Journal of Engineering Physics and Thermophysics, Vol. 70, No. 1, 1997

FORMATION AND DYNAMICS OF PROPAGATION OF RADIOACTIVE CONTAMINATION IN RIVERS OF BELARUS AFTER THE CHERNOBYL NUCLEAR ACCIDENT

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UDC 547.577.39

The formation of radioactive contamination in rivers in Belarus caused by the Chernobyl nuclear accident is analyzed, the peculiarities of monitoring in the experimental Iput catchment are described, a conceptual model of radionuclide migration in the river system is suggested, calculations are carried out, and experimental data and calculation results on transfer of radioactive contamination of terrestrial surface water are analyzed.

The Chernobyl nuclear accident has resulted in radioactive contamination of water catchments, rivers, and reservoirs. Most of the radioactive fallout entered the catchment areas of the rivers Pripyat, Dnieper, Desna, and their tributaries. Those areas have become and will for a long time be potential sources of radionuclide flows into the Dnieper–Sozh system, and surface water is the main radionuclide transport system and the most ecologically vulnerable secondary source of contamination of ecosystems. This is confirmed by results of radiological monitoring performed in Russia, Ukraine, and Belarus [1-5].

In monitoring of water systems in Belarus, the greatest attention is paid to the conditions of the Pripyat and Dnieper, which flow in areas close to the Chernobyl Nuclear Plant, as well as of the Sozh, Iput, Besed, and tributaries of the Dnieper, which flow in areas that are at a great distance from the accident site and whose catchments are contaminated largely by cesium-137 at levels of from 37 to 1480 kBq/m^2 and above. These rivers present a potential ecological danger for areas that are contaminated at a lower level and whose condition could be much worse as a result of transboundary transfer of radionuclides by water.

Comparison of the data on the annual flows of the Dnieper and Sozh has shown that the largest amount of cesium-137 that is brought to the Kiev Reservoir by the Dnieper is carried by water of the Sozh and its tributary Iput (Fig. 1). For example, the total amounts of cesium-137 carried by the Dnieper at the city of Rechitsa (upstream from the confluence with the Sozh) and by the Sozh (at Gomel) were 2960 GBq and 10,360 GBq, respectively. Thus, the flows of the Sozh and its tributary Iput were responsible for most (about 80%) of the cesium-137 brought to the Kiev Reservoir.

Observations that were carried out at the rivers of the Dnieper–Sozh and Pripyat basins showed that from 1987 to 1993 the annual flow of dissolved cesium-137 had markedly decreased (Fig. 1). For example, the amount of cesium in the Pripyat decreased 8 times, in the Dnieper, 3.5 times, in the Sozh, 10.5 times, in the Iput, 13.5 times, and in the Besed, 6.3 times. However, the preaccident level of radionuclides was considerably exceeded in all rivers and reservoirs studied. In the upstream Dnieper basin, almost the entire radiocontamination flow is formed in its water catchment areas, while in the Pripyat, the main sources of strontium-90 are water-covered areas in the region of the CNP, and water catchments in the upstream basin are responsible for cesium-137 [2, 6].

The contents of suspended particles that appear in streams as a result of erosion washout of fine soil particles from the catchment area and of processes occurring in the bed have an important effect on transport of

State Committee for Hydrometeorology of Belarus, Minsk; Institute of Radioecological Problems, Minsk, Belarus. Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 70, No. 1, pp. 73-80, January-February, 1997. Original article submitted May 16, 1996.

