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VERTICAL TRANSFER OF ^{137}Cs IN PEAT SOILS FALLEN OUT AS A CONSEQUENCE OF THE CHERNOBYL ACCIDENT

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On the basis of measurements of the specific radioactivity of soil specimens sampled with a small step along their deposition, depth profiles are plotted for the curves of the vertical change in ^{137}Cs in peat soils not treated since the Chernobyl accident. A theoretical analysis is provided for the plotted profiles that shows that the vertical transfer of ^{137}Cs in these soils is described adequately within the framework of a model that accounts for the processes of diffusion and convection of the radionuclide in the soil solution and its sorption by the solid phase of the soil. The parameters of the model are determined and a prediction is given for the migration character of ^{137}Cs.

To predict the kinetics of change in the radiation status of territories polluted as a result of the Chernobyl accident and to find methods to eliminate the consequences of the accident it is important to establish the character and mechanisms of migration of radionuclides in various types of soils.

The state and behavior of radionuclides fallen out as a consequence of the Chernobyl accident is studied most intensely for the 30-kilometer zone around the Chernobyl nuclear power station (CNPS) (see, e.g., [1–5]). It is shown in a series of works that territorial zones of the accidental ejection are characterized by various forms of fall-out of radionuclides in a proportion depending on the distance from the CNPS.

Inasmuch as the behavior of radionuclides in soil depends substantially on their fall-out forms and the physicochemical characteristics of the soil it is necessity to determine the migration mechanisms and forms of existence for each of the soil types of various regions of the polluted territories.

Peat soils of meliorated and non-meliorated lands constitute a considerable portion of the radioactively polluted territories. The major element of the radioactive pollution for most areas of the mentioned lands is ^{137}Cs.

In the present work the problem of revealing the regularities of the vertical migration of ^{137}Cs is peat soils of meliorated lands was posed.

Peat lands of the Pinsk Multipurpose Branch of the Belarusian Research Institute of Melioration and Water Resources, which possessed an original pollution level up to 10 Ci/km^2, were chosen. The choice of these lands appeared to be the most suitable for the posed problem because the staff of the aforementioned Branch established experimental checks on these parcels prior to the accident at the CNPS, on which herbs were cultivated till 1992 without any cultivation of land. This made it possible to study the regularities in vertical ^{137}Cs migration in peat soils under conditions of virgin lands. The checks differed from one another by the regimes of artificial flooding (see Table 1) and by the types of herbs cultivated. The sampling of the specimens was carried out from a depth of up to 0.2 m. Initially a soil column 0.125 m in diameter and 0.20 m in height was sampled, which then was cut perpendicularly to its axis into discs 0.025 m thick. Each specimen was formed from three discs cut from three different soil columns at the same depth. The mass of a sample comprised about 1 kg and was comparable with the mass of the standard for radioactivity measurements. To ensure statistical averaging, three specimens were sampled from each depth. At each sampling level the humidity of the soil was determined, which varied from 62 to 72%

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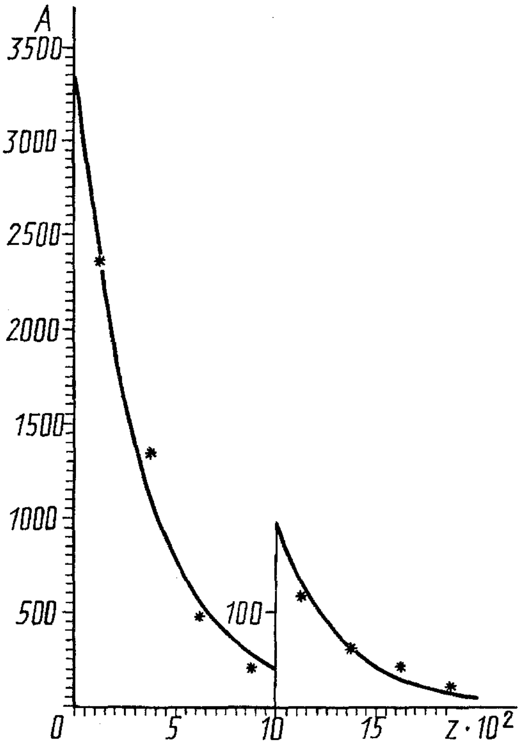
TABLE 1. ^{137}Cs Migration Characteristics in Various Soils

