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MODEL OF TRITIUM DISPERSION BY GROUND WATER

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A three-dimensional model of ground-water contamination in the zone of a steady source of tritium is presented. The model is oriented toward long-term modeling of contamination (for up to several decades) on a large area (of up to several hundred square kilometers) where the contaminant arrives through the roof of the aquiferous stratum by infiltration. The three-dimensional equation of convective diffusion is solved numerically by the method of splitting. The convective component is calculated by the method of particles. The dispersion component of the transfer is calculated using the finite-difference method. A transformation of the vertical coordinate is introduced. A solution of the model problem is presented and an interpretation of the results is given.

Introduction. Tritium is among the radionuclides that are present in technological emissions of nuclear power plants and of the designed ITER research fusion reactor. In comparison with other radionuclides, tritium has a small sedimentation coefficient, which leads to long distances over which it propagates in the atmosphere [1]. On reaching the soil surface, due to the activity of soil bacteria tritium is oxidized to the oxide HTO, after which it gets into the ground water along with the infiltrating water. Although the tritium emissions from nuclear power plants and other units that use this element are small, these units have a long life cycle (of several decades) and cause noticeable tritium contamination of the ground water on a large area over the entire time of their operation and even after shutdown. Therefore, long-term prediction of tritium contamination of the ground water on a large area is needed for justifying the environmental safety of both existing and planned plants that use tritium.

To date, the problems of tritium migration in aquiferous strata have been studied insufficiently. In [2], a model of tritium contamination of ground water due to infiltration was proposed. Using a two-dimensional model, Engesgaard et al. studied tritium migration in an aquiferous stratum of length 3 km and height 50 m. In the review [3], only two references to works in which consideration is given to tritium migration over distances of 20 and 32 km are presented. Here, three-dimensional models were not used for interpreting the observations.

Below, a three-dimensional model of ground-water contamination in the zone of a steady source of tritium is given that is oriented toward long-term modeling of contamination (for up to several decades) on a large area (of up to several hundred square kilometers) in which the contaminant arrives at the roof of the aquiferous stratum by infiltration.

Physicomathematical Model and Formulation of the Problem. The convection-dispersion mechanism of HTO propagation in ground water is described by the equation [4]

$$n\frac{\partial C}{\partial t}+\nabla J+W=0\,, \tag{1}$$

where $\nabla J = \text{div}\,[\bar{V}C-\bar{\bar{D}}\,\text{grad}\,C]$.

Let us select a spatial coordinate system so that it coincides with the principal axes of the dispersion tensor. Then, Eq. (1) in coordinate form is

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$$n\frac{\partial C}{\partial t}=\frac{\partial}{\partial x}D_x\frac{\partial C}{\partial x}+\frac{\partial}{\partial y}D_y\frac{\partial C}{\partial y}+\frac{\partial}{\partial z}D_z\frac{\partial C}{\partial z}+$$

$$+\frac{\partial (V_xC)}{\partial x}+\frac{\partial (V_yC)}{\partial y}+\frac{\partial (V_zC)}{\partial z}+W. \qquad (2)$$

The dispersion coefficients of Eq. (2) are written as follows [5]:

$$D_\alpha = D^m + D^g_\alpha . \qquad (3)$$

The coefficient of hydromechanical dispersion can be represented as [5]

$$D^g_\alpha = \delta_\alpha |\bar{V}| , \qquad (4)$$

where $|\bar{V}|$ is the modulus of the mean velocity of the water flow.

The coefficient of molecular diffusion in water is of the order of 10^{-5} $m^2 \cdot day^{-1}$ [4]. Experimental values of the coefficients of hydromechanical dispersion D^g that were determined for a variety of actual aquiferous strata [3] are larger than 10^{-3} $m^2 \cdot day^{-1}$. Therefore, in solving practical problems, we can neglect the term D^m in Eq. (3). Thus, to describe HTO migration in ground water, Eq. (2) must be solved under appropriate initial and boundary conditions.