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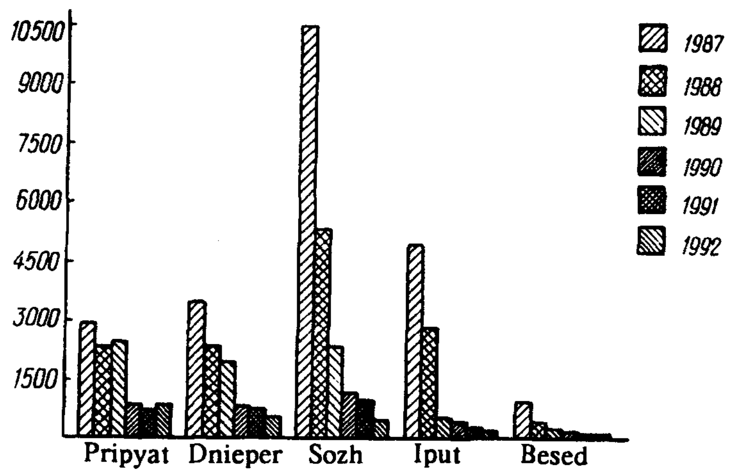

Fig. 1. Dynamics of radionuclide drift by rivers in Belarus (vertical axis, GBq/year).

radionuclides, especially cesium-137. The percentage of cesium-137 in the solid flow can vary from 20 to 80%, depending on erosion of the catchment, turbidity conditions, and the transport capacity of the stream [2, 6, 7].

In 1986-1991 the relative contribution of the flow of radionuclides associated with suspended particles to their total annual amount carried by the rivers remained at a level of 30–40%. The recent trend in the characteristics of the cesium-137 flow indicates that in spite of a decrease in the absolute amounts of the washed-off soluble radionuclide from the catchment surfaces, the erosion characteristics in formation of the surface flow have changed insignificantly. This, in turn, determined the growth of the relative contribution of the flow of this radionuclide on suspended particles to its total water flow [2, 6, 8].

Rivers are not only transport arteries of radionuclide contamination, but also intense accumulators of radionuclides in bottom sediments along river channels, especially in stagnation zones near dams in reservoirs. Along the river channels radionuclides are temporarily buried as a result of motion of sediment ridges, and in reservoirs they are buried as a result of sedimentation of fine silt, which results in mobile local sites of higher radionuclide concentrations.

In order to find the trends in radioactive contamination of Belarusian rivers, after generalization of monitoring data and natural observations, the Iput catchment was chosen as experimental. The Iput river is in the Sozh-Dnieper basin and flows over the Belarus-Bryansk "cesium spot" with contamination levels of 37 to 2200 kBq/m^2. Observations of the radionuclide contents in the water were carried out at two sites: at the village of Vydevo and at the city of Dobrush, Gomel Region.

Choice of control sites and datum plots for observations of the radionuclide contents in the Iput was based on the following criteria:

a) the datum plot and the Vylevo site should characterize a catchment with rather high cesium-137 soil-contamination levels, and in this area the average levels are 2200 kBq/m^2;

b) the Dobrush site is terminal in the Iput, in which integral characteristic of the entire water catchment are determined, and the cesium-137 contamination level is 185 kBq/m^2 in this area.

Since 1991 at the control sites of the Iput monthly observations of radionuclide contents in surface water have been carried out. Simultaneously with water sampling, the water flow rate has been measured and the water level observed.

In the water samples, the contents of cesium-137 and strontium-90 were determined in the dissolved state and on suspended particles (Table 1).

In 1991-1993, once a quarter, bottom sediments were sampled in these sites. The samples were analyzed to determine the contents of cesium-137, strontium-90, and plutonium-238, 239, and 240 (Table 2). Samples of bottom sediments were analyzed to determine the contents of radiocesium with an ADCAM semiconductor gamma spectrometer (ORTEC) with a measurement error within 30%. Strontium-90 and plutonium-238, 239, and 240

TABLE 1. Results of Analysis of Surface Water Samples to Determine Cesium-137 and Strontium-90 in Dissolved State and Sorbed on Suspended Particles (observation period is May-October, 1992)