No. of check	Check flooding regime	Saline pH	Ash content, %	A_0, Bq/kg	$D \cdot 10^{12}$, m^2/sec	$\beta \cdot 10^8$, sec^{-1}	$\alpha \cdot 10^{10}$, m/sec
1	Without flooding	6.2	11.2	118.9	13.1	1.55	1.69
2	Flooding for 10 days in spring	6.2	24.1	119.3	12.0	1.55	1.68
3	Flooding in winter	6.1	16.0	152.9	7.3	1.43	1.73
8	Flooding in summer and autumn for 5 days	6.2	14.1	137.0	5.9	1.42	1.88
11	Flooding for 5 days in autumn	6.4	17.4	141.5	3.2	1.35	1.80



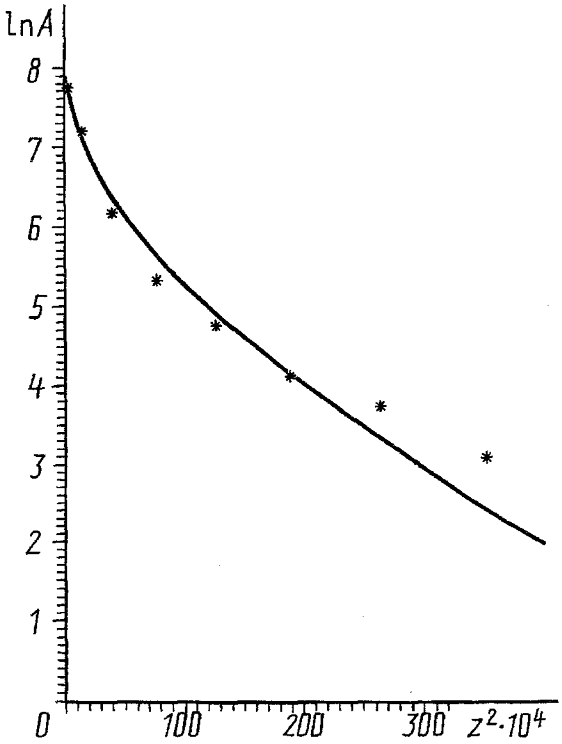


Fig. 1. Change in the specific activity of the peat soil from check No.1 by ^{137}Cs (points, experiment; line, calculation according (11)). A, Bq/kg; z, m.

Fig. 2. Dependence of the logarithm of the specific activity by ^{137}Cs on the square of the depth of check No.1 (points, experiment; line, calculation). z^2, m^2.

depending on the sampling level and the check number. For each check the humidity of the soil at the level of 0.2 m was 2–4% higher than at the surface. Sampling of the specimens was carried out in 1991 and 1992.

A gamma-spectrometer with an NaI(Tl) scintillation counter and a multichannel amplitude analyzer was used for measurements of the radioactivity of the samples.

The results of the measurements showned that the activity of the peat soil by ^{137}Cs decreases rather rapidly with depth. Figure 1 shows as an example the profile of the vertical change in the specific radioactivity for check No. 1 according to data of 1991. The character of the change in the specific activity with depth for the other checks is the same as for the specified one. The maximum activity is possessed by the upper layer. It should be noted that in moving from sample to sample with the same deposition depth the activity value varies within certain limits exceeding the accuracy of the measurements. Such a scatter can be connected with both the scatter in the values of the parameters of the physicochemical characteristics of the soils under analysis and fluctuations in the density of the fall-out radionuclides. Despite this fact the data obtained in the experiments make it possible to determine with reasonable reliability the regularities and to evaluate possible mechanisms of vertical ^{137}Cs migration in the soil type under investigation.

Migration of radionuclides along a vertical soil profile in the general case, as is known [6], can be determined by many factors. The decisive ones are connected with diffusion processes of free ions in the soil solution and colloid soil particles on which radionuclides are sorbed, as well as with convective transfer of matter. The latter can be caused by filtration of atmospheric precipitates into the depth of the soil and capillary streams of moisture upward to the surface due to gradients of the humidity and temperature of the soil. The migration character of radionuclides can be influenced appreciably by their sorption by soil particles.

In the case of transfer of matter that takes place solely as a result of diffusion the change in the specific activity (A) of the soil with depth (z) is described, as is known, by a Gaussian function, i.e., the dependence of ln A on the square of the depth is linear:

$$\ln A = \ln \frac{C}{\sqrt{\pi D t}} - \frac{z^2}{4Dt}, \tag{1}$$

where D is the diffusion coefficient of the radionuclide in the soil; t is time from the instant of the accident at the CNPS; C is a constant that determines the activity of the fallen-out radionuclide.

