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MODEL OF ANALYSIS OF THE GROUNDWATER-PROTECTION LEVEL IN SHALLOW LAND BURIAL OF RADIOACTIVE WASTE

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A mathematical model of transfer of radionuclides in the aeration zone has been considered. The analytical solution for this model in a generalized form suitable for practical application has been obtained. A procedure enabling one to determine the optimum size of the protection zone and in which the maximum concentration of radionuclides does not exceed the maximum permissible levels for a specific kind of radionuclides has been proposed.

The possibility of contaminating groundwater from the earth's surface is largely determined by the protection level of water-bearing horizons. The latter depends on many factors, which may be subdivided into two groups: natural factors and anthropogenic ones. The first group includes the depth to the level of groundwater, the presence in sections and thicknesses of low-permeability rocks, the lithology and sorption properties of rocks, and the relation of the levels of the investigated and overlying water-bearing horizons. Among the second group are the existence conditions of contaminants on the earth's surface and, accordingly, the character of their penetration into the groundwater, the chemical composition of contaminants, and, as a consequence, their migration capacity, the sorptive power, the chemical resistance, the decay time, and the character of interaction with rocks and groundwater.

The groundwater-protection level may be characterized qualitatively and quantitatively. In the first case, one primarily considers only natural factors, whereas in the second case one considers both natural and anthropogenic factors. A detailed assessment of the groundwater-protection level with allowance for the distinctive feature of moisture transfer in the aeration zone and for the character of interaction of the contaminant with rocks and groundwater requires, as a rule, that a hydrogeochemical model of the processes of penetration of the contaminant into the water-bearing horizon be created. Qualitative assessment may be carried out in the form of the sum of arbitrary points or based on assessment of the time in which the water filtered from the surface reaches the water-bearing horizon (the distinctive features of moisture transfer in the aeration zone and the process of interaction of the contaminant with rocks and groundwater are disregarded). The point assessment of the groundwater-protection level has been developed in detail by Gol'dberg [1]. The sum of points, which depends on the occurrence conditions of groundwater and the thicknesses of low-permeability deposits and their lithologic composition, determines the degree of protection of groundwater. As far as the lithology and filtration properties of low-permeability deposits are concerned, one recognizes three groups — sand loams, light loams (k = 0.1–0.01 m/days), and heavy loams and clays ($k < 0.001$ m/days) — and an intermediate group: a mixture of rocks of different groups (k = 0.1–0.001 m/days).

The degree of protection of groundwater may be determined directly from the time of filtration of the contaminated water from the earth's surface to the water-bearing horizon. For the conditions of a homogeneous section of the aeration zone and a constant level in the storage, this time may be determined [2] as

$$t = \left(\frac{nH}{k}\right)\left[\frac{m}{H} - \ln\left(1 + \frac{m}{H}\right)\right], \tag{1}$$

and, in the presence of a protective shield of low-permeability rocks at the storage base, as

$$t_1 = \left(\frac{nH}{k}\right)\left[\frac{m}{H}\left(1 - \frac{m_1}{H}\right)\left(\frac{k}{k_1} - 1\right)\ln\left(1 + \frac{m}{H + m_1}\right)\right]. \tag{2}$$

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The basic infiltration parameters are characterized within the framework of filtration theory [3], in accordance with which the velocity of motion of moisture in unsaturated grounds is described by Darcy's law. Under the assumption that water seeps into the ground under a constant head H, for the filtration rate we write

$$V = k \frac{H + m}{m}, \tag{3}$$

but if there is a protective barrier of thickness m_1, we write

$$V = \frac{H + m}{\dfrac{m_1}{k_1} + \dfrac{m - m_1}{k}}. \tag{4}$$

To assess safety in shallow land burial of radioactive waste one often assumes that the individual effective equivalent irradiation dose is received when contaminated water is drunk. In this case, it becomes urgent to predict the state of groundwater in the zone of influence of storages for the entire period of their potential hazard. The concentrations and fluxes of radionuclides in aqueous media are used as safety indicators. In turn, the problem of searching for the zone of influence is reduced to determination of the size of protective barriers at exit from which the radionuclide concentration in water is lower than the Republic permissible levels (RPLs) or is equal to them.

