THE DYNAMIC OF THE MODEL PARAMETERS OF ^{137}Cs VERTICAL MIGRATION IN SOIL.

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Abstract

Knowledge of the distribution in soil of radionuclides deposited following the release their to the environment is important for a reliable assessment of external dose and root uptake by plants. In order to predict the downward migration of radionuclide we should know the evolution in time the parameters of the model describing distribution of the radionuclide in soil layer. In this paper diffusion model was applied to describe distribution of ^{137}Cs-activity concentration in soil profile of 3 sites located on soddy-podzolic and soddy soils. And the convection-diffusion equation was applied to 3 sites situated on peaty-gley ang peaty-boggy soils. Soil samples have been collected during 1989-2001 years at the southern of Belarus. Earlier, some authors studied the dynamic of the parameters of convection-diffusion model and pointed out a decrease with time these parameters in several soils. However, the period of investigation was short and they didn't give an exact relationship between parameters and time. Our analyses of the dynamic of diffusion (D) and convection (V) coefficients showed that D didn't change for sites with soddy soils during the period of investigation. The prognostic calculations of ^{137}Cs-activity concentration in soil layer with constant value D were carrying out for these sites. At the same time D and V for sites with peaty soil decreased with time. This decrease was well described by power function ($f=at^b$). Obtained dependencies D(t),V(t) were used for prediction of ^{137}Cs activity concentration in soil layer of sites with peaty soil.

1. Introduction

The descriptions of the radionuclides migration in terms of different models and estimations of model parameters have been carried out in some papers (see /1-5/). In particular, in /4/ the assessments of model parameters during the period 1989-1994y for typical soils of Belarus have been obtained. The authors have noted that parameters showed tendency to decrease with time. However, the information about value and character of this decrease didn’t indicate. At the same time the knowledge of the dynamic of the model parameters is needed to construct the long-term prediction and to elaborate the protective measure.

The purpose of the present work was to compare the results of descriptions of ^{137}Cs vertical migration in soil obtained in term of three models: compartment model, diffusion model and convection-diffusion model during the long period of time; and to study the behavior of models parameters in order to determine their time dependencies.

2. Materials and methods

Six sites in the southern part of Belarus were chosen to study the dynamic the characteristics of vertical migration of ^{137}Cs in soil. Soil samples were taken every year from 1989 to 2001. Information of the sites is given in Table 1. The sites are situated near the villages Kruky, Lesok, Nezhihov, Kylazhin and Radin(Gomel region of Belarus). The objects of observation were soddy-podzolic and peaty-gley soils selected on polluted territories of 30 km zone of Chernobyl NPP. ^{137}Cs content was defined using gamma-spectrometer AFORA LP 4900B with the semiconductor detector DGDK-160B.

Table 1. Characteristic of Experimental sites

Site situation (distance from NPP, km)	Type of soil	pH aq	pH KCL	Organic carbon (%)	Loss of ignition (%)	Density of ^{137}Cs contamination Bq/km$^2\cdot10^{-12}$	Content in soil, mg*100g^{-1}	
							Ca^{2+}	Mg^{2+}
Kruky (20)	Soddy-podzolic	5,2	4,1	0,9	2,8	23,5	24,2±1,9	3,5±1,0
Lesok (19)	Soddy-podzolic	4,9	3,9	1,4	7,5	5,3	52,0±12,1	–
Nezhihov-1(25)	Soddy	4,9	3,9	4,6	15,9	3,9	45,7±5,2	8,5±0,6
Nezhihov-2(25)	Peaty-boggy	6,0	5,3	18,9	52,7	3,9	738±42,0	80,8±3,9

Kylazhin (20)	Peaty-gley	6,5	5,0	12,3	19,0	21,7	90,6±6,5	20,6±1,6
Radin (21)	Peaty-gley	7,1	6,4	13,4	38,6	6,1	853,3±55,0	84,4±4,4