An analysis of the experimental data showed that the observed dependence of ln A on z^2 (Fig. 2) is not linear. The existingcelebrated approach (e.g., [3, 7, 8]) to the description of the nonlinear dependence of ln A on z^2 based on the assumption of two rapidly and slowly migrating components determined by the exchange and nonexchange forms of existence of radionuclides is purely formal since the processes of migration of these forms, if any, are not additive because they are interrelated: the exchange form, for example, is partially converted to the nonexchange form in the migration process.

The most adequate description of the observed dependence of the variation in ^{137}Cs concentration with depth, as was shown by the analysis, can be given within the framework of the diffusion-sorption-migration model. The decisive role of the sorption process in ^{137}Cs migration is indicated by the fact that the transition to the nonexchange form is a characteristic feature of the behavior of this radionuclide in soil.

Soil can be represented as composed of two phases [9], the soil solution and the solid soil phase. To give a quantitative description of ^{137}Cs migration on the basis of the specified model we will assume that diffusion and convective transfer take place in the soil solution, whereas the solid phase sorbs ^{137}Cs irreversibly without saturation. Then vertical ^{137}Cs migration in the soil solution will be described by the equation

$$\frac{\partial C_1}{\partial t} = D \frac{\partial^2 C_1}{\partial z^2} - \alpha \frac{\partial C_1}{\partial z} - \beta C_1, \tag{2}$$

where C_1 is the radionuclide concentration in the soil solution at the depth z; D is the effective diffusion coefficient; α is the rate of convective radionuclide transfer; β is the rate constant of sorption in the solid phase. The convective transfer rate (α) will be positive or negative depending on the flux direction (into the depth or to the surface).

In addition to (2), balance equations should be written for transfer and sorption of matter from the soil solution to the solid phase:

$$v_2 \frac{\partial C_2}{\partial t} = \beta C_1 v_1, \tag{3}$$

where C_2 is the ^{137}Cs concentration in the solid phase at the instant t at the depth z; ν_1 and ν_2 are the exchange portions of the liquid and solid phase, which depend, strictly speaking, on the depth.

The total specific activity of the soil is then determined in the following way:

$$A = (C_1\nu_1 + C_2\nu_2)/\rho \,, \tag{4}$$

where ρ is the mean soil density.

The solutions of the Eqs. (2), (3) were sought for the initial and boundary conditions

$$D\frac{\partial C_1}{\partial z}\Big|_{z=0} - \alpha C_1\big|_{z=0} = 0 \,, \tag{5}$$

$$C_1\,(z\,,\,0) = C\delta\,(z\,,\,0)\,, \tag{6}$$

$$C_1\,(z\,,\,t) = 0\,, \tag{7}$$

$$z \to \infty\,,$$

where C is the number of radioactive ^{137}Cs atoms fallen-out onto unit area at the instant of the accident; $\delta(z,\,0)$ is the delta function. The conditions (5) and (6) are written by recognizing that the ^{137}Cs radionuclide fallen out onto the soil is leached out rapidly.

The solution of Eq. (2) has the following form:

$$C_1\,(z\,,\,t) = C\left\{\frac{1}{\sqrt{\pi Dt}}\exp\left[-\beta t - \frac{(z-at)^2}{4Dt}\right] - \right.$$
$$\left. - \frac{\alpha}{2D}\exp\left(-\beta t + \frac{\alpha z}{D}\right)\operatorname{erfc}\left(\frac{z}{2\sqrt{Dt}} + \frac{\alpha}{2}\sqrt{\left(\frac{t}{D}\right)}\right)\right\}. \tag{8}$$

From Eq. (3) it follows that

$$C_2\,(z\,,\,t) = \beta\frac{\nu_1}{\nu_2}\int_0^t C_1\,(z\,,\,t)\,dt\,. \tag{9}$$

Substituting (8) and (9) into (4), we obtain an expression for the specific activity of the soil:

$$A\,(z\,,\,t) = A_0\left\{\frac{1}{\sqrt{\pi Dt}}\exp\left[-\beta t - \frac{(z-at)^2}{4Dt}\right] - \right.$$
$$\left. - \frac{\alpha}{2D}\exp\left(-\beta t + \frac{\alpha z}{D}\right)\operatorname{erfc}\,(z/2\sqrt{Dt} + \alpha\sqrt{t/D/2})\right\} +$$
$$+ A_0\beta\int_0^t\left\{\frac{1}{\sqrt{\pi Dt}}\exp\left[-\beta t - \frac{(z-at)^2}{4Dt}\right] - \right.$$
$$\left. - \frac{\alpha}{2D}\exp\left(-\beta t + \frac{\alpha z}{D}\right)\operatorname{erfc}\,(z/2\sqrt{Dt} + \alpha\sqrt{t/D/2})\right\}dt\,, \tag{10}$$