Numerical Methods of Solution of the Problems. Equation (2) is solved numerically using the method of splitting [6] into three stages [7]. At the first stage, the equation of convective transfer in horizontal planes

$$n\frac{\partial C}{\partial t}=\frac{\partial (V_xC)}{\partial x}+\frac{\partial (V_yC)}{\partial y} \qquad (5)$$

is solved for each z_i, $i = \overline{1, N_z}$. At the second stage, the equation of diffusion (dispersion) scattering in horizontal planes

$$n\frac{\partial C}{\partial t}=\frac{\partial}{\partial x}D_x\frac{\partial C}{\partial x}+\frac{\partial}{\partial y}D_y\frac{\partial C}{\partial y}+W \qquad (6)$$

is solved for each z_i, $i = \overline{1, N_z}$. At the third stage, the equation of convective and diffusive transfer in the vertical direction

$$n\frac{\partial C}{\partial t}=\frac{\partial}{\partial z}D_z\frac{\partial C}{\partial z}+\frac{\partial (V_zC)}{\partial z} \qquad (7)$$

is solved for all (x_i, y_i), $i = \overline{1, N_x}$, $j = \overline{1, N_y}$.

The solution of Eq. (5) by classical schemes of the finite-difference method or the finite-element method entails a nonremovable error, called numerical diffusion, that increases with calculation time [4]. The model that is presented in the current work is intended for long-term prediction. For such calculation times, the numerical diffusion introduces errors that render the modeling results unsuitable for practical use. The numerical diffusion in the solution of Eq. (5) is eliminated using the method of particles in a "PIC" cell [8]. Equations (6) and (7) are solved using the finite-difference method. Allowance for the rate of vertical convective transfer is carried from stage 1 to stage 3, since $V_z h_z/2$, where h_z is the step of the spatial grid along the Z axis, is comparable to and, more frequently, much smaller than D_z. This appreciably decreases the numerical diffusion [4] and at the same time markedly reduces the number of particles [7].

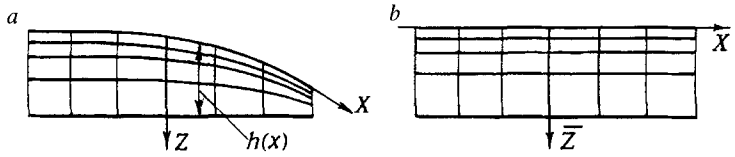

Fig. 1. Calculation grids: a) in the physical region; b) in the calculation region.

The method of particles imposes a constraint on the time step, which must not exceed the time of passage of a particle over one calculation cell [8]. We now compare this step with the time step needed to provide stability of the solution of Eq. (6) by an explicit scheme:

$$\tau_{conv} = \frac{\Delta h}{V}, \tag{8}$$

where V is the water-flow velocity;

$$\tau_{stab} = \frac{\Delta h^2}{D} = \frac{\Delta h}{V}\frac{\Delta h}{\delta}. \tag{9}$$

The model that is described in the current work is intended for calculating tritium migration over large areas, and therefore, the characteristic dimension of a calculation cell in a horizontal plane is not smaller than several hundred meters. From the data of [3] it follows that the dispersion δ for actual aquiferous strata most frequently does not exceed several tens of meters. Thus, the factor $\Delta h/\delta$ on the right-hand side of expression (9) is much larger than unity, and therefore $\tau_{stab} >> \tau_{conv}$. Hence it becomes clear that to solve Eq. (6) it is expedient to use an explicit scheme, which in comparison with an implicit one requires considerably fewer calculations [9].

Tritium contamination of ground water results from infiltration, and therefore, concentration gradients along the Z axis in the initial period of time are large. To preclude substantial computational errors with large gradients, a rather fine-mesh grid along the Z axis must be selected, and therefore, the time step that is used to solve system (5)-(7) is much larger than the stability condition of an explicit scheme for Eq. (7). Hence, it is reasonable to use an implicit scheme to solve Eq. (7).

Aquiferous strata on a large area must be modeled with allowance for variations in the height of the aquiferous stratum, which can range from several tens of meters to several meters. To put it differently, this means that the calculation region in a vertical plane is nonrectangular. For calculations by the finite-difference method, it is brought to rectangular form (see Fig. 1).

The nonrectangular physical region is transformed into rectangular one in the following manner. For each node x_i, $i = 1, N_x$, of the X–Y plane the coordinate Z is transformed by the formula [10]

$$\bar{Z}(x) = \frac{h(x)}{Z_{max}} Z(X), \tag{10}$$

where $Z(X)$ is the vertical coordinate in the physical region.