3. Results and discussion

3.1. Vertical distribution of ^{137}Cs in soil

The cumulative distributions of ^{137}Cs in top soil horizons are submitted on Fig.1. Data shows that distribution of ^{137}Cs in peaty soils strong different from distribution in soddy soils. In first case contents of ^{137}Cs in 0-1cm and 0-5cm soil layers in 1989 are 20% and 60-80%, respectively. During period of investigation contents of ^{137}Cs in 0-1cm and 0-5cm soil layers changed at most 5-10%. While contents of ^{137}Cs in 0-1cm layer of soddy soils decreased up to 30-40% and contents of ^{137}Cs in 0-5cm soil layer decreased by 10%. Therefore, redistribution of ^{137}Cs in soddy soils was mainly in 0-5cm soil layer. According to represented data, migration of ^{137}Cs within 0-5cm layer in soddy soil was going on more intensively than in peaty soils. But in peaty soil the center of activity inventory (defined as $X_a = \sum_{i=1} A_i^0 x_i^0$, where $x_i^0 = (x_i + x_{i-1})/2$ is the center of i-th soil layer) was situated deeper than center of inventory in soddy soils. So, migration of ^{137}Cs was going on more intensively in peaty soils from 1986 to 1989 y.

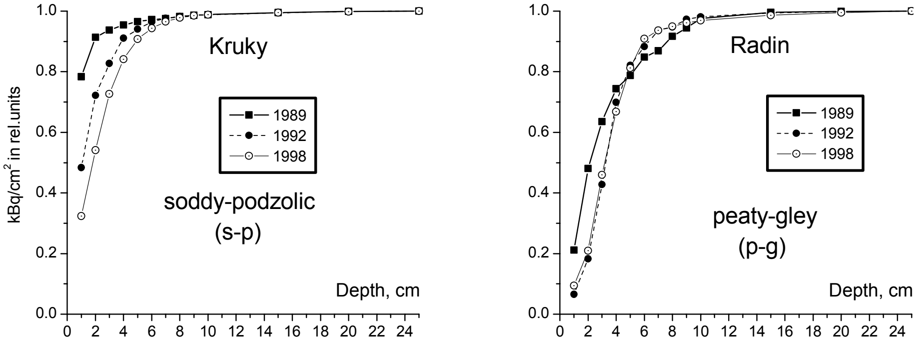

Fig.1. Cumulative distributions of ^{137}Cs in soil profile of sites represented different soils.

3.2. Compartment model

To evaluate the migration rate of ^{137}Cs in various soil layers, a compartment model was used(see /6/). In the general case, the system of differential equations of the compartment model describing radionuclide migration in soil is written as

$$\frac{dy_1}{dt} = -k_{12} y_1 - \lambda y_1 \tag{1}$$

$$\frac{dy_{n>1}}{dt} = k_{n-1,n} y_{n-1} - k_{n,n+1} y_n - \lambda y_n \tag{2}$$

where $y_n(t)$ is the content of radionuclide in the soil layer n at time t, λ is the decay constant, k_{ij} – the coefficient of radionuclide transfer from i-th layer to j-th layer.

The calculated recurrent expressions for functions $y_n(t)$ are written as follows:

$$y_1(t) = y_1(0) \cdot e^{-(k_{12}+\lambda)t} \tag{3}$$

$$y_{n>1}(t) = y_1(0)\left(\prod_{i=1}^{n-1} k_{i,i+1}\right) \sum_{i=1}^{n} \frac{e^{-\left(k_{i,i+1}+\lambda\right)}}{\prod\limits_{\substack{j=1 \\ j\neq i}}^{n} \left(k_{j,j+1} - k_{i,i+1}\right)} \tag{4}$$

After substituting the experimental data for $y_1(0)$ and $y_n(t)$ the above expressions are transformed into the system of transcendental equations that are applied for computing the values of transfer parameters $k_{i,i+1}$. The number of compartments used for calculations of $k_{i,i+1}$ was 3.