Study of the processes and parameters of infiltration and mass transfer in the aeration zone is based on the laws of the theory of infiltration and migration of a radioactive impurity; this theory gives a physicomathematical description of different mechanisms of complex processes of a hydrodynamic and physicochemical character.

To describe the processes of migration of radionuclides in a porous medium we selected a convective-diffusion model developed for the liquid and solid phases of the rock skeleton [4], in accordance with which the concentration of the radioactive impurity is described by the equation

$$S = \frac{1}{2} \exp(-\beta\tau) \exp\left[-\left(\frac{u - \tau}{2\sqrt{\tau}}\right)^2\right] \left[\operatorname{erfcx}\left(\frac{u - \tau}{2\sqrt{\tau}}\right) + \operatorname{erfcx}\left(\frac{u + \tau}{2\sqrt{\tau}}\right)\right], \tag{5}$$

and at $\tau > u$ it takes the form

$$S = \frac{1}{2} \exp(-\beta\tau) \left\{2 - \exp\left[-\left(\frac{u - \tau}{2\sqrt{\tau}}\right)^2\right] \left[\operatorname{erfcx}\left(-\frac{u - \tau}{2\sqrt{\tau}}\right) - \operatorname{erfcx}\left(\frac{u + \tau}{2\sqrt{\tau}}\right)\right]\right\}, \tag{6}$$

where erfcx (x) = exp (x^2) erfc (x) is the scaled residual error function.

For the prescribed distance from the contamination source, the time of attainment of the maximum concentration is determined from the condition

$$-\beta S + \frac{1}{2\sqrt{\pi}} \frac{u}{\tau^{3/2}} \exp(-\beta\tau) \exp\left[-\frac{(u - \tau)^2}{4\tau}\right] = 0. \tag{7}$$

Thus, solving the transcendental equation (7) (S values are computed from (6) for τ with a fixed β and different u), we determine the distance from the contamination source where the maximum concentration in an aqueous solution corresponds to the RPL for this radionuclide.

For long-lived radionuclides such as ^{137}Cs, ^{90}Sr, ^{241}Pu, and ^{241}Am, prediction assessments must be considered during a time interval (of the order of hundreds and thousands of years) in which the basic parameters of the model may substantially change. Based on the literature and experimental data, we have analyzed a correct selection of the necessary parameters of the model for the territories of the Gomel Region where 85 burial sites of radioactive deactivation waste (BSDWs) are located.

Natural factors related to the movement of intrasoil moisture — filtration flow, capillary flow, and thermal and moisture transfer — may be considered as general ones. The difference between the kinds of flow lies only in the

TABLE 1. Properties of the Aeration-Zone Grounds

Ground	ρ, g/cm^3	θ	n	Natural humidity of the ground
Aeration zone	1.72	0.537	0.348	0.075
Deactivation waste	1.35	0.484	0.321	0.131

forces generating the flow and in the direction of motion of moisture. Without taking into account the mechanisms causing the moisture to move in the soil, we will assume that the three kinds of soil-moisture flow may be replaced by the average total equivalent flow. Its direction and value depend on the season, the time of day, weather conditions, and the properties of the soil. A time-variable total flow is inconvenient for consideration for many reasons, primarily in connection with the great difficulties of its measurement. Therefore, in considering long time intervals (for example, those measured in years), there is sense in considering the flow to be, on the average, constant during a season, a year, or a few years.

The structure of the balance of groundwater and surface water under natural conditions is of particular importance in determining the filtration rate. The regional structure of motion, the balance, and the resources of the groundwater of Gomel Poles'e have been formed by the method of mathematical modeling [5].

The model has been tested by matching the model and actual values of the heads of the groundwater and the model and actual moduli of ground flow into rivers [6]. According to the data on the structure of the groundwater balance, the rate of infiltration feed is taken to be 0.89 m/year in this area.