A partial derivative with respect to Z changes as follows [10]:

$$\frac{\partial}{\partial Z} = \left(\frac{\partial \bar{Z}}{\partial Z}\right)\frac{\partial}{\partial \bar{Z}} = \frac{h(x)}{Z_{max}}\frac{\partial}{\partial \bar{Z}}. \tag{11}$$

The coordinate Z is used only in solving one-dimensional equation (7), and therefore, variables must be changed only in this equation. On substituting in (7) the coordinate Z and the partial derivative with respect to

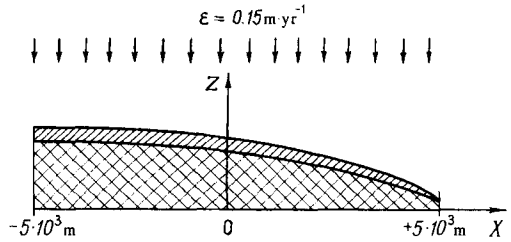

Fig. 2. Diagram of the aquiferous stratum.

Z with the aid of Eqs. (10) and (11), respectively, we obtain a rectangular calculation region and a rectangular grid (see Fig. 1b).

The algorithm of solution of Eq. (2) using the above method lies in the following. Before the first time step, each particle is conferred a value of the activity equal to the activity of the calculation cell in which the particle is located. One time step consists of the following substeps:

1. New coordinates of particles are calculated (Eq. (5)).
2. The collection of activities is calculated anew: the activity of each calculation cell is determined as the arithmetic mean of the activities of the particles that are located in the cell.
3. The diffusion (dispersion) scattering in horizontal planes is calculated (Eq. (6)).
4. The diffusion (dispersion) scattering and the convective transfer in the vertical direction are calculated (Eq. (7)).
5. The activity of each particle that is located in the calculation cell changes by the magnitude of the change in the activity of this calculation cell due to the diffusion (dispersion).

In accordance with the general principle of the method of splitting [6], the same time step is employed in each substep. The initial conditions for each substep are the results of the solution from the preceding substep.

Results of Numerical Modeling. Using the model that is proposed in the current work the problem that was formulated in Scenario 1.3 published by the tritium working group that carries out investigations within the framework of the international scientific program BIOMASS (Biosphere Modeling and Assessment Methods) under the auspices of the International Atomic Energy Agency [11] was solved. This scenario suggests that the specific activity of HTO in ground water 20 years after the start of contamination be determined. The aquiferous stratum is infinite along the Y axis, the left boundary of the stratum X = –5000 m is a water divide, and the right boundary X = +5000 m is a runoff from a height of 1 m. The base of the aquiferous stratum is strictly horizontal. The filtration coefficient is $K = 10^{-4}$ m·sec^{-1} and the porosity is n = 0.4. The infiltration supply ε = 0.150 m·yr^{-1} arrives at the stratum through its roof. Figure 2 schematically illustrates a section of this stratum. At the initial instant of time, the aquiferous stratum is not contaminated by HTO. The HTO flow to the stratum roof is steady and is specified by the following expression:

$$F\,(\text{Bq}\cdot\text{m}^2\cdot\text{yr}^{-1}) = \begin{cases} 2\cdot 10^2, & L \le 0.5 \text{ km}, \\ \dfrac{10^2}{L\,(\text{km})}, & L > 0.5 \text{ km}, \end{cases} \tag{12}$$

where $L = (x^2 + y^2)^{1/2}$. The stratum dispersions are assigned and are equal to δ_x = 50 m, δ_y = 5.0 m, and δ_z = 0.5 m.

Prior to calculating the HTO concentrations from Eq. (2), we must determine the change in the stratum height and filtration rate along the X axis. These parameters can be obtained from the solution of the problem of zero-pressure one-dimensional filtration with constant infiltration. The filtration (mass-balance) equation is of the form [12]

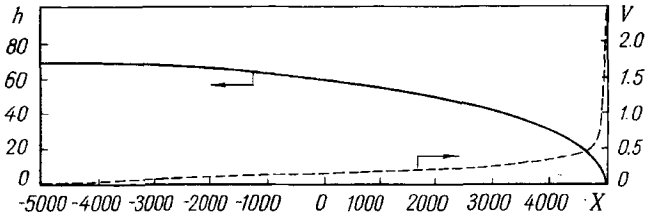

Fig. 3. Height of the aquiferous stratum and filtration rate along the X axis. h, X, m; V, m·day^{-1}.