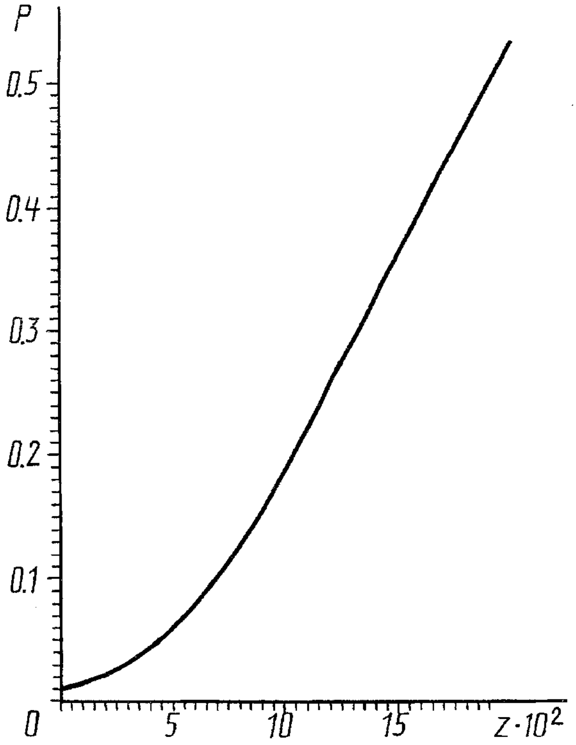


Fig. 3. Change in the relative fraction of the ^{137}Cs concentration with depth in the soil solution.

where $A_0 = Cv_1 e^{\lambda t}/\rho$; λ is the radioactive decay constant for the given isotope.

The parameters D, α, β, and A can be determined from a comparison of the experimentally observed profile of the distribution curves of ^{137}Cs in the soil (see Fig. 1) with that calculated according to (10).

The values of the parameters found in this manner for all of the checks are listed in Table 1. In addition, according to the obtained values of the parameters such characteristics of migration as the relative fraction of ^{137}Cs in the soil solution

$$P = \frac{C_1 v_1}{C_1 v_1 + C_2 v_2} \tag{11}$$

and the distribution coefficient [4]

$$K_p = \frac{C_2}{C_1} \frac{m_1}{m_2}, \tag{12}$$

can be obtained, where m_1 and m_2 are the mass fractions of the soil solution and the solid phase per unit volume of soil.

The dependence of ln A on z^2 calculated according to (10) mostly corresponds to the observed one (Fig. 2). A certain deviation of the experimental data from the results of calculations toward higher values is possibly connected with the background of global fall-out.

As is seen from Fig. 3, the calculated ^{137}Cs concentration in the soil solution changes nonmonotonically, which is connected with the character of manifestations of the competing processes of diffusion and convection at various depths of the soil. The value of the coefficient P for the upper layer is about 1%, which is close to the observed values [4]. The calculated distribution coefficient (12) for the upper layer of the soils under investigation is about 150.

To predict the character of change in the penetration depth of radioactive pollution into the peat soils with time the thickness of the layer that contains 90% of all radioactivity ($z_{90\%}$) was calculated as a function of time. It was obtained from the relationship

$$\frac{\int_0^{z_{90}} A(z, t)\, dz}{\int_0^{\infty} A(z, t)\, dz} = 0.9, \tag{13}$$

where $A(z, t)$ is determined by the expression (10).

The calculations showed that the dependence of the thickness of the layer that contains 90% of the activity ($z_{90\%}$) as a function of time is characterized by a curve with a plateau. Thus, 6–7 years after the accident the thickness of the specified layer reaches a certain value (approximately 0.07 m), which then varies only slightly. This is evidenced by the fact that the profiles of the curves of the vertical ^{137}Cs distribution plotted according to the results of measurements of 1991 and 1992 differ only slightly.

The results obtained in the experiments also show that short-term flooding of parcels in various seasons affects the distribution of radionuclides in the soil only slightly.

NOTATION

A, specific activity of soil; P, relative fraction of ^{137}Cs concentration in the soil solution; K_p, distribution coefficient; $z_{90\%}$, thickness of the layer that contains 90% of all activity.

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