RESULTS OF TEN YEARS STUDY OF CHERNOBYL NPP RELEASE FALLOUT PROPERTIES AND BEHAVIOUR IN SOILS

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1. INTRODUCTION

Radioactive contamination of territories of Ukrainian and Belorussian Polesye as a result of ChNPP accidental release is characterized by high level of un-homogenity of fallout properties (physico-chemical properties, radionuclide composition etc.), density of the territory contamination by long-lived radionuclides. On the other hand, the soil-plant cover of contaminated territory is presented by large set of soils, characterized by contrast physico-chemical and water-physical properties.Peculiarities of the behaviour of different radionuclides, represented initially by various components of radioactive fallout, in soils, as a first link of migration chains are considered.

2. METHOD

The agreed-upon radioecological, radiochemical, soil-chemical and other methods of investigations have been used: *in situ* observation as well as a model laboratory experiments for the estimation of intensity and possible mechanisms of radionuclides vertical transfer; sequential extraction procedures and isotopic dilution method for estimation of the dynamics of radionuclides mobile forms in soils; α- and γ-spectrometry, autoradiography, track-radiography and other nuclear-physical methods for study of properties and radionuclide composition of ChNPP release fallout and their transformation in soils, etc.

Experimental sites for *in situ* observation were choosen with taking into account the following criteria:

- landscape-geochemical conditions, including physico-chemical propertes, granulometric and mineralogical composition of soils;
- physico-chemical properties of fallout (ratio of fuel and condensed components) at various tracks;
- density of the territory contamination by long-lived radionuclides;
- type of lands (natural and improved meadows, fallow lands, agricultural lands).

Most typical soils, represented the soil cover of contaminated territory of Ukraine have been used in model experiments.

3. RESULTS

3.1. Feature of fallout

Survey of the contaminated territories, carried out during the first months after the accident, has shown the great unhomogeneity of soil-plant cover radioactive contamination as well as ununiformity of fallout radionuclide composition (activity and isotopic ratio of ^{95}Zr-^{95}Nb, ^{140}Ba-^{140}La, ^{103,106}Ru, ^{141,144}Ce, ^{134,137}Cs et al) at various tracks of fallout. The presence of hot particles in fallout was shown by the autoradiography of tree leaves.

The further research demonstrated obviously enough that the radioactive fallouts are presented mainly by two components (fuel and condensed ones), contamination of the territory is a superposition of the mentioned components tracks, components ratio depends on the direction and the distance from ChNPP [1,2].

The territories of the "remote" tracks of fallouts are characterized by contamination with condensed component, represented mainly by radioisotopes of Iodine, Caesium and, particularly, Ruthenium. According to their physico-chemical properties these fallouts are similar to global ones [3].

The territories of the "close-in"tracks of fallouts (R<50-60 km) are characterized by the contamination with the superposition of condensed and fuel components of fallout's. The latter is represented small particles of high burn-up uranium oxide fuel with composition similar to that of the fuel in the reactor core but with some depletion of the radioisotopes of more volatile chemical elements (iodine, ruthenium and caesium), some particles were spherical, others were angular shards. Apart from uranium oxide and fission products, many hot particles also contained zirconium and traces of iron, molybdenum, nickel, copper, zinc, silica, aluminium and lead. Within the ChNPP 30-km zone fuel particles were estimated to account for more than 75% of the total radioactive contamination on the ground. Essentially all the radiostrontium and plutonium were associated with particles at the time of deposition.[1,2,4].

Some physico-chemical and nuclear-physical properties of "chernobyl" fuel particles (radionuclide, substantial and granulometric composition, burn-up etc.) have estimated. Data on experimental relationship between i-th radionuclide's activity and ^{144}Ce activity in the fuel particles at the time of the accident are shawn in Table I [5].