On the basis of the coefficients $k_{i,i+1}$ the rate of radionuclide motion $v(i)$ in i-th soil layer can be evaluated. The quantity $v(i)$ is written as $v(i)=\Delta x_i/\tau_i$, where $\tau_i = ln(2)/k_{i,i+1}$ is the residence half-time of the radionuclide in the i-th layer. Thus the compartment model gives the possibility to estimate the variation of the radionuclide migration behavior with the depth of the soil profile. It must be stressed that $v(i)$ is not exactly the rate of radionuclide transport but only a quantity that is used to characterize approximately the rate of transport.

The estimated values of *v(i)* for some years are showed at the Fig.2. According to obtained data the radionuclide migration rates *v(i)* tended to decrease with time. This behavior of *v(i)* had both the sites on peaty and the sites on soddy soils. At the same time both types were characterized by different behavior of *v(i)* with increasing layer depth. In case of soddy soils the rate *v(i)* increased with depth. For peaty soils the maximum in depth dependence of *v(i)* was observed. Thus the distribution of the quantity *v(i)* in soil profile can be considered as the individual migration characteristic of the site. So, the compartment model doesn't available for long-term prognoses of ^{137}Cs activity concentration in soil layers because of change of migration rates *v(i)*with depth.

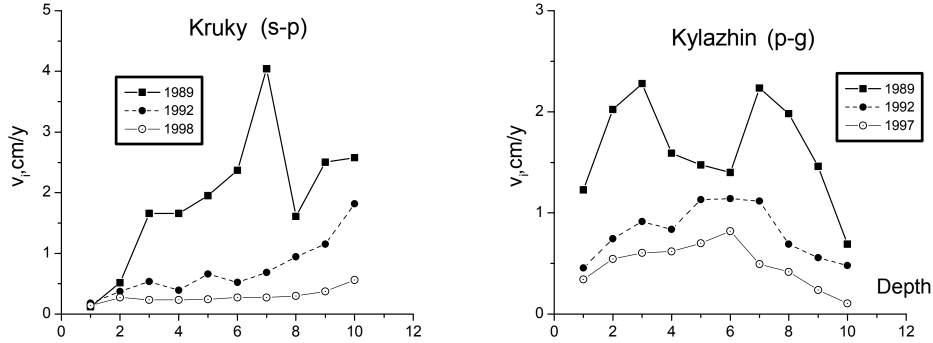

Fig. 2. ^{137}Cs migration rate v_i in the soil layers of the sites.

The obtained values of *v(i)* have been used to calculate model average migration rate $V_0^{CM} = \sum_{i=1} A_i^0 v_i$, where $A_i^0 = A_i \big/ \sum_j A_j$ is relative contents of radionuclide activity in *i*-th layer. The values of v_0^{CM} have been compared with empirical average migration rate defined as $V_a = X_a/\Delta t$. The variation of ratio R= V_a/V_0^{CM} was about 10%. The agreement between the quantities V_0^{CM} and V_a gives foundation to use compartment model for description radionuclides migration in soil layers.

Table 2. Values of empirical V_a and model V_0^{CM} average migration rate(cm/y)

Sites	1989		1990		1991		1992		1993		1994		1995		1996		1997		1998	
	V_a	V_0^{CM}	V_a	V_0^{CM}	V_a	V_0^{CM}	V_a	V_0^{CM}	V_a	V_0^{CM}	V_a	V_0^{CM}	V_a	V_0^{CM}	V_a	V_0^{CM}	V_a	V_0^{CM}	V_a	V_0^{CM}
Kruky	0.36	0.33	0.31	0.32	0.48	0.45	0.30	0.33	0.37	0.34	0.23	0.24	–	–	0.22	0.23	0.17	0.18	0.20	0.22
Lesok	0.33	0.27	0.35	0.38	0.53	0.56	0.17	0.14	0.33	0.29	0.29	0.28	0.24	0.23	–	–	0.14	0.15	0.16	0.17
Nezhihov-1	0.44	0.46	0.46	0.49	0.21	0.19	0.21	0.20	0.30	0.22	0.18	0.19	0.25	0.25	0.20	0.21	–	–	–	–
Kylazhin	1.68	1.58	1.30	1.24	0.97	1.00	0.72	0.76	0.91	0.82	0.67	0.54	–	–	0.30	0.31	0.51	0.47	–	
Radin	1.05	1.18	0.57	0.57	0.45	0.47	0.61	0.54	0.45	0.45	0.44	0.44	–	–	0.37	0.34	0.30	0.30	0.31	0.29
Nezhihov-2	1.57	1.55	1.06	1.13	–	–	0.99	0.93	0.60	0.63	0.68	0.61	–	–	–	–	0.70	0.64	–	–