Site	Sampling date	Average monthly flow rate, m^3/sec	Content, Bq/m^3			
			cesium-137		strontium-90	
			solution	suspension	solution	suspension
Dobrush	25.05	49.9	110	470	40	20
Dobrush	26.06	20.9	280	330	20	40
Dobrush	30.07	12.9	330	300	20	10
Dobrush	26.08	9.5	770	390	30	30
Dobrush	15.09	9.5	340	220	20	10
Dobrush	14.10	15.6	580		50	
Vylevo	27.05	52.0	460	430	90	30
Vylevo	27.06	23.4	320	610	10	50
Vylevo	29.07	15.5	330	280	30	10
Vylevo	26.08	11.2	790	450	40	40

TABLE 2. Specific Activity of Radionuclides in Bottom Sediments in Iput Channel Between Vylevo and Dobrush (Oct. 15, 1992)

Sampling site	Check point	Radionuclide contents, Bq/kg		
		cesium-137	strontium-90	plutonium-238–240
Dobrush (in front of dam)	p. 1	24,100	18.77	0.78
	p. 2	16,600	5.24	0.59
	p. 3	24,100	15.26	0.77
	p. 4	18,400	7.17	0.91
	p. 5	83,600	14.43	
	p. 6	11,400	18.82	
	p. 7	32,300	8.14	0.34
Vylevo	p. 1	20,300	0.01	0.29
	p. 2	4310	6.51	0.1
	p. 3	2520	0.01	0.2
	p. 4	8080	15.26	0.23
	p. 5	7610	9.62	0.64
	p. 6	5050	0.01	

TABLE 3. Results of Granulometric Analysis of Bottom Sediments and Activity of Cesium-134 and 137 in Individual Fractions

Point No.	Fraction, mm	Content, %	Radionuclide activity, Bq/kg	
			cesium-134	cesium-137
2	> 1.0	7,8	68.5	1036.0
	1.0–0.5	10.4	51.4	673.4
	0.5–0.2	25.9	57.4	902.8
	< 0.2	55.9	104.7	1435.6
4	> 1.0	4.3	170.2	1957.8
	1.0–0.5	10.3	75.9	1139.6
	0.5–0.2	29.8	101.0	1280.0
	< 0.2	55.6	185.7	2856.4
6	> 1.0	–	–	–
	1.0–0.5	1.9	178.0	2471.6
	0.5–0.2	5.5	187.6	2671.4
	< 0.2	92.6	595.7	9324.0