TABLE I. Experimental relationship between i-th radionuclide's activity (A_i) and ^{144}Ce activity ($A_{144\,(FHP)}$) in the fuel particles at the time of the accident

Radionuclide	$A_i/A_{144\,(FHP)}$	Radionuclide	$A_i/A_{144\,(FHP)}$
^{90}Sr	0.05	^{137}Cs	0.04
^{95}Zr	2.3	^{141}Ce	1.34
^{103}Ru	1.08	^{144}Ce	1.0
^{106}Ru	0.26	^{154}Eu	0.0015
^{125}Sb	0.006	^{155}Eu	0.0017
^{134}Cs	0.02	$^{239,240}Pu$	0.0004

The fraction of radionuclides, initially contained in fuel particle matrix, in total activity of these ones in soils at the moment of accident, using the data on soil contamination with refractory radionuclides (^{144}Ce etc., taking into account the radioactive decay) can be estimated by the following equations [5] :

$$q(^{137}Cs)=(^{137}Cs/^{144}Ce)_{FP}/(^{137}Cs/^{144}Ce)_{soil}=0.04/(^{137}Cs/^{144}Ce)_{soil}$$

$$q(^{90}Sr)=(^{90}Sr/^{144}Ce)_{FP}/(^{90}Sr/^{144}Ce)_{soil}=0.05/(^{90}Sr/^{144}Ce)_{soil}$$

3.2. Transformation of fallout in soils

Study of the dynamics of fallout transformation in soils has been carried out with the use of sequential extraction techniques, isotopic dilution method, ultrafiltration, dialysis etc. In general, content of ^{137}Cs exchangeable forms in soils is decreasing with time, different decrease rate is noted for conditions, characterised by different soils conditions as well as various initial forms of fallout (ratio of fuel component of fallout to condensed one). Significantly lower content of radiocaesium exchangeable forms as well as higher intensity of its decrease is noted for groups of hydromorphous soils, characterised by higher value of CEC and higher content of clay minerals [6]. However, this phenomena is not an absolute one and has some exceptions. Increase of the content of ^{137}Cs exchangeable forms in soils of some meadows in 30-km since 1990-1991 and its further decrease have been observed [6,7]. It should be noted also that less content of radiocaesium in soils of ChNPP immediate zone is connected not with high content of fuel component in fallout only, but with high sorption by solid phase of hydromorphous soils also.Content of radionuclides fractions, less strongly binded with soils solid phase components, decreases with time. Rate of decrease depends both on radionuclides initial physico-chemical forms of fallout and on soils properties [7].

Comparison of the data on radiocaesium mobile forms in mineral and organic soils with radionuclide transfer factor from these soils to plants allows to make a conclusion about the impossibility of the direct use of mentioned data for the prediction of radiocaesium biological availability. However, the information on radiocaesium mobile forms content in soils could be used as a criteria of its state dynamic in soils.

Dynamics of the content of radiocaesium exchangeable forms in soils depends on above-mentioned factors.

Presence of the fuel particles in the fallout of Chernobyl accidental release has modified the including intensity of the radionuclides, represented by fuel component, to migration chains in terrstrial ecosystems. The mentioned modification depends on both the climatic conditions and on the fuel particles properties [2-4].

The latest data, obtained in 1993-1995, demonstrate, that in some soils there is high quantity of undestructed fuel particles (up to 60-80% in accordance to the data of autoradiography and assessments of the content of ^{90}Sr ion-exchangeable forms in soil).

Significant dependence of the dissolution velocity of fuel particles on the level of physico-chemical transformation of particles matrix (incinerated or non-incinerated fuel; particles, subjected to leaching in soil in natural conditions, etc.) and characteristics of the media (pH, RedOx-potential etc).is shown in model experiments.

3.3. Radionuclides transfer in soils

Close intensity of vertical transfer of different radionuclides (^{144}Ce, ^{90}Sr, $^{134,137}Cs$ et al) in soils on "fuel" tracks of fallout during first 2-3 years after the accidental release has been observed. Later the separation of radionuclides, differing in their physico-chemical properties, in soil profiles has been found. Abnormally high migrative ability of Caesium radioisotopes in hydromorphous organic soils has been observed.

Dynamics of the vertical transfer parameters of ChNPP release radionuclides in soils, calculated with use of the convective-diffusional and quasi-diffusional models of transfer, is discussed. Values of ^{137}Cs quasi-diffusion coefficient vary from $(1\text{-}4)\cdot10^{-9}$ cm^2c^{-1} for mineral automorphous soils (soddy-podsolic loamy-sand, grey soils etc) to $2\cdot10^{-8}$ cm^2c^{-1} and higher for organic hydromorphous soils (peaty-boggy, peaty and similar).

