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NUMERICAL SIMULATION OF MIGRATION OF RADIONUCLIDES IN SOIL AFTER RADIOACTIVE FALLOUT

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The authors consider migration of radionuclides in soil with moisture transfer taken into consideration. It is assumed that in their motion the radionuclides can be adsorbed in accordance with Henry's nonequilibrium law. A numerical procedure is suggested for solution of the problem. Results of mathematical simulation of migration of the radionuclides are presented and can be used to predict soil contamination.

1. Introduction. Nuclear accidents can be accompanied by fallout of radionuclides into the earth's atmosphere, followed by their propagation under the action of the air mass. Here the heaviest radionuclide fractions settle out, forming a radioactive trace on the earth's surface. Subsequently, from the earth's surface the radionuclides can penetrate into soil, contaminating it. Predicting this process in an urgent problem and is of great interest.

Actually, some attempts to calculate penetration of radionuclides into soils and rocks had been made previously in some works to predict possible radioactive contamination in connection with subterranian nuclear explosions (see, for example, [1-5] and their references). However, the solution of this problem differs substantially from the aforementioned one by the fact that in a subterranian explosion pressure filtration is the main mechanism of propagation of radionuclides, while in the first case migration of radionuclides is presumably caused by moisture-transfer processes. Up to the present, mathematical simulation has not actually been used to investigate migration of radionuclides in soils with moisture transfer taken into consideration. In what follows, a solution of this problem is considered.

In order to predict correctly the development of the processes that accompany beyond-design nuclear accidents, it is necessary to use models to simulate both the individual physicochemical processes and the whole set of successive pnenomena.

It is known [6] that mathematical simulation is a set of operations consisting of the following stages: 1) development of computational models based on investigation of particular physicochemical processes; 2) their verification on available experimental setups; 3) analysis of variants associated with prediction of the development of the processes and optimization of possible consequences.

Methods of evaluation of radioactive fallout into the atmosphere and its subsequent propagation within the environment due to a nuclear accident are given in [7, 8].

The present work is aimed at predicting migration of radionuclides in soil with account for moisture transfer, nonequilibrium adsorption, and convective diffusion.

2. Physicomathematical Model and Formulation of the Problem. Let a radioactive trace have been formed on the earth's surface and the radionuclides that fell out filter into the soil, under the action of precipitation. Essentially, the problem is divided into two: one on simulation of moisture transfer in the soil and one on migration of the radionuclides. Since the weight fraction of the radionuclides is extremely small in comparison with the moisture, without substantial error the radionuclides can be assumed to be a dynamically neutral impurity [9]. This is a model in which the velocity is determined by the motion of the main component (the moisture, in the

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present case), the impurity has no noticeable effect on the phenomenon, and the smaller the amount of the impurity, the more accurate the approximation of a dynamically neutral impurity.

Actually, propagation of radionuclides in soil depends on both the type of soil and the special features of the interaction of the radionuclides with the soil complex. For example, after the Chernobyl nuclear accident fuel particles decayed substantially (up to 60%) in sands, while in chernozem this decay was much lower [10]. It is known that ^{137}Cs is incorporated into the crystal lattice of minerals and is irreversibly transformed into a sorbed state, while ^{90}Sr is sorbed by soil by an ion-exchange mechanism, and therefore, the percentage of water-soluble and exchange forms of ^{90}Sr is increased [11].

Thus, vertical migration of radionuclides depends on the processes of transformation of chemical forms and the free-moisture content in the soils, and therefore, the mathematical model of migration of radionuclides includes moisture-transfer equations and transfer equations for radionuclides with account for the kinetics of sorption, diffusion, and radioactive decay.

