MODEL OF MIGRATION OF RADIONUCLIDES IN A RIVER SYSTEM

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O. M. Zhukova,^a N. M. Shiryaeva,^b N. K. Myshkina,^b É. D. Shagalova,^a V. V. Denisova,^b and V. V. Skurat^b

The authors suggest a model of transfer of radionuclides in a river system, which relies on the principle of the chamber model, for the case of hydraulically stationary and chemically equilibrium conditions of interaction of radionuclides in the systems "water-suspensions" and "water-bottom sediments." The model is based on analytical solutions of a system of equations for different conditions of ingress of radioactive contaminants into the river system: in the inlet cross section; with fallout of radiactive aerosols on the water surface; with ingress of radionuclides with surface flow from a contaminated water catchment. The model is verified using the data of radiation monitoring carried out on an experimental water catchment of the Iput river.

The accident at the fourth unit of the Chernobyl Nuclear Power Plant resulted in contamination of the water-catchment areas of the Pripyat and Dnieper rivers and their tributaries. It is precisely these areas that have become the landscape sources of formation of radionuclide flows into the Dnieper–Sozh system on the territory of Belarus, and the terrestrial surface water has become the main radionuclide transport system and the most environmentally vulnerable secondary source of contamination of ecosystems. This is confirmed by results of radiation monitoring carried out in Russia, Ukraine, and Belarus [1–4].

As of now, we have a rather large volume of experimental data on the dynamics of transport and accumulation of radionuclides in elements of river systems which requires generalization and must be used for prediction using the methods of mathematical modeling. The data gathered can also be used rationally to verify the models and predict radioactive contamination of water objects in the case of emergency situations at nuclear power plants near the frontiers between Belarus and Russia, Lithuania, and Ukraine.

At present, specialists have different approaches to solution of problems of transfer of radionuclides by river flows. There are many mathematical models of different degrees of perfection for solving such problems, in particular, one- and two-dimensional models of river-channels, flows, sedimentation and transport of suspensions, and transfer of radioactive contamination. However, the capabilities of the majority of such models are still rather limited both because of the complex nature of the studied natural environment and the processes occurring in it and the deficiency and low quality of initial data on the hydromorphology of rivers, on solid flow and erosion-accumulation processes in water catchments, and on the mechanisms and kinetics of interaction of nuclides in the systems "water-soil of water catchment" and "water-suspensions-bottom sediments." Therefore, in formulation of specific environmental-hydrological problems one has to orient oneself, in the first approximation, to development and use of simplified models which do not require the provision of detailed information for their realization but allow for the main factors of transfer of radionuclides in the system "water catchment-river system" [5–7].

^aRepublic Center of Radiation Control and Monitoring of the Natural Environment, State Committee for Hydrometeorology of the Republic of Belarus, Minsk, Belarus; email: us206@fax.by.mecom.ru; ^bInstitute of Radioenvironmental Problems, National Academy of Sciences of Belarus, Minsk, Belarus; email: irep@sosny.bas-net.by. Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 74, No. 4, pp. 70–76, July–August, 2001. Original article submitted January 16, 2001.

In this work, a multichamber model based on the convective mechanism of transfer of radioactive contamination in soluble form, on suspensions, and by bottom suspended particles with account for granulometric composition of bottom sediments under hydraulically stationary and chemically equilibrium conditions is presented for investigation of the characteristics of migration of radionuclides in rivers. This model is noted for its simplicity and high speed, requires minimum information provision, and can be realized with the lowest numerical error. At the same time, this model allows one to reveal the main qualitative regularities of dispersion of radionuclides in river systems.

The model was developed due to the necessity of predicting radioactive contamination of the rivers in Belarus which flow through the most contaminated areas of the Republic. In this case, direct fallout of aerosols on the water surface, ingress of radionuclides from the contaminated area of water catchments with water flow and particles of soil during rains or thawing of snow, local supply of radionuclides by contaminated water of tributaries, etc. were the sources of radioactive contamination of water objects.