Parameters of diffusional and directional transfer of ^{90}Sr and ^{137}Cs in initial water-soluble form as well as directional transfer of fuel particles in some soils, typical for the Ukrainian Polesje, was studied in model column experiments. It was shown that diffusion coefficients of radionuclides, depending of soils moisture content vary in a following limits: ^{90}Sr - soddy-podzolic sandy soil (SS): $(2.1\text{-}5.1)\cdot10^{-7}$ cm^2c^{-1}, soddy-podzolic loamy-sand soil (LSS): $(1.0\text{-}1.7)\cdot10^{-7}$ cm^2c^{-1}, peaty soil (PS): $(0.06\text{-}1.3)\cdot10^{-7}$ cm^2c^{-1}; ^{137}Cs - soddy-podzolic sandy soil: $(0.5\text{-}1.8)\cdot10^{-8}$ cm^2c^{-1}, soddy-podzolic loamy-sand soil: $(0.9\text{-}1.4)\cdot10^{-8}$ cm^2c^{-1}, peaty soil: $(0.17\text{-}1.3)\cdot10^{-8}$ cm^2c^{-1} (Table II). The rate of directional transfer of fuel particles is much lower than that for radionuclides, introduced in soil column in the

TABLE II. Diffusion coefficients of ^{137}Cs, ^{90}Sr and ^{239}Pu in different soils, depending on their moisture content

Soil	Radionuclide	$D^a\cdot10^{-8}$, $\cdot cm^2/c$ with moisture content, % of total moisture content		
		30	60	100
SS	Strontium-90	20.0±1.0	26.0±3.0	35.0±5.0
LSS	Strontium-90	9.0±1.0	16.0±3.0	15.0±1.0
PS	Strontium-90	0.6±0.1	3.7±0.2	13.0±1.0
SS	Caesium-137	0.50±0.04	0.70±0.15	0.9±0.4
LSS	Caesium-137	0.8±0.1	1.0±0.1	1.7±0.5
LS[b]	Caesium-137	n/determ.	n/determ	0.28±0.08
ChL[b]	Caesium-137	0.27±0.10	0.63±0.20	n/determ.
PS	Caesium-137	1.7±0.3	7.9±3.0	31.6±5.0
LSS	Plutonium-239	0.081±0.020	0.077±0.023	0.19±0.03
ChL	Plutonium-239	0.067±0.022	0.069±0.07	n/determ.
LS	Plutonium-239	n/determ.	n/determ.	0.086±0.020

[a]D - diffusion coefficient; [b]LS - soddy-podsolic loamy soil; [b]ChL - loamy chernozem.

water-soluble form. Values of the effective quasi-diffusion coefficient of fuel particles with real size distribution on 1988 (place of sample selection - R=3 km, W) are: for soddy-podzolic sandy soil: 3.0 $\cdot 10^{-10}$ cm^2c^{-1}, for soddy-podzolic loamy-sand soil: 4.0 $\cdot 10^{-10}$ cm^2c^{-1}, for peaty soil: 2.3 $\cdot 10^{-10}$ cm^2c^{-1}. Values of transfer parameters of Plutonium isotopes are closer to those for ^{137}Cs and are depend on destruction level of fuel in particular soil conditions.

Classification of Ukrainian Polessye soils according to migrative mobility of ^{137}Cs in these ones was proposed.

3.4. Modelling and forecast

Mathematical model of radionuclides transfer in soil profile is developed. Model allows to take into account the role of physico-chemical forms of radioactive fallout, of various forms of radionuclide transfer in soil profile (radionuclides in the matrix of fuel particles; radionuclides in soil solution in the form of free ions and complex compounds; sorbed form of radionuclides) and their transformation in the redistribution of radionuclides in soil profile [8].

$$\frac{\partial C1(x,t)}{\partial t} = \frac{\partial}{\partial x}\left[D(x,t)\frac{\partial C1x,t)}{\partial x}\right] - V(x,t)\frac{\partial C1(x,t)}{\partial x} - \left(b(x,t)+g(x,t)\right)C1(x,t) - \frac{b(x,t)C1(x,t)}{Kd(x,t)} + a(x,t)C4(x,t) - \lambda C1(x,t)$$

$$\frac{\partial C2(x,t)}{\partial t} = \frac{\partial}{\partial x}\left[D(x,t)\frac{\partial C2(x,t)}{\partial x}\right] - V(x,t)\frac{\partial C2(x,t)}{\partial x} + g(x,t)C1(x,t) - \lambda C2(x,t)$$

$$\frac{\partial C3(x,t)}{\partial t} = b(x,t)\left(C1(x,t) - \frac{C3(x,t)}{Kd(x,t)}\right) + \lambda C3(x,t) \qquad (1)$$