To describe migration of radionuclides we used a system of equations of the form [9]

$$\frac{\partial c}{\partial t} + \frac{\partial a}{\partial t} + \frac{\partial vc}{\partial z} + \lambda (a + c) = \frac{\partial}{\partial z} D(v) \frac{\partial c}{\partial z}, \tag{2.1}$$

$$\frac{\partial a}{\partial t} = \beta c - (\lambda + \beta\gamma) a, \tag{2.2}$$

which includes decay of the radionuclides, convective diffusion, and nonequilibrium adsorption. In (2.1) the following expression [9] is used for $D(v)$:

$$D(v) = D_0 + D_1 v.$$

It is assumed that at the initial time there is no radionuclide in the flow or in the adsorbed state:

$$c(z, t = 0) = 0, \quad a(z, t = 0) = 0, \tag{2.3}$$

and at $z = 0$ its concentration changes with time:

$$c(z = 0, t) = \Psi(t).$$

At a rather long distance from the surface its concentration is also zero:

$$c(z = L, t) = 0. \tag{2.4}$$

In the system of equations (2.1) and (2.2) the velocity of propagation of the radionuclides v coincides with the velocity of filtration transfer of water and is determined in terms of the of the gradient of the head H:

$$v = -k_w \operatorname{grad} H, \quad H = \frac{P_w}{\rho g} - z. \tag{2.5}$$

For the moisture-transfer coefficient of a homogeneous porous medium, the following power relation is used [12]:

$$k_w = k \left(\frac{w}{w_*}\right)^{\sigma}. \tag{2.6}$$

A linear relation is ordinarily used as the relation $P_w(w)$ [13]:

$$P_w = -a + bw, \tag{2.7}$$

where a and b are constant coefficients to be determined experimentally.

The basic moisture-transfer equation is obtained from the continuity equation

$$\frac{\partial w}{\partial t} + \operatorname{div} v = 0 \tag{2.8}$$

after substitution of (2.4)-(2.6) into it. It is reduced to the following quasilinear equation for the saturation w:

$$\frac{\partial w}{\partial t} = \frac{\partial}{\partial z}\left[\frac{k\beta}{\rho g}\left(\frac{w}{w^*}\right)^{\sigma}\frac{\partial w}{\partial z}\right] - k\frac{\partial}{\partial z}\left(\frac{w}{w^*}\right)^{\sigma}. \tag{2.9}$$

As boundary conditions for Eq. (2.8) we assume that on the earth's surface at $z = 0$ the wettability is total:

$$w(z = 0, t) = w^*. \tag{2.10}$$

At a rather long distance L from the earth's surface, it is assumed that

$$w(z = L, t) = 0. \tag{2.11}$$

It is assumed that at the initial moment the saturation in the porous medium is also zero:

$$w(z, t = 0) = 0. \tag{2.12}$$

Moisture transfer in the ground is simulated by the solution of boundary-value problem (2.9)-(2.12), then the velocity $v(z, t)$ is found from formulas (2.5) and (2.7), and subsequently the problem on the propagation of radionuclides is solved in accordance with the system of equations (2.1) and (2.2).

3. Method of Solving the Problem. It is impossible to solve analytically the problems that are formulated above in the general case, and therefore, we calculated the propagation of radionuclides in the ground with account for moisture transfer by a numerical method. For this purpose, in the above problem the differential operators were replaced by difference ones and the problem itself was reduced to a system of algebraic equations, which was solved on a computer.

Assuming

$$u = H + \frac{a}{\rho g}, \tag{3.1}$$

it is possible to write Eq. (2.9) in the form

$$\frac{\partial u}{\partial t} = \frac{\partial}{\partial z}\left[\kappa\left(\frac{w}{w^*}\right)^{\sigma}\frac{\partial u}{\partial z}\right], \quad \kappa = \frac{kb}{\rho g}. \tag{3.2}$$

The above relation coincides with the well-known equation of nonlinear heat conduction that has been considered repeatedly in the literature [14, 15].

