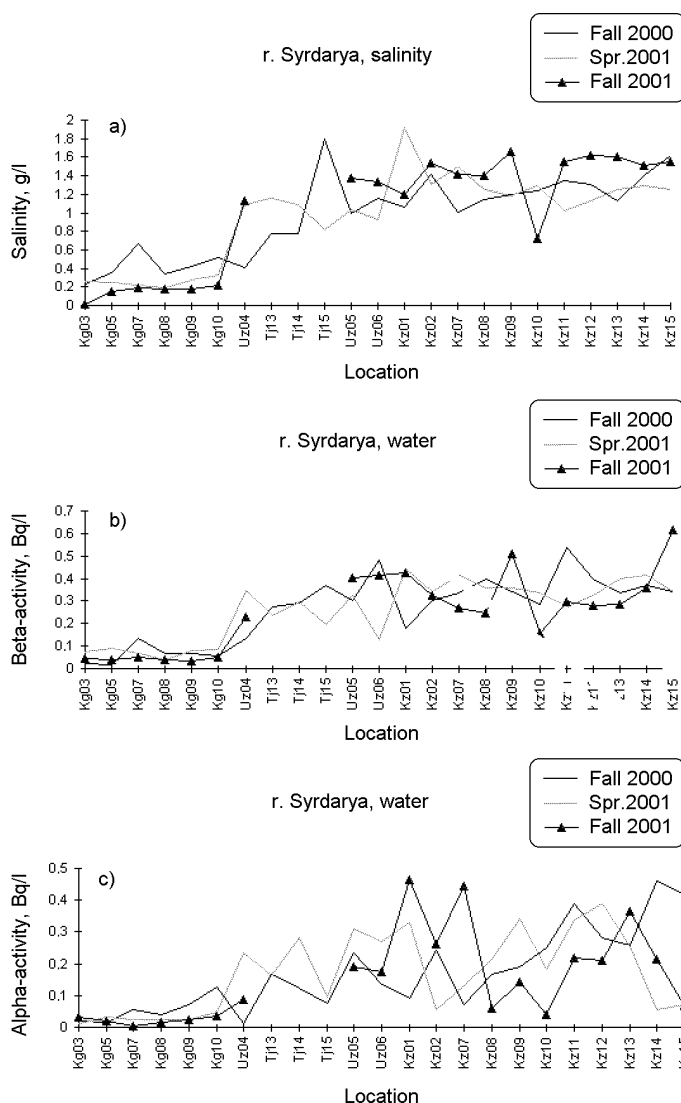


Fig. 2. Salinity (a) and total β - (b) α -activity (c) of water in the Syrdarya

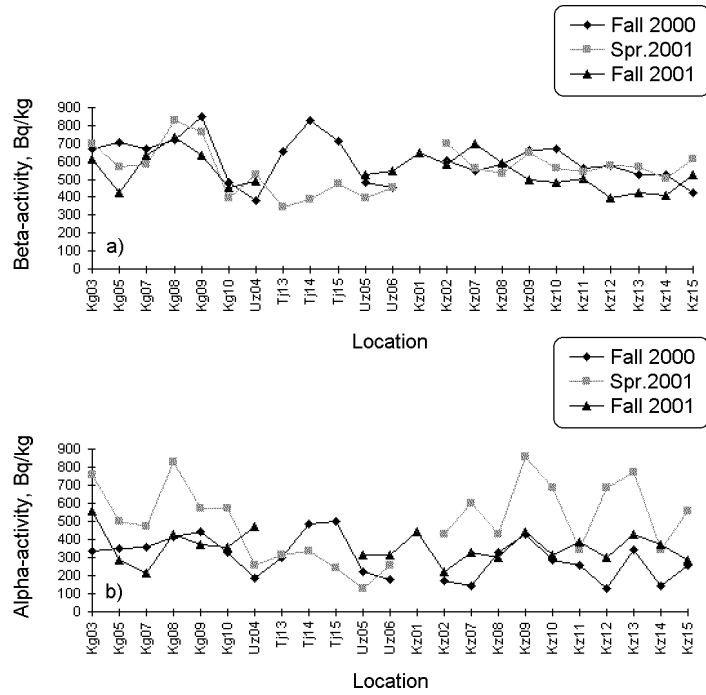


Fig. 3. Total β - (a) and α -activity (b) of soil along the Syrdarya

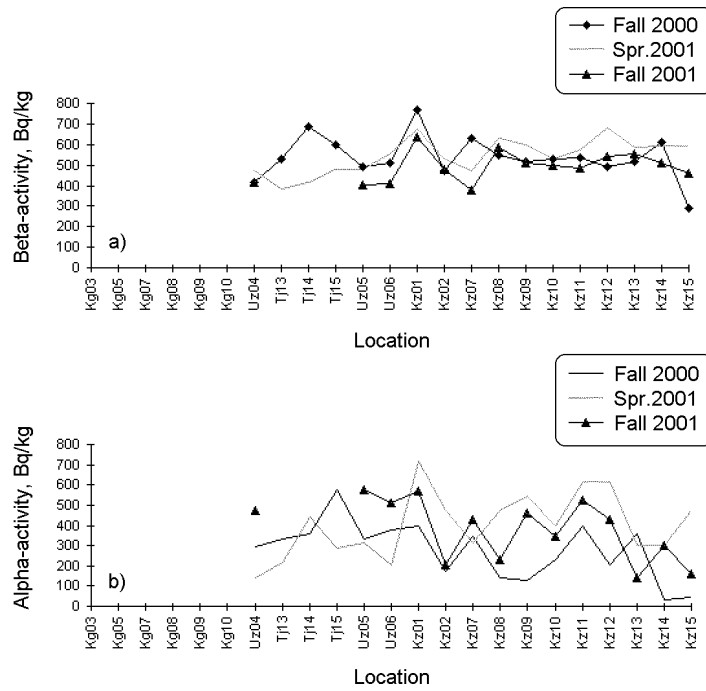


Fig. 4. Total β - (a) α -activity (b) of bottom in river Syrdarya

The salinity and activity of water increases from the upper to the lower river. The considerable increase of total β - α activity and salinity is seen for points UZ-04 (Karadarya), UZ-06 (Chinaz) in Uzbekistan, TJ-13 (Kayrakkum reservoir), TJ-14 (Khodzhand), and TJ-15

(Isfara) in Tajikistan, and for points KZ-01, KZ-02 (Chardarya reservoir), KZ-07 (Turkestan), KZ-09, KZ-10 (Shyily), and KZ-11 (Kyzylorda) in Kazakhstan.

The total β - α activity of soil is shown in Fig. 3. The activity of soil varies between 200 and 850 Bq/kg for the

different regions. The activity of soil increases for the lower Syrdarya – KZ-09, KZ-10 (Shyily) – in Kazakhstan for the fall sampling period.

The total β - α activities of bottom sediments in the Syrdarya are presented in Fig. 4. The bottom sediments total activity increases at points TJ-14 (Khudzhand), KZ-01 (Chardarya), KZ-09, and KZ-10 (Shyily).

The total β - α activity and salinity of Amudarya river water is shown in Fig. 5. Salinity and total β -activity increase in the lower Amudarya River. The same situation is seen for total α -activity.

The α -activities of bottom sediments and soil samples from the Amudarya River are shown in Fig. 6. Total activity of these samples increase the TJ-01, TJ-02 (Varzob river) and TJ-03 (Kafimigan river) in Tajikistan. The migration of activity to the lower Amudarya was not observed.