Data on the density and porosity of 222 ground samples taken in 48 states of the USA have been given in [7]. It has been shown that the experimental data on the density, total porosity, porosity in the natural state, and humidity of the grounds obey a lognormal distribution law. In creating a system of observation of the migration of radionuclides from the storage sites of radioactive waste to the groundwater [8], samples of the ground in the aeration zone were taken at the sites of drilling of hydrological wells. Statistical processing of the experimental data shows that, for the total porosity, porosity in the natural state, and humidity of the aeration-zone grounds, they also obey a lognormal distribution law. The results of the statistical processing are presented in Table 1.

Between the total porosity and the density of the ground skeleton, there is the simple relation

$$\theta = \left(1 - \frac{\rho}{2.65}\right). \tag{8}$$

According to the experimental data, the humidity in the natural state is equal to 0.1–0.15 in sand loams and to 0.01–0.1 in loams.

The process of mixing of the liquids is quite satisfactorily described by the diffusion equation with a convective term [9]; the total diffusion coefficient for both gases and liquids is determined by an expression of the kind

$$D = \frac{nD_{\text{v}}}{\psi} + \frac{\gamma V}{n}. \tag{9}$$

On the average, we have $\gamma = 0.1$ cm for comparatively fine-grained sands [9]. According to the experimental data of [10], ψ for uncemented rocks is within 1.4–1.7, and for cemented and cracked rocks we have $\psi = 2.0$–4.0. The value of γ varies from 0.08 to 0.16 m.

The coefficients γ and ψ have been experimentally determined under natural conditions in [11] by using ^{3}H and ^{3}He as tracers. Values of $\psi = 1.73 \pm 0.56$ and $\gamma = 0.07 \pm 0.05$ have been obtained, which is in good agreement with the data enumerated above.

To select the molecular diffusion coefficient in an aqueous solution we analyzed the experimental data of [12]. A linear relationship between the diffusion coefficients of the cations D_{v} and the limiting equivalent electrical conductivity ε in aqueous solutions at 25°C has been found:

$$D_{\text{v}} = A\varepsilon, \tag{10}$$

TABLE 2. Recommended Values of K_d, cm^3/g

Radionuclide	Sands	Loams	Clays	Organics	Soils [17]	Soils [18]
^{137}Cs	280	4600	1900	270	1100	1000
^{90}Sr	15	20	110	150	27	—
^{241}Am	1900	9600	8400	11200	810	—
Pu	550	1200	5100	1900	1800	5000

here $A = 0.02668$ for monovalent cations, 0.01332 for bivalent cations, and 0.008901 for trivalent cations respectively. The quantity A is easily obtained from the expression $A = A_{\text{mon}}/m$.

If ε in aqueous solutions at 25°C for the cations is unknown, we may use its value of $0.77 \cdot 10^{-9}$ m^2/sec for significant bivalent cations and of $0.58 \cdot 10^{-9}$ m^2/sec for the trivalent ones.

From the analysis of [13], it follows that the diffusion coefficients in the absorbed phase are two orders of magnitude smaller than those in the free solution.

The distribution constant K_d is an empirical quantity. It depends on physicochemical processes occurring in the liquid and solid phases, the physicochemical properties of these phases, and experimental conditions. As follows from numerous experimental data, the values of K_d differ by several orders of magnitude, even for identical physicochemical properties of the grounds studied [14, 15]. Therefore, researchers were facing the problem of systematizing the experimental data obtained and providing recommendations for using them in prediction assessments of the migration of radionuclides.

An attempt at such an analysis has been made in [16]. Sheppard and Thibault used the data on K_d, published in the literature. After their classification by the types of ground, the data were statistically processed for each group of grounds. A lognormal distribution law was used. The recommended values of K_d as compared to [17, 18] for long-lived isotopes ^{137}Cs, ^{90}Sr, ^{241}Am, and Pu are presented in Table 2.