Formula (3.2) in the dimensionless variables

$$z = Lz', \quad t = \frac{L^2}{\chi}t', \quad u = \frac{bw^*}{\rho g}u', \quad \chi = \frac{L\rho g}{bw^*} \tag{3.3}$$

has the form (the primes are omitted)

$$\frac{\partial u}{\partial t} = \frac{\partial}{\partial z}\left[(u + \chi z)^{\sigma}\frac{\partial u}{\partial z}\right]. \tag{3.4}$$

With account for the dimensionless variables the initial and boundary conditions are written as

$$u(z, 0) = -\chi z, \tag{3.5}$$

$$u(0, t) = 1. \tag{3.6}$$

$$u(L, t) = -\chi L. \tag{3.7}$$

Using the grid functions $u_j^n \cong u(z_j, t^n)$, where $z_j = jh_2$ (h_2 is the coordinate step), $t^n = n\tau$ (τ is the time step), from (3.4) we arrive at the difference equation

$$\frac{\tau}{h_z^2} u_{j-1}^{n+1} \left(\frac{u_j^n - u_{j-1}^n}{2} + \chi z_{j-1/2} \right)^\sigma -$$

$$- u_j^{n+1} \left[1 + \frac{\tau}{h_z^2} \left(\frac{u_j^n - u_{j-1}^n}{2} + \chi z_{j-1/2} \right) + \frac{\tau}{h_z^2} \left(\frac{u_{j+1}^n - u_j^n}{2} + \chi z_{j+1/2} \right) \right] +$$

$$+ \frac{\tau}{h_z^2} u_{j+1}^{n+1} \left(\frac{u_{j+1}^n - u_j^n}{2} + \chi z_{j+1/2} \right)^\sigma = - u_j^n, \tag{3.8}$$

$$j = 1, 2, ..., J - 1; \quad n = 0, 1, ..., N - 1,$$

with the conditions

$$\begin{gathered} u_j^0 = -\chi z_j, \quad j - 0, 1, ..., J; \quad u_0^{n+1} = 1, \quad n = 0, 1, ..., N - 1; \\ u_j^{n+1} = - L\chi, \quad n = 0, 1, ..., N - 1. \end{gathered} \tag{3.9}$$

The system of equations (3.8) with conditions (3.9) was solved using factorization and iteration methods on each time level.

To solve the problem on the propagation of the radionuclides with account for moisture transfer, in the system of equations (2.1) and (2.2) dimensionless variables in the following form were used:

$$z' = \frac{z}{L}, \quad t' = \frac{\chi}{L^2} t, \quad D' = \frac{1}{\chi} D, \quad v' = \frac{L}{\chi} v, \quad \beta' = \frac{L^2}{\chi} \beta, \quad \alpha' = \frac{L^2}{\chi} \lambda. \tag{3.10}$$

Changing over to difference equations in the system (2.1) and (2.2), we obtain the algebraic equations

$$a_j^{n+1} = \frac{a_j^n + \tau\beta c_j^{n+1}}{1 + \tau\lambda + \tau\lambda\beta}, \quad j = 0, 1, ..., J, \quad n = 0, 1, ..., N - 1; \tag{3.11}$$

$$\left(\frac{\tau}{h_z} v_{j-1}^{n+1} + \frac{\tau}{2h_z^2} (D_{j-1}^{n+1} + D_j^{n+1}) \right) c_{j-1}^{n+1} - \left(1 + \tau\lambda + \frac{\tau\beta + \tau^2\beta\lambda}{1 + \tau\lambda + \tau\beta\gamma} + \frac{\tau}{2h_z^2} (D_{j-1}^{n+1} + 2D_j^{n+1} + \right.$$

$$\left. + D_{j+1}^{n+1}) \right) c_j^{n+1} + \left(\frac{\tau}{h_z^2} (D_j^{n+1} + D_{j+1}^{n+1}) \right) c_{j+1}^{n+1} = - c_j^n - \frac{\beta\gamma}{1 + \tau\lambda + \tau\beta\gamma} a_j^n \tag{3.12}$$

with the conditions

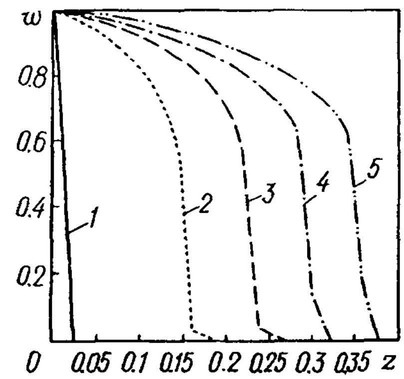

Fig. 1. Moisture distribution in soil at different times: 1) $t = 0$, 2) 2 years, 3) 4 years, 4) 6 years, 5) 8 years. z, m.