$$\frac{d}{dx}\left(h\frac{dh}{dx}\right)+\frac{\varepsilon}{K}=0, \tag{13}$$

where h is the hydraulic pressure head, m; ε, K, m·day^{-1}; x, m. The boundary conditions are

$$x=0 \text{ m}: \quad \frac{dh_1}{dx}=0; \tag{14}$$

$$x=+10\,000 \text{ m}: \quad h_2=1 \text{ m}. \tag{15}$$

The filtration rate is determined as

$$V=-K\frac{dh}{dx}. \tag{16}$$

Condition (14) describes the water divide, condition (15) describes the runoff from a height of 1 m, and Eq. (16) is Darcy's law.

The solution of Eq. (13) with boundary conditions (14) and (15) is of the following form:

$$h(x)=\left(-\frac{\varepsilon x^2}{K}+h_2^2+\frac{\varepsilon L^2}{K}\right)^{1/2}, \tag{17}$$

where L = 10,000 m,

$$\frac{dh}{dx}=\frac{1}{2}[h(x)]^{-1/2}\left[-\frac{\varepsilon x}{K}\right]. \tag{18}$$

Using relation (18) it is possible to calculate the field of filtration rates along the X axis from Eq. (16) and the height of the aquiferous layer from relation (17). Figure 3 presents calculation results.

Knowing the height of the aquiferous stratum $h(x)$ and the field of filtration rates, it is possible to proceed to the calculation of the HTO concentration in the aquiferous stratum by the algorithm that was presented in the previous section. The dimension of the calculation region along the Y axis was taken to be (–5000 m, +5000 m). The boundary conditions at the roof of the aquiferous stratum corresponded to expression (12), and the HTO flow at the base was equal to zero. The diffusion and dispersion flows at the lateral surfaces of the aquiferous stratum were taken to be equal to zero, and the convective flows corresponded to the field of filtration rates. This assumption is quite admissible, since, as is shown below, the substance propagation by diffusion and dispersion is not in excess of one step of the calculation grid.

Figures 4 and 5 present modeling results. The first figure shows profiles of specific activities of HTO on the streamlines that pass through the points with the coordinates x = 0, y = 0, z = 0 and 2, 20, 50 m 20 years after the start of contamination, and the second figure presents isolines on the surfaces that are formed

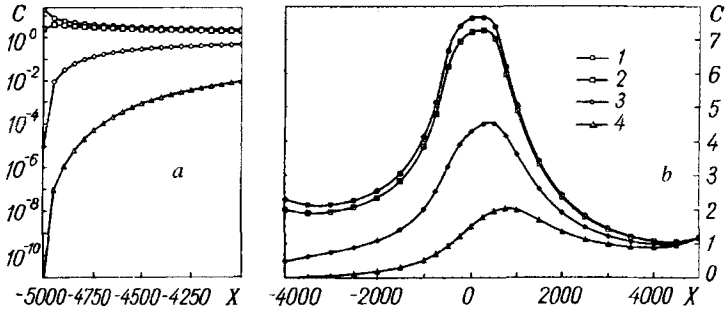

Fig. 4. Specific activity of tritium oxide (HTO) in the ground water on different streamlines 20 years after the start of contamination: a) in the region of a water divide; b) beyond it; 1) Z = 0; 2) 2 m; 3) 20; 4) 50. C, Bq/liter.

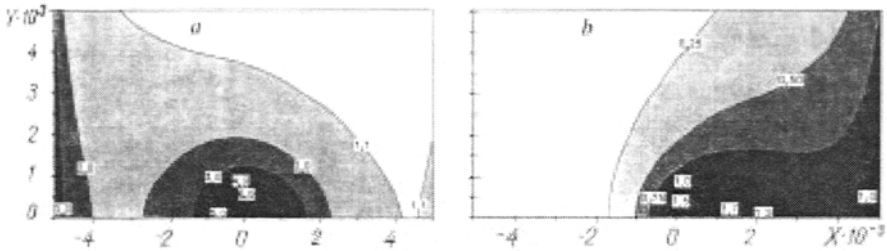

Fig. 5. Isolines of the specific activity of tritium oxide (HTO) in the ground water (Bq/liter) on surfaces parallel to the streamlines [a) z = 2 m; b) 50] 20 years after the start of contamination.

by parallel streamlines passing through the points with the coordinates x = 0, y = 0, and z = 2 m (Fig. 5a) and x = 0, y = 0, and z = 50 m (Fig. 5b).

