PROBABILITY ESTIMATION OF THE DISTRIBUTION OF EXPECTED RESULTS FOR PREDICTION MODELS OF RADIONUCLIDE MIGRATION IN POROUS MEDIA

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A method based on the statistical analysis of estimates of the uncertainty of the input parameters of a prediction model is proposed. Distributions of expected data on the time of attainment of maximum concentrations of radionuclides were obtained by the Monte Carlo method for a concrete model of radionuclide migration in porous media. It is shown that, for this model, the logarithm of maximum concentrations of radionuclides and the time of their attainment obey the uniform-distribution law.

At present, much thought is being given to the analysis of errors in measuring and uncertainty in estimating the safety of places of burial of radioactive wastes (PBRW). We take the word uncertainty to mean both the scatter of calculation data and the lack of understanding of possible processes and parameters. The indicated uncertainty can be analyzed qualitatively and quantitatively. In the case where the safety of a PBRW is analyzed qualitatively, it is necessary to substantiate the model being used and the parameters being selected, consider an alternative model and possible parameters, and, in certain cases, perform an alternative estimation of the safety of the PBRW. A quantitative analysis of this uncertainty represents a probability prediction, in which random variables are used as model parameters and possible distributions of these parameters are prescribed. In this case, a random sampling of parameters is performed by the Monte Carlo method or the Latin-hypercube method and possible concentrations of radionuclides are calculated for simple models. Complex models are not used as a rule because, in this case, it is necessary to perform calculations for a large number of different variants and these calculations give probability distributions of radionuclide concentrations. An example of such investigations is presented in [1].

According to the IAEA recommendations [2], the uncertainties in estimating the safety of a PBRW can be divided into two types: the uncertainties of the first type are associated with the errors in determining the parameters of radionuclides and their distributions, and the uncertainties of the second type are unknown parameters. In the latter case, the parameters of radionuclides and their possible distributions are taken from literature sources or are selected for other reasons.

The above-described method of determining the uncertainties in estimating the safety of a PBRW makes it possible to obtain quantitative estimates of this safety, which is of great importance for making substantiated decisions on the PBRW. In accordance with this method, the safety of a PBRW is estimated by the following scheme. With the use of a model defining an actual PBRW most adequately, the most probable distribution of the activity of radionuclides in the environment is calculated. For the same model, the probability distributions of radionuclide concentrations are calculated using a model for analysis of uncertainties. In this way, the probable properties of engineering barriers and of a radioactive source can be estimated.

The migration of radionuclides in a porous medium was defined using a convective-diffusion model for the liquid and solid phases of the skeleton of a rock [3]. According to this model, the concentration of a radioactive impurity is determined as

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

At $\tau > u$, Eq. (1) takes the form

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

where $\mathrm{erfcx} = \exp(-x^2) \, \mathrm{erfc}(x)$.

For a given distance from the radiocontamination source, the time of attainment of a maximum concentration of radionuclides is determined from the condition $\frac{\partial S}{\partial \tau} = 0$. Then, taking into account (1), we obtain

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

The solutions of the problem being considered, obtained for definite parameters, determine the zone of influence of a radioactive source. This zone is determined by the distance from the radioactive source at which the maximum concentration of radionuclides in an aqueous solution corresponds to the Republican Admissible Levels for these radionuclides. Thus, the problem is reduced to the solution of the transcendental equation (3) (the values of S are calculated by (1) with respect to τ at a definite value of β and different values of u).

The above-described model can be used for defining the processes occurring in the environment. To perform calculations with this model, it is necessary to know a large number of parameters characterizing the environment. Every so often it is impossible to obtain complete information on these parameters and the parameters themselves can change in wide ranges depending on the state of a natural object. For the model being considered, such parameters are ρ, K_d, n, D_{eff}, and V. The coefficient of radioactive decay λ of radionuclides is assumed to be constant.

In the case where the probability approach is used, each parameter appearing in formula (3) is assumed to be a random variable and the quantities $S_{\max}$ and $\tau_{\max}$ are considered as functions of these variables. The main difference of the probability model from the determinate model is that the determinate model makes it possible to determine unique values of $S_{\max}$ and $\tau_{\max}$ and the probability model gives a variety of possible values of these quantities.