The multichamber model used for modeling transfer of radionuclides in the river system incorporates subdivision of the river system into k sequentially located chambers. Each chamber comprises two interacting layers, namely, the upper layer V_w , which contains water and suspended particles, and the lower layer of volume V_b , i.e., the active layer of bottom sediments. Mutual exchange occurs between these components of the river medium, which contain radionuclides in soluble, exchange, and nonexchange forms: processes of sedimentation-turbidity of suspended macroparticles and those of sorption-desorption of radionuclides which are related to ion-exchange processes in the systems "water–suspensions" and "water–bottom sediments." Radionuclides enter the considered volumes in fallout of radioactive aerosols on the water surface, with the water of tributaries, surface and ground flows, etc. [4].

The structural scheme of calculation has three main blocks which describe [4]:

- (1) flow-rate characteristics of the river system (hydrological block);
- (2) transfer of suspended and entrained sediments;
- (3) transfer of radionuclides in soluble form, on suspensions, and in bottom sediments.

In the general case, in the isolated sections of the river such processes are determined by a system of ordinary differential equations of conservation of mass of water, concentration of suspended and entrained sediments, and flow of radionuclides in soluble form, on suspensions, and entrained by sediments:

$$\frac{dV_{\mathrm{w}i}}{dt} = Q_{\mathrm{w.ent}i} - Q_{\mathrm{w}.i} + Q_{\mathrm{w.tr}i} + Q_{\mathrm{w.gr}i} + Q_{\mathrm{w.c}i}, \qquad (1)$$

$$\frac{dV_{wi}S_{mi}}{dt} = Q_{w.enti} S_{m.enti} - Q_{wi} S_{mi} + Q_{w.tri} S_{m.tri} + Q_{w.ci} S_{m.ci} , \qquad (2)$$

$$\frac{dV_{bi}}{dt} = Q_{b.enti} + F_i Q_{r.bi} - Q_{bi} , \qquad (3)$$

$$\frac{dV_{wi}C_{wi}}{dt} = Q_{w.enti} C_{w.enti} - Q_{wi} C_{wi} + Q_{w.tri} C_{w.tri} + Q_{w.gri} C_{w.gri} + Q_{w.ci} C_{w.ci} - a_{w.ri} (K_{d.ri}C_{wi} - C_{ri}) - a_{w.bi} (K_{d.bi}C_{wi} - C_{bi}) - \lambda_d V_{wi}C_{wi}, \qquad (4)$$

$$\frac{dV_{wi}S_{mi}C_{ri}}{dt} = Q_{w.enti} S_{m.enti}C_{r.enti} - Q_{wi} S_{mi}C_{ri} + Q_{w.tri} S_{m.tri}C_{r.tri} + Q_{w.ci} S_{m.ci} C_{r.ci} C_{r.ci} + Q_{w.$$

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$$+ a_{w,ri} (K_{d,ri}C_{wi} - C_{ri}) - F_i Q_{r,bi} C_{ri} - \lambda_d V_{wi} S_{mi} C_{ri} , \qquad (5)$$

$$\frac{dV_{bi}C_{bi}}{dt} = Q_{b.enti} C_{b.enti} - Q_{bi} C_{bi} + a_{w.bi} (K_{d.bi}C_{wi} - C_{bi}) + F_i Q_{r.bi} C_{ri} - \lambda_d V_{bi} C_{bi} .$$
(6)

In this case, we consider a quasistationary model of migration of radionuclides under chemically equilibrium conditions of their interaction in the systems "water-suspensions" and "water-bottom sediments" with the following assumptions:

(1) the flow rate of the water in control volumes of the river is constant;

(2) the characteristic time of run-off from the water catchment is greatly in excess of the time of turnover of the water in the river;

(3) sorption equilibrium between the water and the suspension and the water and the bottom sediments sets in instantaneously for exchange forms of the radionuclides;

- (4) equilibrium between turbidity and sedimentation takes a very short time;
- (5) the flow rate of the bottom sediments in control volumes of the river is constant;

(6) the river channel is not deformed.