$$\overset{0}{c_j} = \overset{n+1}{c_j} = \overset{0}{a_j} = 0\,, \tag{3.13}$$

$$\overset{n+1}{c_0} = \Psi\,(t^{n+1})\,. \tag{3.14}$$

In calculation of the radionuclide concentration, on each time level a factorization method was also used.

Difference schemes (3.8), (3.11), and (3.12) are stable and have a first order of approximation. They were used in developing numerical methods for simulation of moisture and radionuclide transfer in a porous medium.

4. Results of the Numerical Simulation. For preparation of the programs for the aforementioned methods, use was made of the Mathcad 6.6 Plus system.

The computation programs were tested by an analytical solution of simplified problems. It is known [3] that at a constant moisture-transfer rate, the problem on the propagation of a radionuclide can be represented in the form of an analytical formula. For example, if at the initial time the radionuclide concentration is specified as a delta-function, the analytical solution of problem (2.1)-(2.4) has the form [4]

$$G\,(z,\,t) = G_1\,(z,\,t) + G_2\,(z,\,t)\,, \tag{4.1}$$

where

$$G_1\,(z,\,t) = \frac{z}{\sqrt{4\pi D^2 t}} \exp\left\{-\,t\,(\lambda + \beta) - \frac{(z - vt)^2}{4Dt}\right\}, \tag{4.2}$$

$$G_2\,(z,\,t) = \frac{z\beta\sqrt{\gamma}}{\pi D} \exp\left\{-\,t\,(\lambda + \beta\gamma)\right\} \times$$

$$\times \int\limits_0^t \frac{I_1\,[2\beta\sqrt{\gamma\tau\,(t - \tau)}}{\tau\sqrt{t - \tau}} \exp\left\{-\,\frac{(z - v\tau)^2}{4D\tau} - \beta\,(1 - \gamma)\,\tau\right\} d\tau\,. \tag{4.3}$$

Here $I_1(z)$ is a Bessel function of an imaginary argument of the first kind. This solution was used for testing the numerical method used for computation of the propagation of radionuclides. The test shows that the calculations agree well with the analytical solution.

The program used for simulation of moisture transfer in the ground was tested by the known analytical solution of the problem on nonlinear heat conduction [16] obtained in terms of self-similarity variables.

In mathematical simulation of prediction of the propagation of radionuclides in soil, the main difficulty consists in uncertainty of the parameters of the problem, since the models of moisture transfer and propagation of

