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DISPERSION MODEL AND PROGRAM FOR CALCULATION OF RADIOCHEMICAL CONTAMINATION IN RIVERS WITH CONSIDERATION FOR INTERACTION OF RADIONUCLIDES, SEDIMENT, AND SUSPENDED MATTER

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A mathematical model and the RIVER-01 software package fc. simulation of river transport of radiochemical contaminants are presented. Test calculations are carried out, and the effects of various factors are investigated.

Introduction. The process of radionuclide transport in rivers is determined generally by a number of parameters. Natural waters comprise complex physicochemical systems containing mineral and organic compounds of differing dispersions. The water composition depends mainly on the type of soil it contacts, the action of living organisms, hydrological regime, season, temperature, and illumination. In addition, the water composition changes with time within the limits of a single basin on various portions of the river.

Capture of radionuclides by suspensions and deposits (sediment) depends on the type of suspension, fraction size, soil sorption capacity, radionuclide type, flow rate, and alluviation characteristics, including the sediment granulometric composition.

Presently, several approaches to and models of the river transport of contamination exist [1-5]. The choice depends on the particular problem, the scale of contamination transport, the level of information required, etc. An attempt to develop a detailed mathematical description of the systems leads to a number of parameters in the equations of the model. The present-day level of ecological studies does not allow us to find many of the necessary parameters. In addition, one should bear in mined that many of them are determined with a large error, and the application of a complicated model that takes into account fine processses is usually not justified. This remark by no means negates the expediency of practical applications of complicated multidimensional transport models in cases when the usage efficiency and expenses for software development are reasonable.

Mathematical Model. We propose a so-called finite-volume model for calculation of radiochemical contamination transport with river flow. The river is presented as a finite number of completely mixed volumes containing three components: water, suspended matter, and sediment (Fig. 1). The mean concentrations for each of the volumes are determined, and transfer coefficients for correlating neighboring elements (volumes) are established. The mass balance, or transfer, between the volumes is described on the basis of time-dependent linear differential equations with constant coefficients, and for each cross-section of the river the corresponding system of equations can be written as follows:

$$\frac{d}{dt}(M_1C_1) = \Delta(qC_1) + k_{21}C_2 + k_{31}C_3 + k_{12}C_1 + k_{13}C_1 + \lambda M_1C_1 ,$$

$$\frac{d}{dt}(qSC_2) = \Delta(qSC_2) + k_{12}C_1 + k_{32}C_3 + k_{21}C_2 + k_{23}C_2 + \lambda M_2C_2 , \quad (1)$$

$$\frac{d}{dt}(M_3C_3) = k_{13}C_1 + k_{23}C_2 + k_{31}C_3 + k_{32}C_3 + \lambda M_3C_3 ,$$

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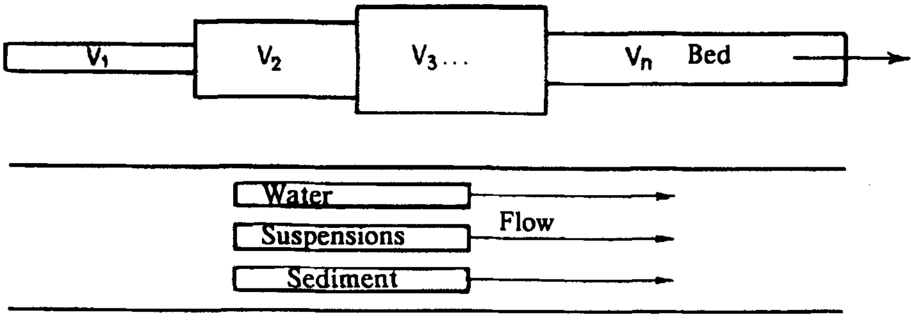

Fig. 1. Finite-volume model and components participating in calculations of admixture transport in river.

$\Delta(qC_1) = (qC_1)_{i+1} - (qC_1)_i$ is the difference in the content of the j-th component in neighboring cross-sections of the river.