$$\frac{\partial C4(x,t)}{\partial t} = \frac{\partial}{\partial x}\left[D4(x,t)\frac{\partial C4(x,t)}{\partial x}\right] - (a(x,t)+\lambda)C4(x,t)$$

where: $C_1(x,t)$ - concentration of radionuclide in soil in the form of free ions at the depth x at the moment t; $C_2(x,t)$ - concentration of radionuclide in soil in the form of soluble complex compounds at the depth x at the moment t; $C_3(x,t)$ - concentration of sorbed radionuclide forms in soil at the depth x at the moment t; $C_4(x,t)$ - concentration in soil of radionuclide in fuel particles at the depth x ast the moment t; D(x,t) - effective diffusion coefficient of soluble forms at the depth x at the moment t; V(x,t) - effective velocity of convective transfer of radionuclide with soil moisture at the depth x at the moment t; $D_4(x,t)$-effective diffusion coeffiecient of fuel particles at the depth x at the moment t; b(x,t) - intensity of sorption of soluble radionuclide forms by soil at the depth x at the moment t; a(x,t) - intensity of fuel particles festruction at the depth x at the moment t; λ - constant of decay. Total concentration of all considered radionuclide forms in the soil layer C(x,t) is described by the ratio:

$$C(x,t) = C_1(x,t) + C_2(x,t) + C_3(x,t) + C_4(x,t).$$

The example of model application for the calculation of predictive assessments of ^{90}Sr, ^{137}Cs and $^{239,240}Pu$ redistribution in the profile of soddy-podzolic sandy-loamy soil on the fuel track of ChNPP fallout (fallow land), is presented below (Fig....., Tables). Fallout characteristics: ~100% of ^{90}Sr and $^{239,240}Pu$ and about 80% of ^{137}Cs were within fuel particles. Parameters were estimated with the condition that the values of V, D_4 and a are the same for all radionuclides. The values of the mentioned parameters are estimated as follows: V=0.86±0.02cm/year, D_4 =0.06±0.02cm^2/year and a=0.06±0.01 1/year. The values of other parameters are presented below.

TABLE III. Parameters of transfer model

Radionuclide	D, cm^2/year	b, 1/year	g, 1/year	K_d cm^3/g
^{90}Sr	30±7	0.4±0.3	2.4±1.2	10
$^{239,240}Pu$	12.9±3.7	0.87±0.25	0.06±0.04	550
^{137}Cs	11.4±0.6	24.0±0.3	0.58±0.03	300