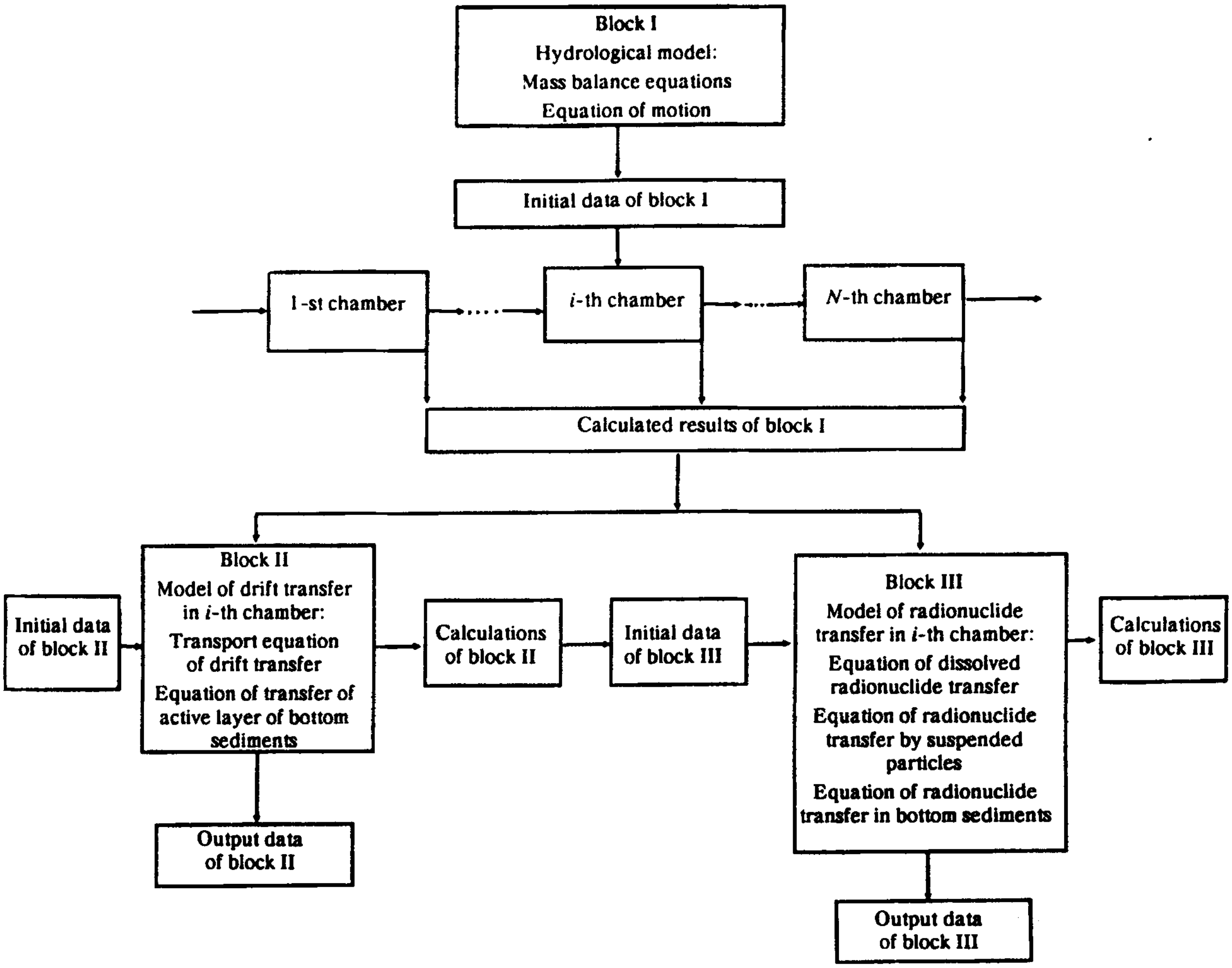

Fig. 2. Flow chart for calculation of radionuclide transfer in river system.

were determined by the standard radiochemical procedures approved by the State Committee for Hydrometeorology. The measurement error for strontium-90 was about 30%.

In 1992 a study of the granulometric composition of the active layer of bottom sediments was carried out in the terminal site of the Iput, and the activity of cesium-134 and 137 was determined for individual fractions of the sediments. The granulometric composition was analyzed with sieves. In the composition of the sediments, four groups of fractions were found: larger than 1 mm, 1–0.5 mm, 0.5–0.2 mm, and smaller than 0.2 mm. From every group, weighed quantities were chosen, in which the contents of cesium-134 and 137 were determined (Table 3).

On the basis of experimental data obtained in the Iput catchment, a mathematical model was developed for forecasts of the behavior of radionuclides in the river system. In construction of the migration model the entire river system was divided into N sequentially located chambers, each of which comprised two interacting layers, namely, an upper layer with volume V_1, which contained water and suspended particles, and a lower layer with volume V_2, which was an active layer of bottom sediments. These components of the river medium that contain radionuclides in soluble, exchange, and nonexchange forms are involved in mutual exchange: processes of sedimentation and stirring-up of suspended macroparticles, and radionuclide sorption-desorption associated with ion exchange processes in the suspension–water, and water–bottom-sediment systems. Moreover, the radionuclides are permanently brought to these volumes by water of the tributaries and groundwater and surface flows.