Equations (1) are written correspondingly for water, suspended matter, and sediment. Let us consider the derivation of dependences for calculation of coefficients k_{mn} by the example of radionuclide transfer from water into suspension and back. The water–suspension system is considered to be in the equilibrium state, and the time of passing to this state (relaxation time τ_{12}) is known. If the equilibrium distribution of radionuclides between water and suspensions k_d is determined as the ratio of the specific radionuclide concentration on suspensions to their concentration in water, then one can write

$$k_d = \frac{C_{susp} M_1}{M_2 C_w} = \frac{C_{susp} M_1}{(C_1 - C_{susp}) M_2}, \tag{2}$$

where M_1 is the mass of the water, M_2 is the mass of the suspension, C_1 is the initial concentration in the water, and C_{susp} is the portion that is transferred onto the suspension. Radionuclide concentration on the suspension can be determined in terms of the relaxation time τ_{12} as $k_{12}C_1\tau_{12}/M_1 = C_{susp}$; hence, in view of (2), we obtain

$$\frac{k_{12} C_1 \tau_{12}}{M_1} = \frac{k_d M_2 C_1}{M_1 + k_d M_2}, \quad \text{hence} \quad k_{12} = \frac{k_d M_2 M_1}{(M_1 + k_d M_2)\tau_{12}}. \tag{3}$$

In a similar manner we can obtain expressions for the coefficients for concentration of radionuclides passed from suspensions to water:

$$k_{21} = \frac{M_1 M_2}{(M_1 + k_d M_2)\tau_{12}}, \quad k_{21} = \frac{k_{12}}{k_d}; \tag{4}$$

$$k_{13} = \frac{M_1 M_3}{(M_1 + k_d M_3)\tau_{13}}, \quad k_{31} = \frac{k_{13}}{k_d}. \tag{5}$$

Depending on whether or not a portion of admixtures has precipitated in the given portion of the river, variation of its concentration in the given volume takes place. Formulas for evaluation of the corresponding transfer coefficients are as follows:

$$k_{23} = \max(0, q\Delta S), \tag{6}$$

$$k_{32} = \max(0, -q\Delta S). \tag{7}$$

In the general case, coefficients k_d are functions of a particular physicochemical process and can differ for water–suspension and water–sediment systems; however, for simplicity, we did not discriminate between them in the present work. Consideration of the specific features of these coefficients is not complicated in mathematical implementation and, when necessary, is easily realized in the model. In addition, a number of simplifying assumptions are used in the model: for each of the volumes the shore line is considered straight, it is assumed also that the sediment is not mixed on reaching the equilibrium condition (although, in practice, numerous processes of precipitation and turbidization take place downstream), sediment slippage is neglected, the masses of components participating in the process are considered constant in time for each of the portions, processes of leaching of a portion of the adsorbed material out of the sediment, and sediment motion downstream are not taken into account.

Initial Data and Implementation of the Model. The model presented is rather simple in implementation and does not require a great amount of initial information; however, it is highly sensitive to input data. To start calculations, one needs information on turbidity for each portion (concentration of suspended matter in the water), to divide the river into portions with constant, wherever possible, properties, and to determine experimentally the coefficients of equilibrium distribution of radionuclides between the sediment and water. Usually, k_d values are determined by the formula [2]

$$k_d = \frac{V}{M}\left(\frac{C_0}{C_\infty} - 1\right), \tag{8}$$

where V is the sediment volume, M is the mass of the water, C_0 is the radionuclide concentration in the water at time zero, C_∞ is the concentration after establishing equilibrium.

As has been noted, the model is sensitive to variations in k_d, since this parameter includes in an integrated manner a number of the above-discussed factors.

To determine the water–suspension mass $M_1 + M_2$ in a given volume, we calculated the sum of water volumes with identical characteristics multiplied by the corresponding densities. With the known concentration of suspensions in the water, the corresponding masses of the water and suspensions are easily determined. The sediment mass can be found by two methods.

In the first one, the sediment mass is determined from the flow rate of the bed load, and the sediment is considered a bottom layer that can move by jumping and rolling. In order to determine the flow rate of bed loads, their initial dragging rate is calculated. One possible expression for their evaluation proposed in [6] is as follows

$$V_{\text{init}} = 4.6 d_{\text{av}}^{1/3} H^{1/6}, \tag{9}$$

and flow rates of the bed load G_{bed} on each river section were determined by the dependence [6]

$$G_{\text{bed}} = 2.08\,(V_{\text{av}}/V_{\text{init}})\,(V_{\text{av}} - V_{\text{init}})\,(d_{\text{av}}/H)^{0.1}\,d_{\text{av}}. \tag{10}$$

By summing the flow rates over all subdivisions of the cross-section and knowing the flow rate of the river in a given portion and its length, one can determine the mass of the bed load.