The quantity of the radionuclides ^{226}Ra , ^{40}K , ^{232}Th , and ^{238}U in all samples was investigated. Of particular note are the sampling points at Bekabad and Yangiabad in Uzbekistan and at Chardara Reservoir, Shyily, and Dzhusaly in Kazakhstan. At these points, the quantities of radionuclides are higher than in other points. The amount of radionuclides ranges from 90 to 920 Bq/kg

for ^{40}K , from 30 to 150 Bq/kg for ^{226}Ra , from 7 to 70 Bq/kg for ^{232}Th , and from 5 to 180 Bq/kg for ^{238}U . The amount of uranium and thorium decay products is highest in stations near uranium mines.

Activation analysis data are shown in Table 4.

Results of elemental analyses are shown in Tables 5 to 8. Widely differing natural and geochemical conditions, as well as presence of a number of specific sources of environmental contamination, are the reason for the very high variability of the elemental results.³ It should be noted that in most cases elevated average concentrations are connected with extremely high concentrations of some elements in samples taken in vicinity of sources of contamination.

Significantly elevated levels of Ag, Au, Cr, Cu, Mg, Ni, and U, in comparison with world-average data, were found in the liquid phase of river water. Most of these elements are produced in the region. Slightly elevated levels were found for Ba, Co, Cs, Sc, and Zn. Levels close to world averages were found for Hg, La, and Rb. Lower than average levels were found for Fe and Se.

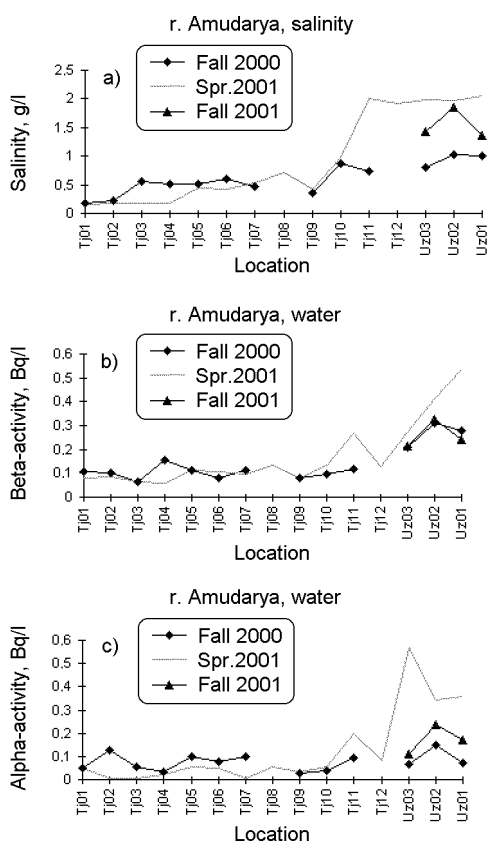


Fig. 5. The salinity (a) and total β - (b) α -activity (c) of water in river Amudarya

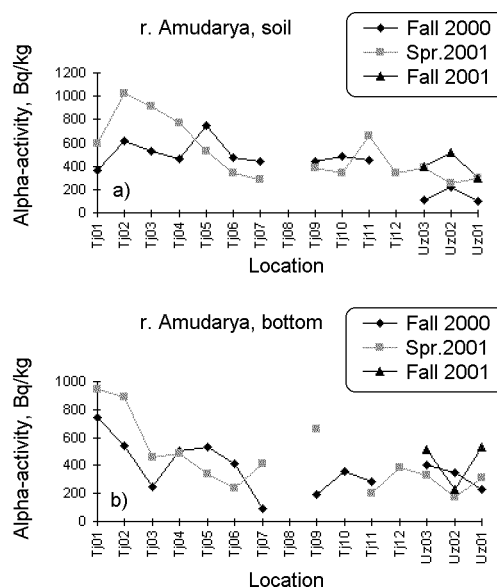


Fig. 6. Dynamics of alpha-activity of soil (a) and bottom sediments (b) for river Amudarya