In the Regulations of the Washington Health Department [19], the following values of K_d have been recommended: 50 for ^{137}Cs, 25 for ^{90}Sr, and 200 cm^3/g for ^{241}Am and Pu. Using the data of [20, 21] and applying the lognormal distribution law, we obtained the geometric mean values of K_d for ^{137}Cs (162 cm^3/g) and for ^{90}Sr (5 cm^3/g).

From an analysis of the literature and experimental results, we take the following values for the computational model: $V = 0.89$ m/year, $\rho = 1.72$ g/cm^3, $n = 0.2$, $\gamma = 0.001$ m, $\psi = 1.73$, $D_v = 2.06$ m^2/year and $K_d = 200$ cm^3/g for ^{137}Cs, and $D_v = 0.79$ m^2/year and $K_d = 10$ cm^3/g for ^{90}Sr. According to these data, we take $D_{\text{eff}} = 1.16$ m^2/year, $R = 1721$, $\lambda = 0.023$ 1/year, and $\beta = 2.303$ for ^{137}Cs and $D_{\text{eff}} = 0.04$ m^2/year, $R = 87$, $\lambda = 0.0241$ 1/year, and $\beta = 0.004243$ for ^{90}Sr.

Transition from $u_{\max}$ to the coordinate specifying the size of the protective barrier is determined by the expression

$$z_{\max} = \frac{u_{\max} n D_{\text{eff}}}{V}, \tag{11}$$

and conversion to the time of attainment of the maximum concentration levels is written as

$$t_{\max} = \frac{\tau_{\max}\beta}{\lambda}. \tag{12}$$

The concentration for which we determine $z_{\max}$ is found from the formula

$$C_{\max} = \frac{S_{\max} N}{K_{d,w}}. \tag{13}$$

The program of computations was realized in the Microsoft Excel environment. A MATLAB mathematical system is used for obtaining the solution in a graphic version and for numerous calculations. The results obtained according to the model proposed are presented in Table 3. Using the data of the table, we may solve the problem on

TABLE 3. Coordinates and Time of Attainment of the Maximum Concentrations for ^{137}Cs and ^{90}Sr

^{137}Cs			^{90}Sr		
Protective barrier, m	$t_{\max}$, year	$S_{\max}$	Protective barrier, m	$t_{\max}$, year	$S_{\max}$
1.0	130.6	$4.58 \cdot 10^{-3}$	1	24.69	$5.31 \cdot 10^{-1}$
1.5	191.5	$4.73 \cdot 10^{-4}$	2	45.58	$3.17 \cdot 10^{-1}$
2.0	252.1	$5.10 \cdot 10^{-5}$	3	66.02	$1.91 \cdot 10^{-1}$
2.5	321.5	$5.63 \cdot 10^{-6}$	4	86.24	$1.16 \cdot 10^{-1}$
3.0	372.9	$6.33 \cdot 10^{-7}$	5	106.32	$7.09 \cdot 10^{-2}$
3.5	433.2	$7.18 \cdot 10^{-8}$	6	126.32	$4.34 \cdot 10^{-2}$
4.0	493.4	$8.23 \cdot 10^{-9}$	7	146.24	$2.66 \cdot 10^{-2}$
4.5	553.6	$9.49 \cdot 10^{-10}$	8	166.11	$1.63 \cdot 10^{-2}$
5.0	613.7	$1.10 \cdot 10^{-10}$	9	185.93	$1.01 \cdot 10^{-2}$
5.5	674.2	$1.28 \cdot 10^{-11}$	10	205.73	$6.19 \cdot 10^{-3}$
6.0	734.0	$1.50 \cdot 10^{-12}$	15	304.31	$5.55 \cdot 10^{-4}$
6.5	794.6	$1.76 \cdot 10^{-13}$	20	402.51	$5.03 \cdot 10^{-5}$

the size of a protective barrier for which the radionuclide concentration ($S_{\text{m.p.c}}$) in groundwater will be lower than RPLs-99 or equal to them.