The assumptions made allow one to pass from the system of differential equations to a system which comprises algebraic balance equations describing the hydrological block and the block of transfer of suspended and entrained sediments under hydraulically stationary conditions, and the differential equation of change in the concentration of radionuclides in the chambers with time in equilibrium interaction of the radionuclides in the systems "water–suspensions" and "water–bottom sediments." For these conditions, the transfer of radionuclides in the *i*th chamber is written as follows:

$$\frac{dV_iC_i}{dt} = C_{\text{w.ent}i}Q_{\text{w.ent}i} + C_{\text{r.ent}i}Q_{\text{w.ent}i}S_{\text{m.ent}i} + C_{\text{b.ent}i}Q_{\text{b.ent}i} \rho_{\text{b}i} + C_{\text{w.tr}i}Q_{\text{w.tr}i} + C_{\text{r.tr}i}Q_{\text{w.tr}i}S_{\text{m.tr}i} + C_{\text{b.tr}i}Q_{\text{b.tr}i} \rho_{\text{b}i} + C_{\text{w.c}i}Q_{\text{w.c}i} + C_{\text{r.c}i}Q_{\text{w.c}i}S_{\text{m.c}i} + C_{\text{w.gr}i}Q_{\text{w.gr}i} - C_{\text{r}i}Q_{\text{w}i}S_{\text{m}i} - C_{\text{b}i}Q_{\text{b}i} \rho_{\text{b}i} - C_{\text{w}i}Q_{\text{w}i} - \lambda_{\text{d}}V_iC_i,$$
(7)

$$C_{\rm ri} = C_{\rm wi} \,\rho_{\rm bi} K_{\rm d.ri} \,, \tag{8}$$

$$C_{\rm bi} = C_{\rm wi} K_{\rm d,bi} \,, \tag{9}$$

$$C_i = C_{\rm wi} R_i \,, \tag{10}$$

$$R_i = R_{\rm ri} + R_{\rm bi} \,, \tag{11}$$

$$R_{\rm ri} = 1 + S_{\rm mi} \,\rho_{\rm bi} K_{\rm d.ri} \,, \tag{12}$$

$$R_{\rm bi} = \frac{V_{\rm bi}}{V_{\rm wi}} (1 - \Delta_i) K_{\rm d.bi} \,\rho_{\rm bi} \,. \tag{13}$$

With certain initial conditions and specific conditions of ingress of radionuclides into the chambers, the differential equation (7) is solved analytically.

In this stage of investigation, we consider three cases of ingress of radioactive contamination into the river system.

I. Ingress of Radionuclides in the Inlet Cross Section of the River during a Limited or an Unlimited Period of Time. With the initial condition

$$t = 0$$
, $C_{wi}(0) = 0$, $C_{ri}(0) = 0$, $C_{bi}(0) = 0$ (14)

and the condition of ingress of radionuclides

$$C_{\text{ent1}} = C_{0\text{ent1}} \exp\left(-\lambda_{d}t\right) \tag{15}$$

we obtained the analytical solution of Eq. (7)

$$C_{\text{w}i}(\mathbf{I}) = C_{0\text{ent1}} \exp(-\lambda_{d}t) A_{0} \prod_{j=1}^{i} \lambda_{\text{ent1}} \left[B_{i} - \sum_{j=1}^{i-1} B_{ij} \exp(-\lambda_{j}t) - B_{jj} \exp(-\lambda_{i}t) \right] \delta_{1}.$$
(16)

II. Fallout of Radionuclides on the Water Surface of the Entire River or Separate Sections of It. It is assumed that the radionuclides that entered interact with the elements of the river medium instantly. With the initial condition

$$t = 0, \quad C_{0i}(0) = a_{0i}/H_{mi}$$
 (17)

we obtained the analytical solution of Eq. (7) in the following form:

$$C_{\rm wi} ({\rm II}) = \exp\left(-\lambda_{\rm d} t\right) \left\{ \sum_{j=1}^{i-1} D_{ij} \exp\left(-\lambda_{j} t\right) + \left(C_{0i} A_{i} - \sum_{j=1}^{i-1} D_{ij} \right) \exp\left(-\lambda_{i} t\right) \right\}.$$
(18)