Varying the initial data in the determinate model, one can obtain an interval of $S_{\max}$ and $\tau_{\max}$ values; however, it is impossible to determine the probability that "actual" values of $S_{\max}$ and $\tau_{\max}$ will fall in any interval. The probability model allows one not only to obtain an interval of $S_{\max}$ and $\tau_{\max}$ values, but also to determine the probability that the "actual" values of $S_{\max}$ and $\tau_{\max}$ will fall in a definite interval. The most popular approach is the approach in which the less probable, the most probable, and the maximum possible values of parameters, the characteristics of these parameters, and their probability distributions are determined on the basis of available a priori information and experience. The probability characteristics obtained in this way are usually called the "subjective" characteristics since they reflect the experience and preferences of an expert.

The problem being considered is usually solved using normal, log-normal, uniform, and triangular distributions of parameters as well as the β-distribution with form parameters admitting the existence of the most probable value of a parameter. When information making possible the estimation of the chances of obtaining any value of a parameter from a given interval of its values is absent, a uniform distribution is used.

To determine the response of a model to the variations in its input parameters, it is necessary to analyze its sensitivity. For the model being considered, the influence of the change in its input parameters on the values of $S_{\max}$ and $\tau_{\max}$ is illustrated in Table 1. It is seen that this model is less sensitive to the changes in the parameters D_{eff} and n and is most sensitive to the changes in the parameters V, ρ, and K_d. In the case where the errors in the initial data are known, the standard uncertainty $\Delta S_{\max}$ is calculated by the formula

$$\Delta S_{\max} = \frac{dS}{dX_j} \Delta X_j .$$ (4)

For the model selected, the influence of the errors in the initial data on the calculations of $S_{\max}$ can be determined by the following formulas:

TABLE 1. Change in the Input Parameters of a Model in the process of Calculating S_{max} and τ_{max}

Change in parameters	Input parameters				
	ρ	K_d	n	D_{eff}	V
S_{max}					
−40	0.0087	0.0087	0.0016	0.0016	0.00012
−30	0.0062	0.0062	0.0016	0.0015	0.00022
−20	0.0042	0.0042	0.0016	0.0015	0.00041
−10	0.0026	0.0026	0.0015	0.0015	0.00078
0	0.0015	0.0015	0.0015	0.0015	0.0015
10	0.00081	0.00081	0.0014	0.00147	0.0027
20	0.00044	0.00044	0.0014	0.00145	0.0044
30	0.00024	0.00024	0.00136	0.00144	0.0067
40	0.00013	0.00013	0.0013	0.0014	0.0096
τ_{max}					
−40	192.44	192.44	261.08	263.10	363.10
−30	206.19	206.19	261.58	263.22	338.67
−20	222.19	222.19	262.15	263.33	313.92
−10	241.02	241.02	262.79	263.43	288.87
0	263.52	263.52	263.52	263.52	263.52
10	288.16	288.16	264.29	263.58	240.24
20	312.69	312.69	265.02	263.61	220.68
30	337.11	337.11	265.72	263.62	204.02
40	361.43	361.43	266.39	263.59	189.66

$$\frac{\Delta S_{max}}{S_{max}} = -\beta\tau_{max}\left(\frac{R-1}{R}\right)\frac{\Delta\rho}{\rho},$$

$$\frac{\Delta S_{max}}{S_{max}} = -\beta\tau_{max}\left(\frac{R-1}{R}\right)\frac{\Delta K_d}{K_d},$$

$$\frac{\Delta S_{max}}{S_{max}} = -\beta\tau_{max}\left[\frac{R+1}{R} - \left(2 - \sqrt{\pi\tau_{max}}\,\operatorname{erfcx}\left(\frac{(u+\tau_{max})^2}{4\tau_{max}}\right)\right)\right]\frac{\Delta n}{n},$$

$$\frac{\Delta S_{max}}{S_{max}} = \beta\tau_{max}\,\sqrt{\pi\tau_{max}}\,\operatorname{erfcx}\left(\frac{(u+\tau_{max})^2}{4\tau_{max}}\right)\frac{\Delta V}{V},$$

$$\frac{\Delta S_{max}}{S_{max}} = \beta\tau_{max}\left[1 - \sqrt{\pi\tau_{max}}\,\operatorname{erfcx}\left(\frac{(u+\tau_{max})^2}{4\tau_{max}}\right)\right]\frac{\Delta D_{eff}}{D_{eff}}.$$