In the second case the density of the bed load lies within the limits of from 2.55 to 2.80 kg/m^3 [6]. The joint action of the growth of new sediment and radioactive decay in the sediment substantially restricts the thickness of the effective layer to a value of about several centimeters. Moreover, experiments have shown that the main portion of radioactivity is concentrated within the upper 5-cm of the layer. Knowing the geometry of a given river portion, one can easily determine the sediment mass M_3.

The relaxation time τ_{mn} is determined experimentally. It is known that for alkaline solutions with pH 12, time of establishing equilibrium between the water and sediment for Cs^{137} is about 10 min, and for neutral waters with pH 7 it is about 20 min. For Sr^{90} the relaxation time lies in the interval of from 4 to 10 min [4, 5].

Under the condition that the masses M_1, M_2, and M_3 are constant in time, the problem posed can be solved by solving a system of ordinary differential equations with constant coefficients. For this purpose, we used the Runge–Kutta method. The initial conditions for the first portion of the river were set up as follows: $c_1 = c_1^0$, $c_2 = 0$, $c_3 = 0$. For each successive portion the initial conditions were determined from the solution for the preceding

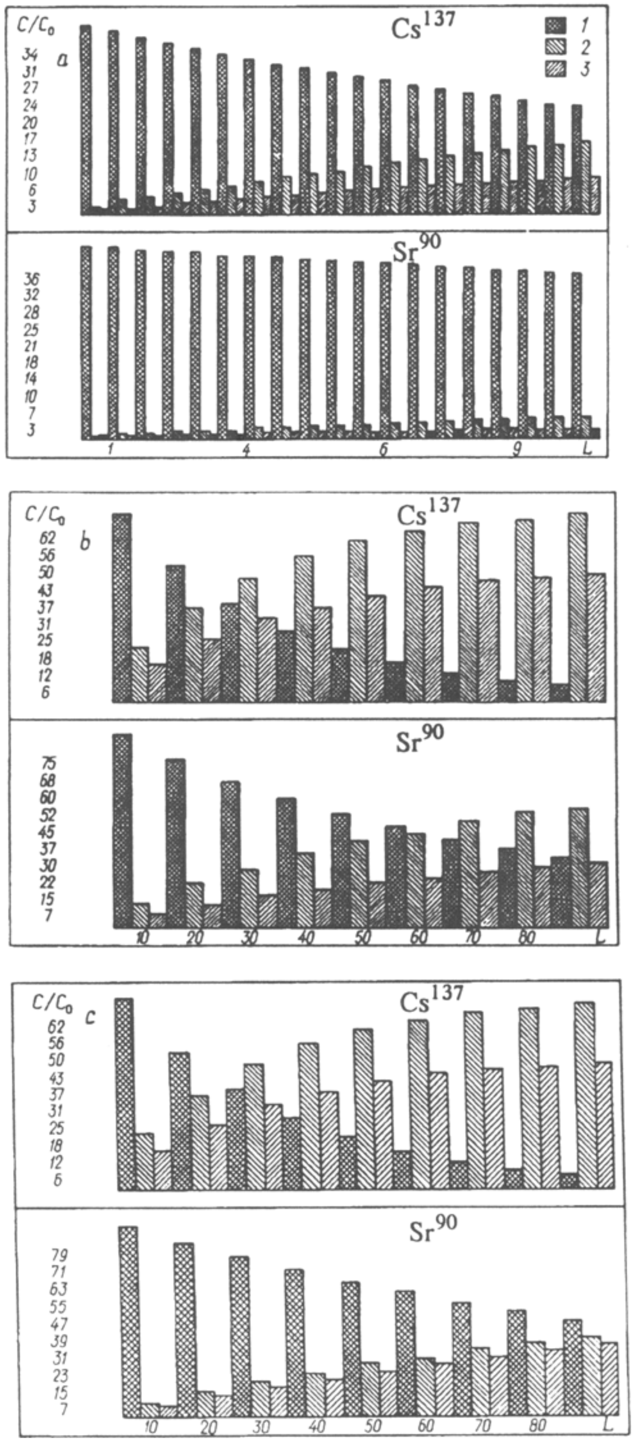

Fig. 2. Diagrams of distribution of contaminating admixtures (C/C_0, %) downstream for various values of parameters (mean flow rate of the river, relaxation rate, and density of suspended particles); a) $L = 10$ km, $n = 20$, $A = 0.1$ Ci/kg, $Q = 0.005$, $V_{av} = 1$ m/sec, k_d for Cs^{137} and Sr^{90} are 3000 and 50, respectively; b) $L = 100$ km, $n = 10$, $A = 0.1$ Ci/kg, $Q = 0.0015$, $V_{av} = 0.5$ m/sec, k_d for Cs^{137} and Sr^{90} are 3000 and 50, respectively; c) $L = 100$; $n = 10$, A for Cs^{137} and Sr^{90} equal 0.1 and 0.01 Ci/kg, respectively, $Q = 0.0015$, $V_{av} = 1$ m/sec, k_d for Cs^{137} and Sr^{90} are 3000 and 50, respectively, τ_{mn} for strontium precipitation into sediment in this variant is two times greater than in variant b). 1) water; 2) suspension; 3) sediment.

one. The finite integration time was determined for each portion as the mean time expended by the flow to pass a preset length.

In summary, the following quantities are considered as initial data:

- the length of the working portion of the river (the number of cross-sections over the length);
- contaminant quantity (radionuclide source);
- the number of portions along the transverse direction with an identical depth and flow rate;
- the flow rate in a given portion over both depth and width;
- the depth of each portion;
- half-lives of radionuclides;
- density of the water–suspension solution;
- coefficients k_d of distribution between water and sediment;
- relaxation coefficients τ_{ij} (water–suspension; water–sediment);
- average concentration of the suspended matter;