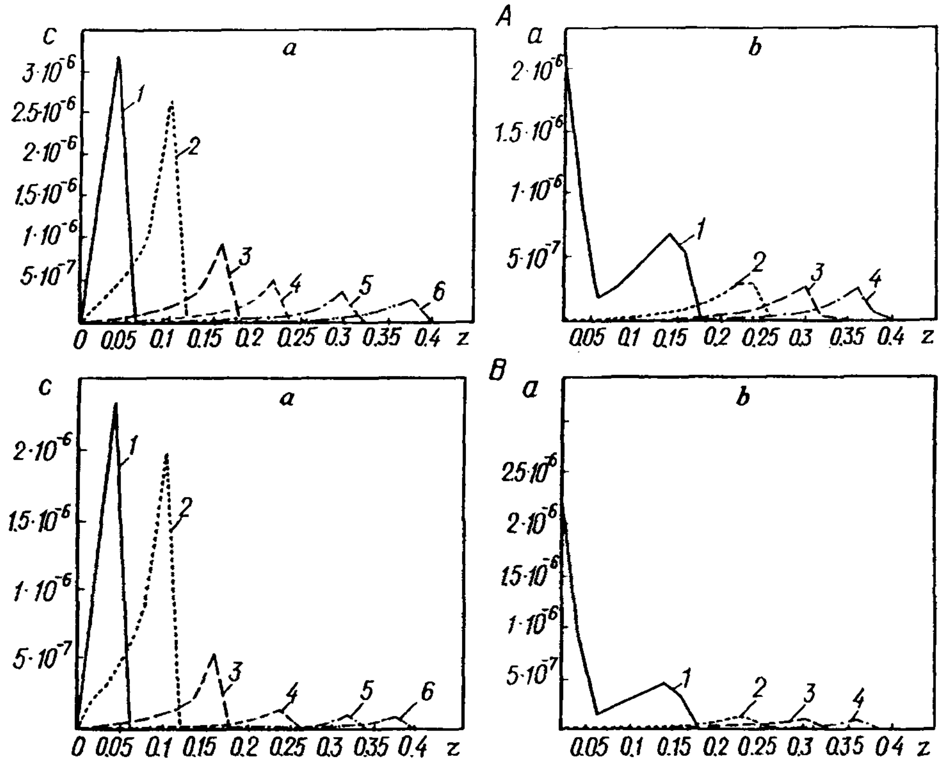

Fig. 2. Concentration of ^{90}Sr (A) and ^{137}Cs (B) in the flow (a) and the amount of adsorbed radionuclide in the medium (b) at different times: a: 1) $t = 0.4$ year, 2) 0.8 year, 3) 2 years, 4) 4 years, 5) 6 years, 6) 8 years; b: 1) $t = 2$ years, 2) 4 years, 3) 6 years, 4) 8 years.

radionuclides given above contain some parameters k, D_0, D_1, β, and γ that can be evaluated experimentally. These data are taken from [17] and some other sources.

In the calculations the following parameters are used: $\sigma = 3.56$; $k = 8.02$ m/yr; $D_1 = 0.034$ m^2/yr; $\beta = 7$ yr^{-1}; $b = 20$ kg/(m·sec^2).

In Fig. 1 one can see results of calculations of free moisture at different times. It is assumed that on the earth's surface the moisture content is maximum and does not change with time. The front of moisture that filters into the soil can be seen in Fig. 1.

Figure 2A illustrates results of calculation of the ^{90}Sr distribution in the flow and in the medium at different times after falling onto the earth's surface. It can be seen from Fig. 2Aa that with time the radionuclide concentration is diffusely smeared in the flow. The total amount of radionuclide is also found to decrease because of radioactive decay. Figure 2Ab shows the adsorbed-radionuclide distribution in the soil at different times. One can see from the figure that in the first stage the total amount of adsorbed ^{90}Sr increases, which can be explained by kinetic adsorption. However, at subsequent moments, because of desorption and radioactive decay, the amount of adsorbed radionuclide decreases. The calculations correspond to experimental data of [18].

In Fig. 2B one can see the ^{137}Cs concentration in the flow and the amount of adsorbed radionuclide in the medium. Results of the computational experiment indicate that the radionuclide concentration in the flow decreases with time, and that in the adsorbed form increases, while the total amount of adsorbed radionuclide increases, starting from a certain moment, which can be explained by radioactive decay of the radionuclide.

The present calculated results allow prediction of the depth of penetration of ^{90}Sr and ^{137}Cs into soil and can be used for predicting possible contamination of ground water with long-lived biologically dangerous radionuclides.

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NOTATION

$c(z, t)$, radionuclide concentration in the flow; $a(z, t)$, amount of radionuclide adsorbed per unit volume of the porous medium; v, flow velocity; β, kinetic adsorption coefficient; λ, decay constant; $D(v)$, longitudinal-diffusion coefficient; γ, inverse of the Henry coefficient; t, time; z, depth; H, head; P_w, capillary pressure; k_w, moisture-transfer coefficient; k, filtration coefficient; ρ, water density; g, acceleration due to gravity; w, moisture content; w^*, maximum moisture content; σ, exponent ranging from 2 to 4; χ, dimensionless parameter.

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