III. Ingress of Radionuclides into the River System with Surface Flow from the Contaminated Water Catchment (or Separate Sections of It). To determine the amount of radioactive contamination entering the river system from the surface of the water catchment, we write the equation of balance of the radionuclides on the *i*th section of the water catchment:

$$F_{\rm c}h_{\rm c}\frac{dC_{\rm s}}{dt} = -C_{\rm w.s}Q_{\rm w.c} - C_{\rm r.s}Q_{\rm w.c}S_{\rm m.c} - C_{\rm w.s}U\alpha F_{\rm c} - \lambda_{\rm d}F_{\rm c}h_{\rm c}C_{\rm s},$$
(19)

$$C_{\rm s} = C_{\rm w.s} \theta_{\rm s} R_{\rm s} \,, \tag{20}$$

$$R_{\rm s} = \left[1 + K_{\rm d.s} \frac{\rho_{\rm s} \left(1 - \mu_{\rm s}\right)}{\theta_{\rm s}}\right],\tag{21}$$

$$C_{\rm r.s} = C_{\rm w.s} K_{\rm d.s} \,\rho_{\rm s} \,. \tag{22}$$

Integrating Eq. (19), we obtain the solution for the *i*th section of the water catchment:

$$C_{\text{w.si}} = \frac{C_{\text{s0i}}}{\theta_{\text{si}} R_{\text{si}}} \exp\left(-(\lambda_{\text{wi}} + \lambda_{\text{d}}) t\right).$$
(23)

With the initial condition (14) and the condition of ingress of radionuclides with surface flow from the water catchment (23), we found the solution of Eq. (7) for the *i*th chamber in the following form:

$$C_{wi} (III) = \exp(-\lambda_{d}t) \left[w_{i} \exp(-\lambda_{wi}t) + \sum_{j=1}^{i-1} w_{i} \exp(-\lambda_{wi}t) - \sum_{j=1}^{i-1} E_{ij} \exp(-\lambda_{j}t) - E_{ii} \exp(-\lambda_{i}t) \right], \quad (24)$$

$$w_i = \frac{C_{s0i}}{\theta_{si} R_{si}} \frac{\lambda_{ci}}{\lambda_i - \lambda_{wi}} \,. \tag{25}$$

Equations (7)–(25) are the main part of the mathematical model describing migration of radionuclides in the river system. For obtaining a complete solution of the problem, the system of equations (7)–(25) must be closed by the equations of transfer of suspended and bottom sediments which are typical of the specific water object studied [8, 9].

When radionuclides enter the river system simultaneously in all the ways considered, their concentration will be equal to the sum of the concentrations:

$$C_{wi} = C_{wi} (I) + C_{wi} (II) + C_{wi} (III) .$$
 (26)

The removal of radionuclides through control cross sections during a fixed period of time is determined as follows:

• in soluble form

$$S_{\mathrm{w}i} = Q_{\mathrm{w}i} \int_{0}^{t_k} C_{\mathrm{w}i} dt , \qquad (27)$$

• on suspensions

$$S_{\rm ri} = S_{\rm wi} S_{\rm mi} K_{\rm d.ri} \rho_{\rm bi} , \qquad (28)$$

• with entrained sediments

$$S_{bi} = S_{wi} \frac{Q_{bi}}{Q_{wi}} K_{d.bi} \rho_{bi} .$$
 (29)

The calculated parameters of removal of radionuclides allow one to determine the coefficients of flow of radionuclides in the closing chamber which are typical of the considered section of the water catchment:

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• the coefficient of liquid flow

$$K_{\rm liq} = \frac{S_{\rm wk}}{\sum_{i=1}^{k} C_{\rm s0i} h_{\rm ci} F_{\rm ci}},$$
(30)

• the coefficient of solid flow

$$K_{\rm s} = \frac{S_{\rm rk} + S_{\rm bk}}{\sum_{i=1}^{k} C_{\rm s0i} h_{\rm ci} F_{\rm ci}},$$
(31)

• the coefficient of total flow

$$K_{\rm com} = \frac{S_{\rm wk} + S_{\rm rk} + S_{\rm bk}}{\sum_{i=1}^{k} C_{\rm s0i} h_{\rm ci} F_{\rm ci}}.$$
(32)

To develop and verify the model suggested, we took the basin of the Iput river as an experimental one. A large body of experimental data, which is a result of the long-standing monitoring and field observations conducted by the State Committee for Hydrometeorology of the Republic of Belarus and the "Taifun" Scientific-Production Association (Russia), have been accumulated for this river basin [10].