In the isolated sections of the river such processes are described by a system of ordinary differential equations of conservation of mass and motion of water averaged over the chamber volume, concentrations of transported drift, and amounts of radionuclides dissolved, adsorbed on suspensions, and entrained by drift. A flow

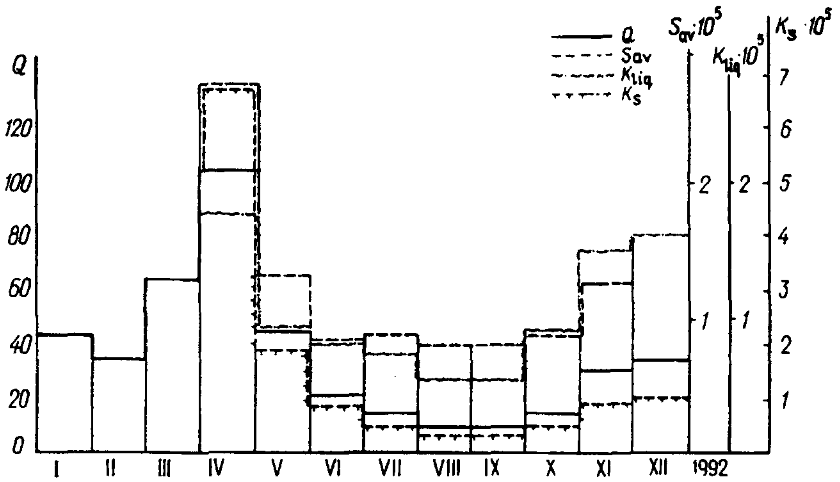

Fig. 3. Dynamics of changes in water flow rate (Q, m^3/sec), average turbidity (S_{av}, m^3/m^3), coefficients of solid and liquid flows of cesium-137 (K_{liq}, K_s) in Iput (Dobrush).

chart for calculation of the dynamic distribution of radionuclides in the chambers and components of the river medium is shown in Fig. 2.

For expeditious forecast, only the steady-state model can be considered with the following simplifying assumptions that allow calculations using minimal information:

1. The characteristic time of run-off from the water catchments is greatly in excess of the water turnover in the river.

2. Sorption equilibrium between the water and suspension and the water and bottom sediments sets in instantaneously for exchange forms of the radionuclides.

3. Equilibrium between stirring-up and sedimentation of suspended drift also takes a very short time.

4. The flow rate of bottom sediments in the river is stabilized rapidly.

5. The river channel is not deformed.

Information about measured flow rates, average water contamination, contamination of bottom sediments in individual fractions, and geometry of control sites and chambers is used as initial data. The measured granulometric composition of the active layer of bottom sediments can be used to calculate the transport capacity of the stream, its partial, average, bottom, and vertical turbidity, and the flow rate of entrained drift on the basis of the Makkaveev–Karaushev diffusion theory of bottom sediments and to find the vertical distribution of radionuclide concentrations in the control sites, for individual components in the i-th chamber and the river system as a whole, the distribution coefficients in the suspension–water and water–bottom-sediment systems, the total amount of radionuclides in the terminal site, and the coefficients of liquid and solid flows from the water catchment.

As was noted earlier, a characteristic feature of the experimental Iput catchment studied is that the river flows over the Belarus-Brest cesium spot. Moreover, sediments that form its bed are of medium size and large sands. Under these conditions, water contamination with strontium is insignificant.

Cesium-137 is transferred by river flow in dissolved form and sorbed on suspended particles and bottom sediments. Therefore, in this stage the main attention was given to cesium-137 transfer. Results of the study carried out in 1992 were initial data for analysis of experimental data and calculations (Tables 1 and 2).

Analysis of the experimental data (Table 1) shows that in the control sites the concentrations of cesium-137 in water are lower than its permissible level in drinking water established in Belarus ($C_{NPL}(Cs^{137}) = 18{,}500$ Bq/m^3). From the results of the analysis of bottom sediment samples (Table 2) taken over the channel of the experimental Iput catchment, it is possible to draw the following conclusions:

1. The maximum content of cesium-137 in the bottom sediments is concentrated in front of the dam which is located at Dobrush. In this section the specific activity of cesium-137 is 11.4–83.6 kBq/kg, which corresponds