The size of the protective barrier of the aeration zone and the time of attainment of the maximum concentrations may be approximated by the following simple dependences:

for ^{137}Cs

$$z = 0.00002x^3 + 0.0011x^2 - 0.2093x - 0.1562\,, \tag{14}$$

$$t_{\max} = 0.032x^3 - 0.47x^2 + 122.43x + 8.5\,, \tag{15}$$

for ^{90}Sr

$$z = 0.0002x^4 + 0.00432x^3 + 0.0423x^2 - 1.8825x - 0.2084\,, \tag{16}$$

$$t_{\max} = 0.0017x^3 - 0.0751x^2 + 20.731x + 4.279\,, \tag{17}$$

where $x = \ln(S_{\max})$.

According to RPLs-99, the limiting concentration in water is 10 and 0.37 Bq/liter for ^{137}Cs and ^{90}Sr respectively.

If we take the distribution coefficients in the deactivation waste to be 500 cm^3/g for ^{137}Cs and 20 cm^3/g for ^{90}Sr, from (13) we have $S_{\max} = 5000/N$ for ^{137}Cs and $S_{\max} = 7.4/N$ for ^{90}Sr.

The maximum concentrations in the deactivation waste located in the Gomel Region area do not exceed 30,000 Bq/kg for ^{137}Cs and are within 20–1300 Bq/kg for ^{90}Sr. Then we have $S_{\max} = 0.17$ for ^{137}Cs, and $S_{\max}$ for ^{90}Sr varies within 0.37–0.006.

A protective barrier of the aeration zone of thickness 0.4 m is sufficient for penetration of ^{137}Cs from the BSDWs into the ground water. The protective barrier of the aeration zone for ^{90}Sr must have a thickness of 2–10 m. Thus, within 40–300 years, the BSDWs of the Gomel Region will be a serious hazard as far as the arrival of ^{90}Sr at the groundwater is concerned.

The model proposed enables one to determine, from the adopted initial data, the size of the protective barriers of aeration zones, which ensure the protection of groundwater against radionuclide contamination. This procedure may be used in the process of designing shallow-land sites for burial of radioactive waste and of selecting the optimum protective barriers for different kinds of radionuclides to be buried.

NOTATION

C, specific activity of the radionuclide in the liquid phase, Bq/liter; C_0, initial specific activity of the radionuclide in the liquid phase, Bq/liter; D, coefficient of dispersion in porous media, m^2/year; D_m, coefficient of molecular diffusion in the rock skeleton, m^2/year; D_v, coefficient of molecular diffusion in the free volume, m^2/year; $D_{eff} = D + \frac{\rho K_d D_m}{n\psi}$, effective diffusion coefficient, m^2/year; H, moisture head, m; k, filtration coefficient, m/year; K_d, distribution coefficient of a water-soluble compound, cm^3/kg; $K_{d,w}$, distribution coefficient in radioactive waste, cm^3/kg; m, thickness of the barrier (m), valence of a cation; n, active porosity of the rock skeleton, m^3/m^3; N, activity of the radionuclide in the solid phase, Bq/kg; $R = \left(1 + \frac{\rho K_d}{n}\right)$, retrogression coefficient; $S = C/C_0$, dimensionless coefficient; t, time; $u = zV/nD_{eff}$, dimensionless coordinate; V, flow velocity, m/year; x, parameters of the functions; z, coordinate in the direction of motion of the liquid; $\beta = \lambda n^2 n D_{eff}/V^2$, dimensionless decay constant of the radionuclide; γ, hydrodispersion coefficient, m; ε, electrical conductivity, m^2/(Ω·mole); θ, porosity; λ, decay constant of the radionuclide, 1/year; ρ, density of the rock skeleton, kg/cm^3; $\tau = tV^2/Rn^2D_{eff}$, dimensionless time; ψ, crookedness coefficient characterizing the inhomogeneity of a porous medium. Subscripts: d, distribution; eff, effective; m, molecular; 0, initial; 1, clay barrier; mon, monovalent; v, free volume; w, radioactive waste; p, piston-type; p.s, piston-type with allowance for sorption; max, maximum; m.p.c, maximum permissible concentration.

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