The simulation of the processes being considered by the Monte Carlo method allows one to construct a mathematical model for S_{max} and τ_{max} with indeterminate parameters. Knowing the probability distributions of the model parameters and the relations between them, we can determine the distributions of S_{max} and τ_{max}. The Monte Carlo method is realized with the use of special mathematical packages (e.g., Microsoft Excel, Matlab, Statistica, and others packages). The indicated simulation represents a series of numerical experiments making possible the obtaining of empirical estimates of the influence of different initial quantities on the results dependent on them.

In the general case, a simulation experiment includes the following stages:

1) the relations between the input and output parameters of a model are represented in the form of mathematical equations or inequalities;

TABLE 2. Comparison of the Main Parameters of a Model Determined Experimentally (Numerator) and the Corresponding Theoretical Distributions (Denominator) for $\ln S_{max}$ and τ_{max}

Distance, m	Average value		Dispersion		Asymmetry		Excess	
	$\ln S_{max}$	τ_{max}	$\ln S_{max}$	τ_{max}	$\ln S_{max}^{*)}$	$\tau_{max}^{*)}$	$\ln S_{max}^{**)}$	$\tau_{max}^{**)}$
10	$\dfrac{-1.6243}{-1.6244}$	$\dfrac{63.98}{63.97}$	$\dfrac{0.00710}{0.00707}$	$\dfrac{10.01}{9.97}$	0.00364	−0.0097	−1.2076	−1.2075
30	$\dfrac{-4.4083}{-4.4086}$	$\dfrac{176.97}{176.95}$	$\dfrac{0.05041}{0.05023}$	$\dfrac{75.77}{75.51}$	0.00399	−0.0086	−1.2075	−1.2075
50	$\dfrac{-7.1330}{-7.1335}$	$\dfrac{288.13}{288.10}$	$\dfrac{0.13065}{0.13020}$	$\dfrac{200.58}{199.90}$	0.00403	−0.0082	−1.2075	−1.2075
70	$\dfrac{-9.8349}{-9.8356}$	$\dfrac{398.61}{398.58}$	$\dfrac{0.24717}{0.24633}$	$\dfrac{383.95}{382.59}$	0.00402	−0.0079	−1.2075	−1.2075
80	$\dfrac{-11.1807}{-11.1815}$	$\dfrac{453.71}{453.68}$	$\dfrac{0.31893}{0.31785}$	$\dfrac{497.55}{495.98}$	0.00401	−0.0074	−1.2075	−1.2075

*) Theoretical distribution of $\ln S_{max} = 0$ and $\tau_{max} = 0$.

**) Theoretical distribution of $\ln S_{max} = -1.2$ and $\tau_{max} = -1.2$.

2) the probability distributions of the key parameters of the model are determined;

3) the computer simulation of the key parameters of the model is performed;

4) the main characteristics of the distributions of the input and output parameters are calculated;

5) the results obtained are analyzed and a decision is made on the basis of this analysis.

The results of the above-described experiment can be supplemented with statistical-analysis data used for construction of prediction models. This complex analysis allows one to obtain the probability distribution of possible values of S_{max} and τ_{max}.

In the simulation method, the first step is determination of the distribution function of each variable influencing the values of S_{max} and τ_{max}. Let us consider the case where the uncertainty in determining the input parameters of the model is maximum. In this case, it is practically impossible to determine the most probable values of parameters from the prescribed interval of their minimum and maximum values, and the uniform distribution is used as the approximate distribution. By way of example, we will simulate the migration of the ^{90}Sr radionuclide in a system with the following parameters: ρ = 1.75–2.1 kg/cm^3, K_d = 7–8.4, n = 0.35–0.42, V = 2.5–3 m/g, and D_{eff} = 0.4–0.48 m^2/g. The radioactive-decay constant λ of this radionuclide is equal to 0.024.