- average sediment particle size.

Results and Discussion. For each contaminant (radionuclide) we calculated its concentration and plotted a diagram of its distribution in water, suspensions, and sediment. The model generally makes it possible to take into account up to 20 different admixtures in water, and Fig. 2 shows distributions of the type by the example of two radionuclides, cesium and strontium, that are known as being most dangerous. In addition, these radionuclides have different behaviors under conditions of water transport and make it possible to simulate highly and poorly soluble admixtures. We are aware of the fact that, to provide a detailed calculation of a river system, one should know a great amount of empirical information on the conditions of river flow (hydrograph), the characteristics of water and soils on each of the portions, flow regimes and transfer processes in the river, etc. At the present stage, we have posed the problem of development of a general mathematical model and its software implementation.

The qualitative character of the diagrams depends on a number of factors (powers of sources of waste disposal, distribution coefficients, relaxation time, and others), and, therefore, Fig. 2 presents parametric studies of the effect of various factors on the process of transport of admixtures downstream.

The distribution coefficient k_d, governs to a large extent the behavior of admixtures in the river. Its values determine the direction of the precipitation–turbidization process. The main contamination either remains in the water or enters suspensions and sediment.

The dependence on particle diameter is also notable, since this parameter enters the expression for calculation of the sediment mass and finally determines the number of precipitated particles. In addition, the particle diameter affects k_d and thus changes the shape of the diagrams.

The proposed RIVER-01 software package is designed for investigations of complex processes of river contamination transport, has a modular structure, and is easily adapted to various transport problems.

NOTATION

C_j, radioactivity of a unit mass of component, Ci/kg; A, activity of contamination source, Ci/kg; k_{mn}, coefficient of transfer from the control volume m to n, kg/sec; S, mean relative concentration of the suspended matter; q, mean water flow rate in river, kg/sec; λ, radioactive decay constant, sec^{-1}; M_j, mass of the j-th component, kg; k_d, coefficient of the equilibrium distribution of radionuclides in water; d_{av}, average diameter of movable fractions, m; H, flow depth, m; V_{stop}, velocities at which particles of bed loads are stopped; V_{av}, average flow rate of the river, m/sec; L, total length of the model portion of the river, km; n, number of elementary portions of the river; Q, relative density of suspended matter in water.

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