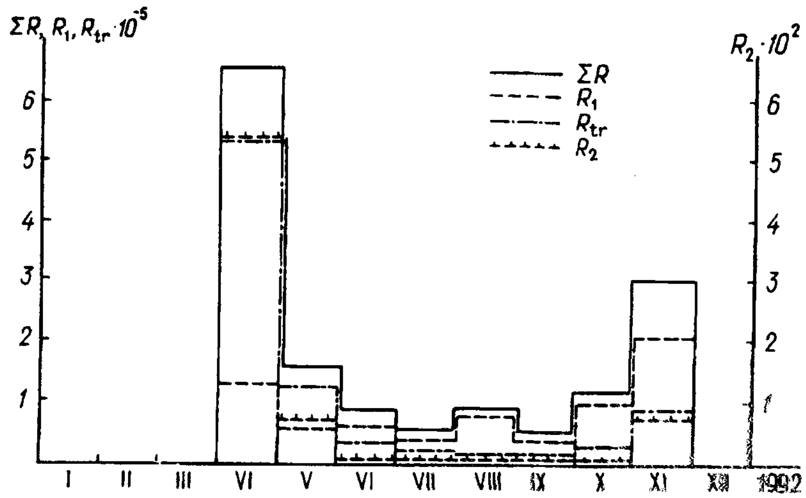

Fig. 4. Dynamics of cesium-137 amounts carried by water in dissolved form (R_1), with transported drift (R_{tr}) and entrained (R_2) by bottom sediments, and total flow (ΣR) (Bq/sec) at terminal site of Iput (Dobrush).

to the level of radioactive wastes. The maximum local content of cesium in the bottom sediments (83.6 kBq/kg) is 200,000 times higher than its preaccident specific activity.

In the section in front of the dam the content of cesium-137 can be 1330 kBq/m^3 in the case of complete stirring-up, which is almost two orders of magnitude higher than the National Permissible Level of cesium-137 in drinking water.

3. The measured specific activity of bottom sediments is almost an order of magnitude higher than the specific activity of the soil in the water catchment. This finding can probably be ascribed to the washout of radionuclides with the solid flow from the catchment, and consequently, cleaning of its soil, unsteady transfer of radioactive contamination by transported drifts, and sorption processes in the water—bottom-sediment system.

4. Radionuclide concentrations in the bottom sediments sampled along the Iput channel at the village of Vylevo are much lower than those concentrations at Dobrush, in front of the dam, in spite of the fact that Vylevo is within the cesium spot, where the soil contamination level is 2220 kBq/m^2.

5. The maximum radioisotope concentrations are found in stagnant areas of the Iput (backwater, a small lake, near the banks), where flow velocities are low. In bottom sediments sampled along the middle of the channel, the specific activity of radionuclides is lower compared with the stagnant areas, which indicates that they are washed from the bottom sediments by the mainstream flow.

6. Data given in Table 3 on the distribution of cesium-137 in the various fractions of the mechanical composition of the active layer of the bottom sediments confirm that this radionuclide is mainly transferred from the finely disperse fraction with a diameter smaller than 0.2 mm.

The study of radionuclide accumulation in the bottom sediments shows that at present the accumulation is largely caused by washing-off of radioactive contamination from the water catchment with suspended particles and subsequent transfer along the channel with the transported drift as well as by exchange processes in the water—bottom-sediment and suspension—water systems. This is confirmed by the data obtained in the Iput water catchment studied. The river section in front of the dam functions as a trap for contaminants and is a local site of dangerous concentration of radionuclides.

In Figs. 3-5 one can see experimental data and calculations of the behavior of the average water flow rates and turbidity, coefficients of liquid and solid flows of radioactive cesium, its concentration, the amount of dissolved cesium with transported drift, and the total flow of water contaminated with cesium-137 through the Dobrush terminal site in 1992.