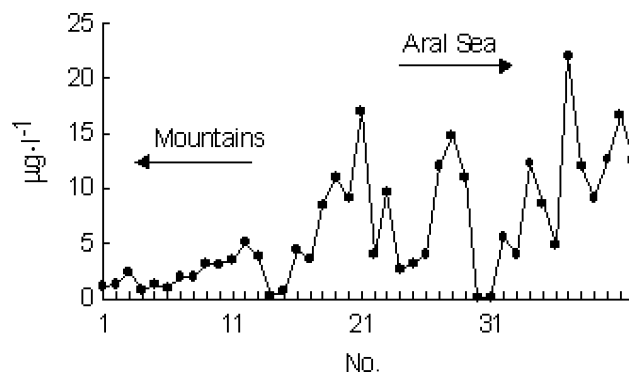


Fig. 7. Concentration of uranium in liquid phase of Syrdarya together with inflows

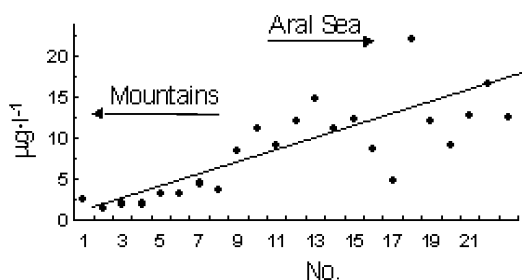


Fig. 8. Concentration of uranium in the liquid phase of the Syrdarya in samples taken on the river only

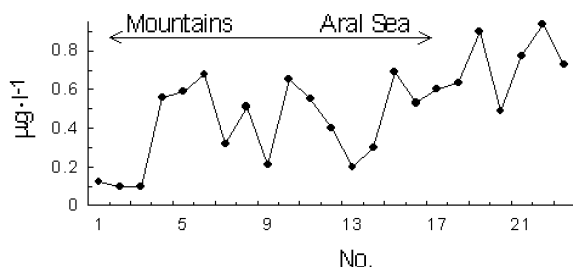


Fig. 9. Concentration of antimony in the liquid phase of the Syrdarya in samples taken on the river only

Bottom sediments and plants also contain higher levels of elements in comparison with existing, but not averaged, data.

Soils were found to have significantly elevated levels of Ag and Ca, in comparison with world-average data. Slightly elevated levels were found for K, Na, Th, and U. Levels close to world averages were found for Co, Cs, La, Mn, Sb, and Sc. Lower than average levels were found for Ba, Cr, Fe, and Rb.

In all types of samples the maximum measured levels are extremely high and are connected with industrial activity.

Figure 7 illustrates the extremely wide range of geochemical and environmental conditions among the sampling points. Figure 7 shows the concentration of uranium in the liquid phase of water in the Syrdarya

together with number of its inflows. Peaks are connected with decommissioned and working mines, mill sites, tailings sites, and factories. The lowest levels are in samples from mountain rivers which are not near uranium production activities. Uranium concentration tends to increase from the mountains to Aral Sea. This trend is much clearer in the data for samples taken on the river without inflows (Fig. 8). The trends of contamination of the Syrdarya by other contaminants is similar. For example, data for antimony are given in Fig. .

Several correlations and peculiarities appear in the sampling data, but limitations of space in this paper allows discussing only a few illustrations.

Conclusions

The distribution and dynamics of β - and α -activity were studied in both the Syrdarya and Amudarya. This investigation shows that Central Asian rivers deliver nearly 10^{13} Bq of total radioactivity to the Aral Sea. Uranium mines influence the process of formation of natural radioactivity in these rivers.

Parallel analysis of collected samples gives extremely important information. It allows determination of elements of interest, such as U and Th along with other toxic elements.

It should be emphasized that a study of such scale has never before been carried out in the region. The results give a much clearer picture of regional contamination than heretofore, because rivers collect information on the status of the environment in a more averaged way than analysis of usual environmental samples taken according to a grid. This allows significant reduction in the number of samples necessary for such large area.

Unfortunately, it is not possible to present all the information obtained in this paper. However, additional data treatment will doubtless lead to important new conclusions.

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