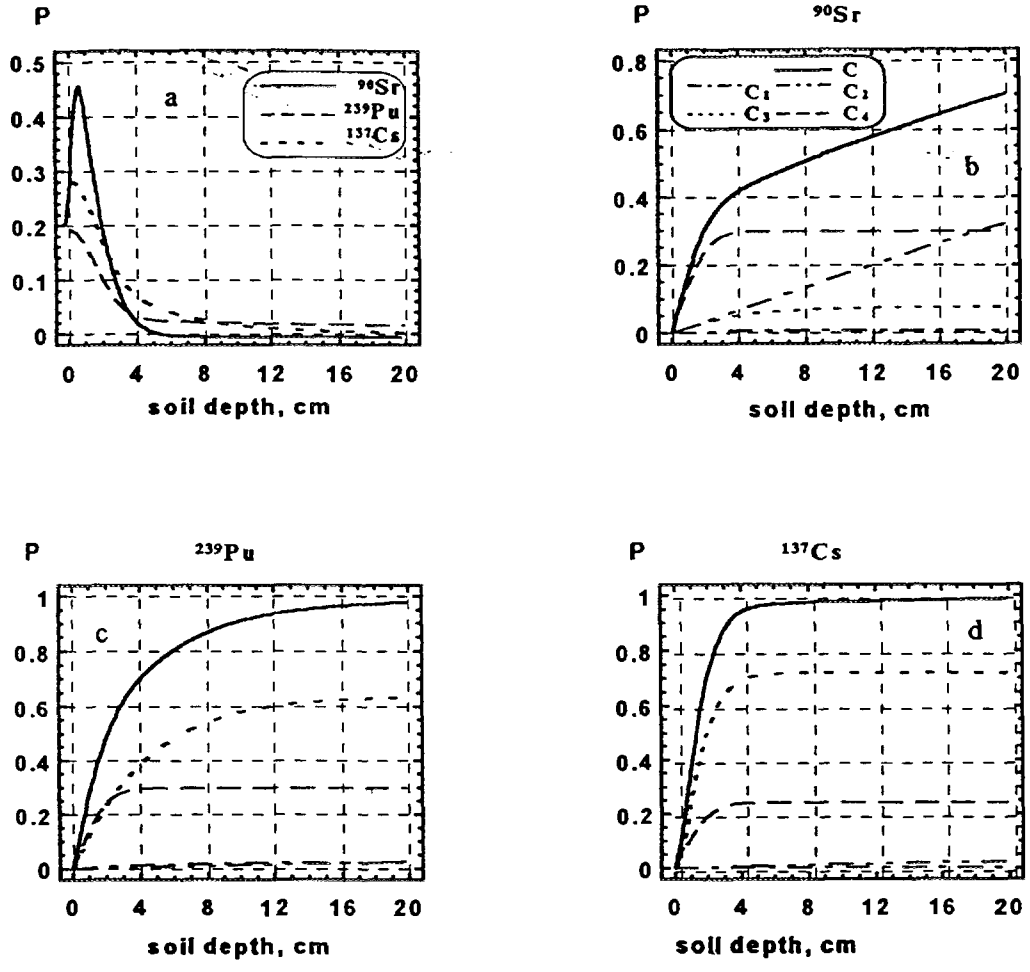

Fig.1 Forecast (on 2006) of the distribution profiles P(x,t) of ^{90}Sr, ^{239,240} Pu and ^{137}Cs in soddy-podsolic loamy-sand soil: a - total distribution (differencial curve); b, c, d - distribution profiles of considered forms of radionuclides transfer (integral curve)

Fig.1 demonstrates in differential form predictive (for 2006) profiles of distribution P(x,20) of ^{90}Sr, ^{239,240} Pu and ^{137}Cs in the soil of the plot (gross content of a specific radionuclide), on Figures 2b, 2c and 2d (for ^{90}Sr, ^{239,240} Pu and ^{137}Cs, respecttively) the profiles of distribution in soil of the considered within the model forms of radionuclides transfer are presented in integral form. Below the predicted values of ablsolute portions of various forms of radionuclides transfer, included to the transfer model, are presented (Table IV).

TABLE IV. Predicted values of ablsolute portions of various forms of radionuclides transfer, included to the transfer model

Form of radionuclide	^{90}Sr	^{239,240} Pu	^{137}Cs
Ion-exchangeable, C1	0.62	0.04	0.035
Watersoluble complex compounds, C2	0.008	0.02	0.003
Sorbed forms, C3	0.072	0.64	0.721
Fuel particles, C4	0.30	0.30	0.241

TABLE V. Migration ability of ^{137}Cs in typical soils of Ukrainian Polesye

Soils	Granulometricc omposition	M[a], $\cdot 10^{-5}$ cm$^2\cdot$s^{-1}	D[b], $\cdot 10^{-5}$ cm$^2\cdot$s^{-1}	V_k[b] $\cdot 10^{-5}$ cm$\cdot$s^{-1}
Organic hydromorphous soils (peat-boggy, peaty etc.)		> 2.0	0.5 - 1.2	0.3 - 1.0
Meadow gley and gleed, meadow and soddy carbonate, soddy gley and gleed	sandy	1.5 - 2.0	0.3 - 0.8	0.3 - 1.7
Soddy gley and gleed	sandy-loam, loamy-sand	1.0 - 1.5	0.8 - 1.0	0.2 - 0.5
Soddy-podsolic, soddy-podsolic gley, weakly humous sand	sandy	0.4 - 1.0	0.3 - 0.7	0.3 - 0.5
Soddy-podsolic, grey podsoled etc.	sandy-loam, loamy-sand, loamy	< 0.4	< 0.4	0.1 - 0.3

[a] - M - quasi-diffusion coefficient, calculated by one-component model of transfer; [b] - D, V_k - diffusion coefficient and directional transfer velocity, respectively, calculated by convective-diffusional model of transfer.