3.3. Diffusion model

The descriptions of activity concentration distribution in profile have been carried out by using diffusion model (see /4/) for three sites on soddy soil. The expression of diffusion model is written as follows:

$$c(x,t) = \frac{1}{\sqrt{\pi \cdot \mathrm{D} \cdot t}} \cdot \exp\left(-\frac{x^2}{4 \cdot \mathrm{D} \cdot t} \right) \tag{5}$$

Here *c(x,t)* is relative activity concentration in depth *x* at time *t*, D is the diffusion coefficient. Fig.3 shows the result of description for Kruky site. There was a good agreement between theoretical and experimental descriptions for sites with soddy soils. The relative error of diffusion coefficient (D) was at the average 16%.

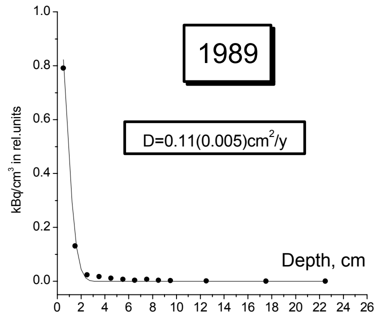

Fig.3.Diffusion model description of ^{137}Cs activity in soil profile of Kruky site.

At the same time, there was considerable deviation experimental data from theoretical description for sites on peaty soils. The diffusion model can not reproduce the shift of activity maximum to lower layer. The average values of D were 0.21 cm^2/year for Radin site, 0.99 cm^2/y for Kylazhin and 1.00 cm^2/y for Nezhihov-2. The diffusion model gives D, which is too large because of the fit ignores the details of the measured activity profile close to the surface.

The investigation of dynamic of diffusion coefficient (D) for sites with soddy soils showed that D didn't change during the period of investigation from 1989 to 2001(see Fig.4.). So, the average value of D can be used to predict the contents of ^{137}Cs activity in soil layers during short-term period. The average values of D were 0.33 cm^2/y for Kruki site, 0.28 cm^2/y for Lesok site and 0.21 cm^2/y for Nezhihov-1.

Table 3. Values of diffusion coefficient D (cm^2/y)

Sites	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998	2001	D_{av}
Kruky	0.11	0.1	0.82	0.21	0.73	0.21		0.32	0.19	0.28		***0.33***
Lesok	0.095	0.093	0.82	0.071	0.6	0.49	0.31		0.07	0.14	0.081	**0.28**
Nezhihov-1	0.096	0.35	0.071	0.099	0.42	0.076	0.35	0.21				***0.21***

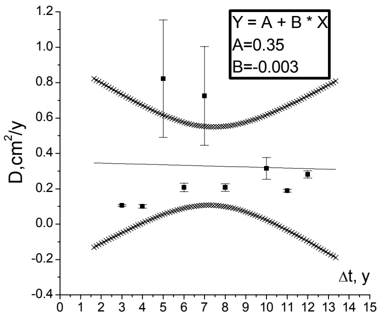

Fig.4. Diffusion coefficient D for Kruky site. Cross(x) lines show 95% confidence limits.