The Iput river is in the Dnieper–Sozh basin and flows over the Belarus–Bryansk "spot" with contamination levels of 37 to 2220 kBq/m² for ¹³⁷Cs and of 0.37 to 31.1 kBq/m² for ⁹⁰Sr. Detailed observations of the level of contamination of the river water and bottom sediments and the removal of radionuclides of the Iput river on the territory of Belarus began in 1987 in control cross sections of the village of Vylevo and the town of Dobrush in front of the dam and behind it (the terminal cross section) and are in progress now. On the territory of Russia, similar observations were conducted in eight control cross sections, unfortunately only in 1991–1993 and they were not sufficiently frequent. These experimental data on the Iput river from its source to the observation point in Dobrush were used to graduate the parameters of the model and to test it.

In verification of the model and in computational studies of the change in the radiation situation in the Iput river basin, we covered virtually the entire water catchment of the river from its source to the terminal control cross section at Dobrush. This allowed us to take into account the effect of the nonuniform density of radioactive fallout on the territory of the water catchment and the particular features of formation of the river flow on the character of contamination of the river system [11–13].

Numerical realization of the model required determination of the analytical content of its second block, namely, mathematical description of the transport of suspended and bottom sediments in a hydraulically stationary formulation of the problem. To calculate the transport of sediments, we used a hydraulic-morphological method. It is characterized by a rather wide set of semiempirical dependences for determination of the flow rate (turbidity) of suspended and entrained sediments on the basis of the data on the flow parameters and the granulometric composition of sediments [8, 14, 15]. In its hydraulic and morphometric characteristics the Iput river belongs to middle rivers. The data of the field observations were used for analysis of the most popular recommendations [8, 14, 15] and also for selection and testing of analytical dependences for calculation of the flow rate of suspended and bottom sediments for rivers of this type. The results of this study are published in [16] and were used in the present work.

Experimental data on contamination of the river system by radionuclides ¹³⁷Cs and ⁹⁰Sr along the Iput river channel and in time were used to graduate the model parameters against chambers [10–13]. Such parameters of the model as the coefficients of distribution in the systems "water–bottom sediments," "water–suspensions," and "water–soil of water catchment," the amount of bottom sediments, the liquid and solid flows from the surface of the water catchment, and feeding of the river with ground water were selected for each section of the river from the ranges determined in the parametric study [17–20] so that the experimental and calculated values of the concentrations of radionuclides in water, on suspensions, and in bottom sediments along the river channel and in time have minimum divergences.



mean; 4) minimum] values of the specific activities of ¹³⁷Cs (a) and ⁹⁰Sr (b) in water-soluble form (C_w), on suspensions (C_r), and in bottom sediments (C_b) along the Iput river channel in 1991. C_w and C_r , Bq/m³; C_b , Bq/kg; L, km.

Comparison of the calculation results with the experimental data on the concentrations of cesium-137 and strontium-90 in water, on suspensions, and in bottom sediments along the river channel obtained in 1991 is given in Fig. 1. It shows that the calculated concentrations of ¹³⁷Cs and ⁹⁰Sr in water-soluble form and in bottom sediments along the Iput river channel are in satisfactory agreement with the data of field measurements, whereas the calculated concentrations of the radionuclides on suspended sediments fall within the range of measured values to a lesser extent. The inconsistency of individual measured and calculated points is caused by both considerable uncertainty of the morphometric characteristics of the channel, the sorption parameters of the elements of the river system and the water catchment, the erosion characteristics of the water catchment, etc. and insufficient accuracy and frequency of experimental measurements. Similar results of comparison of the calculated and experimental values of the concentrations of the radionuclides were obtained for the years 1992–1993 [17].

Particular attention was paid to the agreement of experimental and calculated data in the terminal control cross section in Dobrush, for which the most complete experimental information was available.