Random numbers were obtained using the function of generation of a sample of uniformly distributed Latin-hypercube numbers from the Matlab package containing 500 values for each model parameter. A program for solving the transcendental equation (3) for determining the values of S_{max} and τ_{max} at a definite distance from a radiocontamination source has been developed in Microsoft Excel. This program allows one to perform calculations for any sample of random model parameters. The statistical processing and simulation of the distributions of the S_{max} and τ_{max} values obtained for the distances 10, 30, 50, 70, and 80 m from the radiocontamination source were performed using the XLSTAT program integrated into Microsoft Excel.

The primary processing of the data obtained is usually performed for the purpose of determining a law that would define the distribution of the random model values of S_{max} and τ_{max} most adequately. The correspondence of the experimental distribution to the theoretical one is verified with the use of different goodness-of-fit tests. This verification is necessary to be certain that the theoretical model is not contrary to the data obtained and its use did not lead to the appearance of large errors in the probability calculations. The uniform distribution was used as the approximate distribution.

The χ^2 and Kolmogotorv–Smirnov criteria were used for verification of the correspondence of the empirical distribution function obtained to the theoretical one. The comparison of the experimental distributions with the corre-

sponding approximation distributions, performed with the use of the indicated criteria, has shown that the distribution of the S_{max} values is not uniform. Then we investigated samples of ln S_{max} and τ_{max} values. For the first quantity, the Kolmogorov–Smirnov criterion (the maximum difference between the empirical and postulated distribution functions) calculated at different distances from the radiocontamination source is equal to 0.004543–0.004650; its critical value falls within the interval 0.1016–0.1040. The two-sample significance p-level is equal to unity at a 95% reliability. The calculated χ^2 criterion is equal to 0.32–0.4 (its critical value is 27.587) at a one-sample significance p-level equal to unity at a 95% reliability. Thus, there are no differences between the theoretical uniform distribution and the experimental distribution obtained for ln S_{max}.

For a sample of τ_{max}, the Kolmogorov–Smirnov criterion calculated at different distances from the radiocontamination source is equal to 0.005224–0.004439; its critical values falls within the interval 0.1168–0.0993. The two-sample significance p-level is also equal to unity at a 95% reliability. The calculated χ^2 criterion is equal to 0.64–0.80 (its critical value is 27.587) at a one-sample significance p-level equal to unity at a 95% reliability. The theoretical uniform distribution was identical to the experimental distribution of the τ_{max} values. For comparison, the main parameters of the experimental and theoretical distributions are presented in Table 2.

Thus, the analysis of the proper statistics approximating the experimental distributions of the S_{max} and τ_{max} values has shown that, for the above-described model of radionuclide migration in a porous medium, the distribution of the ln S_{max} and τ_{max} values at all distances from the radiocontamination source is most adequately defined by the normal law.

An analogous approach can be used for solving the inverse problem on determination of errors in the experimentally determined input parameters of a model.

NOTATION

C, specific activity of a radionuclide in the liquid phase, Bq/liter; C_0, initial specific activity of the radionuclide in the liquid phase, Bq/liter; D, dispersion coefficient in porous media, m^2/g; D_m, coefficient of molecular diffusion in the skeleton of a rock, m^2/g; $D_{eff} = \left(D + \dfrac{\rho K_d D_m}{n \psi}\right)$, effective diffusion coefficient, m^2/g; K_d, distribution coefficient of a water-soluble compound, cm^3/kg; n, active porosity of the rock skeleton, m^3/m^3; $R = \left(1 + \dfrac{\rho K_d}{n}\right)$, retrogradation coefficient; $S = C/C_0$, dimensionless specific activity; t, time, year; $u = zV/(nD_{eff})$, dimensionless coordinate; V, velocity of flow, m/year; x, parameters of functions; X_j, parameters of a model; z, coordinate in the direction of liquid motion; $\beta = \lambda n^2 R D_{eff}/V^2$, dimensionless constant of radionuclide decay, λ, constant of radionuclide decay, 1/g; ρ, density of the rock skeleton, kg/cm^3; $\tau = tV^2/(Rn^3 D_{eff})$, dimensionless time; ψ, twisting coefficient characterizing the heterogeneity of the porous medium. Subscripts: d, distribution; eff, effective; 0, initial; max, maximum; m, molecular.

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