Obtained average values were used for prognosis evaluation of ^{137}Cs activity in soil profile considered sites in 2006 year. The results show that redistribution of ^{137}Cs mainly will within 0-10cm layer. ^{137}Cs activity will decrease in layer 0-3cm and increase in layer 3-10cm. The content of ^{137}Cs activity in 0-5 cm soil layer will decrease on 5-10% to 2006 in comparison with values of 1998 year.

3.4. Convection-Diffusion model

To describe the activity concentration distribution in profile on peaty soils, the convection-diffusion model has been applied. The expression used for description of distributions is written as follows:

$$c(x,t) = \frac{1}{2\cdot\sqrt{\pi\cdot \mathrm{D}\cdot t}}\cdot\left\{\exp\left(-\frac{(x-\mathrm{V}\cdot t)}{4\cdot \mathrm{D}\cdot t}\right)+\exp\left(-\frac{(x+\mathrm{V}\cdot t)}{4\cdot \mathrm{D}\cdot t}\right)\right\} \qquad (6)$$

Here *c(x,t)* is relative activity concentration in depth *x* at time *t*, D is the diffusion coefficients, V is the coefficient of convection. Fig.5 shows the result of description for Radin site. The same descriptions have been carried out for Kylazhin and Nezhihov-2 sites too. The relative error of D was in average 20%, the relative error of V was in 2,5 time less (~5%).

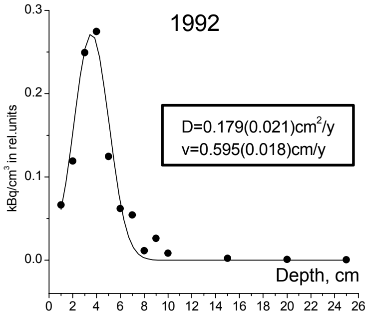

Fig.5. Convection-diffusion model description of ^{137}Cs distribution in soil profile of Radin site.

The analyses of behavior D and V showed that their change with time was well described by power function (f=at^b). Obtained dependencies D(t) and V(t) are submitted on Fig. 6. Relationships between parameters and time are written as: Radin– $\mathrm{D(t)} = 0{,}66\cdot \mathrm{t}^{-0{,}59}$; $\mathrm{V(t)} = 1{,}67\cdot \mathrm{t}^{-0{,}71}$; Kylazhin– $\mathrm{D(t)} = 5{,}02\cdot \mathrm{t}^{-0{,}88}$; $\mathrm{V(t)} = 4{,}42\cdot \mathrm{t}^{-1}$; Nezhihov-2– $\mathrm{D(t)} = 3{,}93\cdot \mathrm{t}^{-0{,}8}$; $\mathrm{V(t)} = 2{,}14\cdot \mathrm{t}^{-0{,}62}$. The coefficients D and V decreased in 7 time for Kylazhin site, and in 3 time for Radin and Nezhihov-2 during the period of investigation.

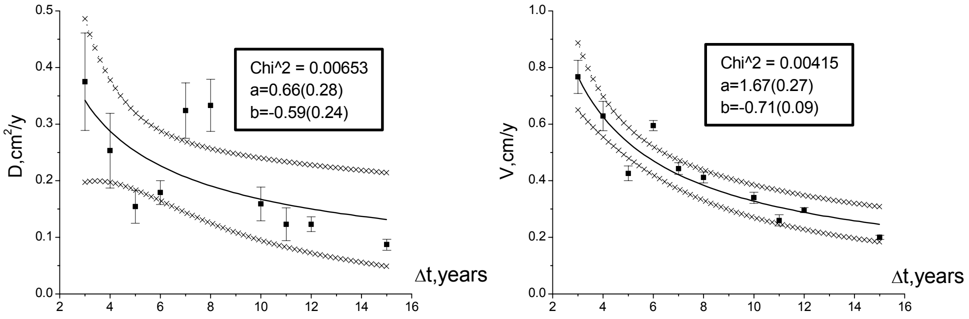

Fig.6. Descriptions by power function F(t)=at^b the time dependencies of D and V for Radin site. Cross(x) lines show upper and lower 95% confidence limits.