Discussion of Results. In the region of the water divide (x = −5000 m), the velocity of the water flow is equal to zero and, according to Eqs. (3) and (4), the mechanism of HTO propagation is totally determined by the molecular diffusion. The depth of propagation of the diffusion wave in 20 years is $L_{dif} = \sqrt{D^m \cdot 20 \text{ years}} \approx 1$ m. Hence it is clear that the contamination cannot reach the stratum base in 20 years. This agrees with the modeling results that are presented in Fig. 4a. The filtration rate in the region x = 0 is $V \approx 40$ m·yr^{-1} (see Fig. 3). Let us compare the magnitudes of the dispersion and convective transfer in this region: $L_{disp} = \sqrt{(V\delta_x) \cdot 20 \text{ years}} \approx 40$ m and $L_{conv} = V \cdot 20$ years ≈ 800 m. The comparison indicates that, in the region x = 0, the transfer along the streamlines is totally determined by convection. The data in Figs. 4b and 5b show that the concentration maximum at the stratum base is displaced 800 m downstream due to convection. We next evaluate the depth of propagation of the dispersion wave along the vertical in the region x = 4500–5000 m. For this region, $V \approx 150$ m·yr^{-1} and $L_{disp} = \sqrt{(V \cdot \delta_z) \cdot 20 \text{ years}} \approx 40$ m. This value is larger than the stratum height in this region, and therefore, the HTO concentration along the vertical must be approximately constant. The results that are presented in Fig. 4b are fully consistent with this inference. The slight increase in the concentration in the region x = 4500–5000 m is attributable to the fact that here the stratum height decreases more rapidly than the HTO flow to the stratum roof.

Conclusion. To solve the equation of convective diffusion, a method is proposed that consists in the following:

1) the convective transfer is calculated using the method of particles;

2) the diffusion (dispersion) scattering in horizontal planes is calculated using an explicit finite-difference scheme;

3) the diffusion (dispersion) scattering and the convective transfer are calculated using an implicit finite-difference scheme;

4) the transformation of coordinates along the vertical axis Z is carried out.

The method proposed permits prediction of tritium contamination of ground water on large territories (of up to several hundred square kilometers) over a long time (of up to several decades). The model can be used as part of a model for prediction of the consequences of environmental contamination by tritium in the zone of a tritium plant.

The solution of the model problem [11] has some special features, namely, a steep concentration profile in the region of $x = -5000$ m, a displacement of the maximum at the stratum base at the center of the calculation region, and a concentration increase in the region $x = 4500$–5000 m. The presence of the above features in the solution permits the use of this problem for testing other programs that model ground-water contamination due to infiltration.

NOTATION

n, porosity; C, activity of tritium oxide (HTO) in ground water; t, time; J, mass flux; $\bar{V}$, vector of the mean filtration rate; $\bar{\bar{D}}$, dispersion tensor; W, rate of β-disintegration of tritium; D_x, D_y, and D_z, coefficients of dispersion along the X, Y, and Z axes, respectively; D^m, coefficient of molecular diffusion; D^g_α, coefficient of hydromechanical dispersion along the coordinate axis α; $\alpha = \{X, Y, Z\}$, coordinate axes; δ_α, dispersion along the coordinate axis α; N_x, N_y, and N_z, number of nodes along the X, Y, and Z axes, respectively; τ_{conv}, time step needed to provide stability of the method of particles; Δh, grid step in the horizontal plane; τ_{stab}, time step needed to provide stability of the solution by the explicit scheme; $Z(x)$, vertical coordinate in the calculation region; $h(x)$, height of the aquiferous stratum at the point with the coordinate x; h_1, hydraulic pressure head at the left boundary ($x = 0$); h_2, same at the right boundary ($x = 10{,}000$ m); Z_{max}, maximum height of the aquiferous stratum; K, filtration coefficient of the aquiferous stratum; ε, infiltration to the aquiferous stratum; F, HTO flow to the stratum roof; L_{dif}, depth of contaminant propagation by diffusion; L_{disp}, same by dispersion; L_{conv}, same by convection.

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