Figure 2 shows a comparison of the data of field measurements made during ten years (1987–1997) and the results of calculations of the concentrations of ¹³⁷Cs and ⁹⁰Sr in water, on suspensions, and in bottom sediments related to the terminal control cross section in Dobrush. The calculations were made for hydraulically constant conditions within the entire period time, whereas under real conditions the flow rate and morphometric characteristics of the channel had both seasonal and yearly variations. Despite this fact, as is seen from Fig. 2, most computation points fall within the range of maximum and minimum yearly values of the



Fig. 2. Change in the calculated (1) and measured [2) maximum; 3) mean; 4) minimum] values of the water flow rate (Q_w) and the specific activities of ¹³⁷Cs (a) and ⁹⁰Sr (b) in water (C_w), on suspensions (C_r), and in bottom sediments (C_b) during 1987–1997 in the terminal control cross section of the river (Dobrush). *t*, years; Q_w , m³/sec; C_w and C_r , Bq/m³; C_b , Bq/kg.

experimental data on the concentrations of ¹³⁷Cs and ⁹⁰Sr. This agreement of the experimental data and the results of the calculations along the Iput river channel and in time (Figs. 1 and 2) led to the conclusion that within the framework of the assumptions made the model adequately and quite satisfactorily describes the processes of migration of radionuclides in the river system and can be used for prediction estimates.

NOTATION

k, number of controlled chambers; t, time; L, distance from the river source; V, $V_{\rm w}$, and $V_{\rm b}$, volumes of the chamber, the water in the chamber, and the active layer of bottom sediments in the chamber of the river system; F, area of the chamber; $H_{\rm m}$, mean depth of the water in the chamber; $F_{\rm c}$, area of the section of the water catchment; $h_{\rm c}$, layer of contaminated soil of the water catchment; $Q_{\rm w}$ and $Q_{\rm b}$, flow rates of the water in the chamber and of the entrained sediments; $Q_{r,b}$, sedimentation-turbidity flow; S_m , mean turbidity of the water in the chamber; $S_{m,tr}$, turbidity of the water coming with tributaries; $S_{m,c}$, turbidity of the water coming with surface flow from the water catchment; C_i , total specific activity of the radionuclides in the river system; C_{0ent} , specific activity of the radionuclides in the inlet cross section of the river; C_w , C_r , and C_b , specific activities of the radionuclides in soluble form, on suspensions, and in bottom sediments of the river system; a_0 , density of fallouts of radionuclides on the water catchment and the water surface of the river section; C_{s} , total specific activity of the contaminated soil of the water catchment; $C_{w.s}$ and $C_{r.s}$, specific activity of the radionuclides in soluble form and sorbed on the particles of the water catchment soil; θ_s , μ_s , and ρ_s , humidity, porosity, and density of the soil of the water catchment; Δ and ρ_b , porosity and density of bottom sediments; $K_{d.r.}$, $K_{d.b.}$, and $K_{d.s.}$, coefficients of distribution of the radionuclides in the systems "water-suspensions," "water-bottom sediments," and "water-soil of water catchment"; $a_{w,r}$ and $a_{w,b}$, rate constants of sorption of the radionuclides from water by suspended particles and bottom sediments; R, common factor of trapping of radionuclides in the river system; R_r , R_b , and R_s , factors of trapping of radionuclides by suspensions, bottom sediments, and soil of the water catchment; λ , constant of convective transfer on the section of the river system; λ_w , constant of convective transfer on the section of water catchment; λ_{ent} , constant of convective transfer of radionuclides that entered the river system in the inlet cross section; λ_c , constant of convective transfer of radionuclides that entered the river system with the surface flow; λ_{d} , constant of radioactive disintegration; S_w, S_r, and S_b, removal of radionuclides through control cross sections during a fixed period of time in soluble form, on suspensions, and entrained sediments; K_{liq} , K_{s} , and K_{com} , coefficients of the liquid, solid, and total flows of radionuclides; U, norm of atmospheric fallout; α , coefficient of underground flow; δ_1 , function controlling the durability or periodicity of ingress of radionuclides in the inlet cross section; A₀, A_i, B_i, B_{ii}, B_{ii}, D_{ij}, E_{ij}, E_{ii}, and w_i, coefficients obtained by integration of Eq. (7). Subscripts: i, chamber number; w, water; r, suspensions; b, bottom sediments; s, soil; c, water catchment; tr, tributary; gr, ground flow; 0, initial value; ent, value at the entrance to the chamber; m, mean value; d, radioactive disintegration; liq, liquid; com, total; 1, first chamber.

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