Analysis of the integral characteristics of the Iput river for 1992 has shown that:

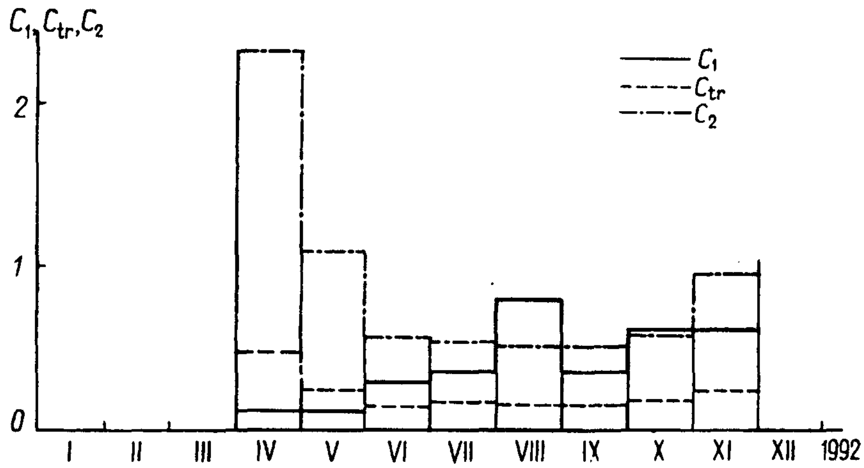

Fig. 5. Dynamics of changes in cesium concentration in dissolved form (C_1), in transported (C_{tr}), and entrained (C_2) (kBq/m^3) sediments at terminal site of Iput (Dobrush).

a) from April to November, 1992 the cesium-137 flow through the terminal site was 148 GBq in dissolved form, 207.2 GBq with transported drift, and the total amount of radionuclides in the flow through the site was 355.2 GBq;

b) the percentage of activity transported by suspended particles varied from 20 to 80% during the entire controlled period in 1992;

c) the moduli of liquid and solid flows of radionuclide varied in the ranges:

$$M_{\text{liq.f}} = (0.71-0.96)\cdot 10^{-3}\ 1/\text{m}, \quad M_{\text{s.f}} = (1.4-3.0)\cdot 10^{-3}\ 1/\text{m};$$

d) for 1992 the annual coefficients of liquid and solid flows for cesium-137 varied in the ranges:

$$K_{\text{liq}} = (0.5-3.0)\cdot 10^{-5}, \quad K_{\text{s}} = (3.3-6.7)\cdot 10^{-5}$$

and were higher in flood seasons.

Thus, analysis of theformation of river contamination in Belarus has shown that the greatest amount of radionuclides is from water catchments in the upstream Dnieper basin, namely, from the catchments of its tributaries Iput and Sozh. The annual amount of the dissolved radionuclides carried by Belarusian rivers substantially decreased from 1987 to 1992. In the controlled rivers transfer of radionuclides with suspended and entrained drift has a substantial effect on migration of the radioisotopes, and the contribution of this transfer form to the total radionuclide flow increases with time. In 1992 percentage of radiocesium carried with transported drift was 20–80% of the total flow. The importance of this conclusion consists in the fact that as a result of unsteadiness of the processes that occur in the river bed, suspended drift is deposited in regions with a slower flow velocity. This, in turn, gives rise to local mobile ecologically dangerous sites of radioactive contamination in bottom sediments of river flood plains, shallows, and reservoirs in front of dams. Therefore, regular monitoring of such sites, forecasting of their appearance, water-protection activities for deactivation of the largest accumulators of radionuclides, localizing the main sources of contamination, and control of migration processes in the soil–water and water–bottom-sediment systems are necessary.

NOTATION

Q, water flow rate; S_{av}, average water turbidity; K_{liq}, K_{s}, coefficients of liquid and solid flows of radionuclides; R_1, R_{tr}, R_2, R, flows of radionuclides in dissolved form, with transported and entrained drift, total flow; C_l, C_{tr}, C_2, concentration of dissolved cesium-137 in water, in transported and entrained drift: $M_{liq.f}$, $M_{s.f}$, moduli of liquid and solid flows.

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