TABLE VI. Effective half-time of ^{137}Cs residence in upper layers of soils

Soils	Granulometric composition	Effective half-time of ^{137}Cs residence in upper layers of soils	
		$T_{1/2}$(0-2 cm)	$T_{1/2}$(0-5 cm)
Organic hydromorphous soils (peat-boggy, peaty etc.)		5.5 ± 0.8 4.0 - 9.0	14.9 ± 1.7 11.2 - 21.6
Meadow gley and gleed, meadow and soddy carbonate, soddy gley and gleed	sandy	5.1 ± 1.3 3.2 - 11.0	12.7 ± 2.7 8.3 - 24.3
Soddy gley and gleed	sandy-loam, loamy-sand	6.6 ± 0.4 5.6 - 7.9	17.8 ± 0.8 15.7 - 20.3
Soddy-podsolic, soddy-podsolic gley, weakly humous sand	sandy	8.3 ± 0.7 6.7 - 11.0	20.3 ± 1.1 17.5 - 24.3
Soddy-podsolic, grey podsoled etc	sandy-loam, loamy-sand, loamy	11.1 ± 0.5 10.0 - 12.7	24.4 ± 0.3 23.0 - 26.0

Results of *in situ* observation of ^{137}Cs vertical re-distribution in soils profile at various tracks of fallout in combination with results of the model experiments are permitted to classify the soils of contaminated territory of Ukrainian Polesye according to migration ability of radiocaesium (Table V). From point of view of migration ability most "critical" are the hydromorphous organic soils of Ukrainian Polesye.

Some forecast assessments of radionuclides transfer in soils are calculated (ecological and effective half-time of the residence in upper layers of soils, dynamics of internal dose rate, formed by Caesium radioisotopes etc.- Table VI, Fig.2). Values of the effective half-time of ^{137}Cs and ^{90}Sr residence in upper 5-cm layer of undisturbed soils vary, respectively, from 8-14 and 20-22 years for organic hydromorphous soils to 20-22 and 6-8 years for mineral automorphous soils, the same parameters for ploughed layer of disturbed soils are, respectively, 16-18 and 24-26 years for organic hydromorphous soils and 22-24 and 12-14 years for mineral automorphous soils.

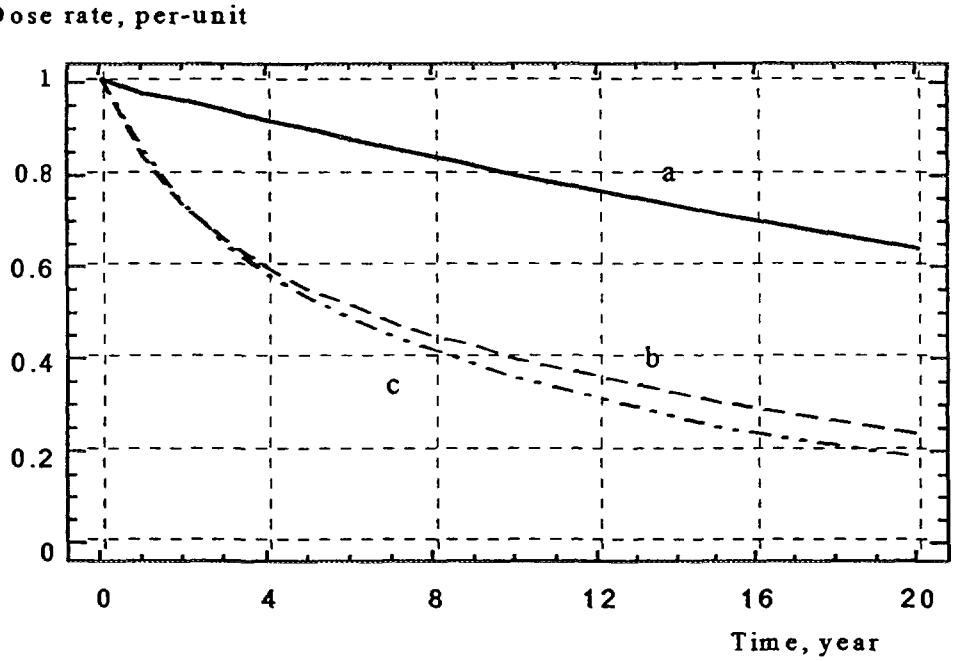

Fig.2 Calculated curves of dynamics of gamma-irradiation dose rate, formed by ^{137}Cs, under soil surface (H=1 m): a - without consideration of radionuclide vertical transfer; b, c - with consideration of radionuclide vertical transfer: b - in soddy-podsolic sandy-loam soil; c - in peaty soil.

Comparable assessments of the intensity of ^{137}Cs vertical transfer in soils and half-life of this radionuclide were demonstrated.

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