Table 4. Values of diffusion D (cm^2/y) and convection V(cm/y) coefficients

Sites	1989		1990		1991		1992		1993		1994		1996		1997		1998		2001	
	D	V	D	V	D	V	D	V	D	V	D	V	D	V	D	V	D	V	D	V
Kylazhin	1.47	1.53	1.97	1.23	1.49	0.67	1.17	0.41	1.05	0.89	0.50	0.76	0.15	0.27	0.81	0.55	–	–	0.29	0.34
Radin	0.38	0.77	0.25	0.63	0.15	0.43	0.18	0.60	0.32	0.44	0.33	0.41	0.16	0.34	0.12	0.26	0.12	0.30	0.087	0.20
Nezhihov-2	1.85	1.20	0.74	0.68	–	–	1.60	0.88	0.63	0.49	0.54	0.69	–	–	0.64	0.50	–	–	–	–

Obtained dependencies D(t),V(t) were used for prediction of ^{137}Cs activity concentration in soil layer of sites with peaty soil. To investigate the possibility of using constant D and V to make reliable prediction, the distributions of ^{137}Cs activity concentration in 2006y were constructed with constant value D and V (1998y). Prognoses with constant parameters D and V predict less content of ^{137}Cs activity in upper soil layers than prognoses with variable D and V. In particular, the median of ^{137}Cs distribution with variable D and V will be 4,1; 4,6; 6,7 cm for Radin, Kylazhin and Nezhihov-2 site, respectively. But with constant parameters median will be more in 1,8 time for Kylazhin, and in 1,4 and 1,5 time for Radin and Nezhihov-2.

Taking into account the significance of upper root-inhabited layer, the prognosis estimations of ^{137}Cs activity concentration in 0-5cm soil layer were carried out. The comparison of prognosis estimations obtained with variable and constant parameters showed that in first case the content of ^{137}Cs will be in 2 time more than in second case.

4. Conclusions

The vertical distributions of ^{137}Cs activity concentration in soil in Gomel region of Belarus have been determined for six sites situated at the territory of 30 km zone of Chernobyl NPP. The data obtained were used to evaluate in terms of the compartment model the radionuclide migration rate $v(i)$ in soil layers and the average migration rate V_0^{CM} in the soil horizon of the sites. The rate $v(i)$ shows the tendency to increase with increasing depth of a soil layer for sites on soddy soils. At the same time there is maximum in dependence of $v(i)$ on depth for sites on peaty soils. The calculated values of the rate V_0^{CM} agree with the assessments of the empirical average migration rate $V_{a.}$. The variation of ratio R= $V_a/\ V_0^{CM}$ is about 10%. The rate $v(i)$ has tendency to decrease during the period of investigation. This fact makes difficult using of compartment model for long-term prognosis of ^{137}Cs activity concentration in soil layers.

The diffusion model describes satisfactorily the measurement results only for sites on soddy soils. The average values of diffusion coefficient D for these sites lie in the range 0,21–0,33 cm^2/year. The diffusion coefficients D didn't change during the period of investigation. Thus obtained average values can use for prognosis evaluation of ^{137}Cs activity in soil profile considered sites. The diffusion model gives too high assessments of diffusion coefficients D for sites on peaty soils.

The convection-diffusion model can reproduce the shift of ^{137}Cs activity maximum to lower layer in sites with peaty soils. The description by this model the distribution of ^{137}Cs activity in soil profile agrees with experimental data. The analysis of behavior of diffusion D and convection V coefficients have been carry out. According to the results the time dependencies of D and V showed the tendency to decrease during the period of investigation. These dependencies were satisfactorily described by power functions. Obtained relations have been applied to predict the contents of ^{137}Cs activity in soil layers. The prognoses constructed without taking the dynamic of D and V into account give the conservative values of ^{137}Cs activity in 0-5cm soil layer.

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