Бюллетень Почвенного института им. В.В. Докучаева. 2019. Вып. 97.
Dokuchaev Soil Bulletin, 2019, 97

UDC 631.4:581.1
DOI: 10.19047/0136-1694-2019-97-165-180

Cite this article as:
Pakshina S.M., Harkevich L.P., Belous N.M., Smolsky E.V., Regularities of migration ^{137}Cs in the alluvial soil, Dokuchaev Soil Bulletin, 2019, V. 97, pp. 165-180, DOI: 10.19047/0136-1694-2019-97-165-180

REGULARITIES OF MIGRATION ^{137}CS IN THE ALLUVIAL SOIL

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Received 28.02.2019, *Revised* 08.04.2019, *Accepted* 28.05.2019

This paper examines the patterns of ^{137}Cs migration in the alluvial soil of the central floodplain of the Iput River (Novozybkovsky district, Bryansk region). The effect of water filtration during flooding, bioremoval by plants and ^{137}Cs radioactive decay on the migration process of ^{137}Cs ions in the soil was studied. The contribution of radioactive decay, filtration of water and bioremoval to the total removal of ^{137}Cs from the soil during the period from 1994 to 2007 was 50–79, 20–50, 0.3–2.2 % correspondingly, depending on the treatment method, the dose of mineral fertilizers and the ratio of nutrients. It was found that the increased removal of ^{137}Cs from the soil layer of the floodplain during double-depth plowing, compared with disking and natural grass stands, is determined by a lower Peclet number, which indicates the prevalence of convective ^{137}Cs transfer in contrast to diffusion in the total solution flow.

Keywords: ^{137}Cs, migration, regularity, radioactive decay, biocarrying out, water filtration.

INTRODUCTION

The work examines the effect of radioactive decay, water filtration during flooding and biocarring out on the ^{137}Cs migration process in root-inhabited alluvial soil layer of a natural meadow, along with at its surface-level and radical improvement.

Currently we know the patterns of soil macronutrients moving and distribution at the absence of vegetation. The work (Volobuev, 1975) describes two patterns of salt migration in soil, which were obtained by generalizing and analyzing numerous data from field-experimental laboratory research on saline soils washing.

The first regularity connects salt mass with the time of salinization – desalinization soil processes and appears as follow:

$$S_t = S_0 \exp(\pm \beta t), \qquad (1)$$

where **t** – time required for soil salt content change from the initial value of S_0 to the value of S_t, **β** – constant for certain migration process conditions, signs “±” refer correspondently to soil salinization-desalinization processes.

The second regularity connects salt mass with the space where soil desalinization process takes place and appears as follow:

$$h = \mu \lg(S_i/S_0), \qquad (2)$$

where **h** – soil thickness divided into **i** layers, S_i – salt supply in the layers $i = 0 - h$, S_0 – salt supply in the surface layer, **μ** – a parameter characterizing filtration ability and soil drainage degree. The value h depends on the amount of filtered moisture (**Q**). Therefore, the value of μ is derived from the function graph $\lg (S_i/S_0) = f(Q)$, with a linear directly proportional view. Parameters **β** and **μ** are constant values for a particular soil, since the values of S_0, S_t, S_i, S_h have a finite value (Volobuev, 1975). The paper (Borzdyko, 2000) has shown for the first time that equation (2) is also correct when describing ^{137}Cs activity distribution over the soil depth at the absence of vegetation.

The papers (Pakshina, 1985, 1989) provide a theoretical justification for the dependencies (1), (2), constructing a model that includes both diffusion and convective ion flows over the soil pores along the solution flow and also a diffusion ion flow in the electrostatic field around the charged pore walls, perpendicularly directed to the pore walls, which describes the ion exchange on the soil-absorption complex (SAC). After the equation was solved for the cases of downward (Pakshina, 1985) and upward (Pakshina, 1989) ion flows the following equation was obtained:

$$C_t = C_0 \exp(-\lambda \upsilon t), \qquad (3)$$

where C_0, C_t – correspondently the initial and the final soil ions concentration, t – time required to reduce ion content from C_0 to C_t, υ – flow rate, λ – mass transfer parameter for certain conditions of the process.

We obtained a link between the parameters β, μ, and λ, which is determined by the following relations:

$$\beta = \lambda \upsilon, \qquad \mu = 1/\lambda. \qquad (4)$$

The model made it possible to decipher the content of parameter λ and express it in a formula that includes only physical values:

$$\lambda = 1.8 \cdot 10^3 \cdot \text{ЕКО} \cdot Pe \sqrt{\frac{Z_1 + Z_2}{2}} / S \cdot T, \qquad (5)$$

where **CEC** – cation exchange capacity, mEq/100 g of soil, **S** – specific soil surface, m^2/g, **T** – absolute soil temperature, **Pe** – Peclet parameter equal to $D/\upsilon r$. **D** – ion diffusion coefficient, υ – solution flow rate, **r** – pore radius, Z_1, Z_2 – salt anion and cation valence (Pakshina, 1985, 1989).

Considering equation (1) the paper (Pakshina, 1994) derived a formula that relates the density of ^{137}Cs soil pollution to the time, which changes under the influence of radioactive decay, water filtration through the soil, biocarrying out and appears as follow:

$$A_к = A_0 \left(e^{-\lambda_1 t} - e^{-\lambda_2 Q_t} - n e^{-\lambda_3 \Sigma в E_т}\right). \qquad (6)$$

Where the first, the second and the third members relate to radioactive decay, water filtration and biocarrying out; λ_1, λ_2, λ_3 – radioactive decay constant, ion mass transfer parameters under moisture filtration and radionuclide biocarrying out by plant root system; **Q**, $\Sigma в E_т$ – amount of moisture filtered through the soil and transpiration, respectively. A_0, $A_к$ – the initial and the final density of soil layer contamination, $A_0 e^{-\lambda_1 t}$ – remaining density of layer contamination after ^{137}Cs radioactive decay for the period **t**, $A_0 e^{-\lambda_2 Q_t}$ – ^{137}Cs removal under water filtration, $A_0 n e^{-\lambda_3 \Sigma в E_т}$ – biocarrying out of plant available ^{137}Cs, **n** – available ^{137}Cs fraction of total activity.

The model (6) describes ^{137}Cs migration from a certain soil layer by three mass transfer mechanisms over time.

The aim of this work is to study the applicability of formula (6) to the process of ^{137}Cs migration in root zone profile with perennial grasses vegetation grown in a floodplain.

MATERIALS AND METHODS

The study of ^{137}Cs migration patterns in root zone profile of perennial grasses was carried out on a meadow-type plot of the central floodplain of the Iput River (Novozybkovsky District, Bryansk Region) for 14 years (1994–2007).

The experimental plot soil is represented by alluvial meadow, low efficient, medium in humus content sandy soil on sandy-loam alluvium and has the following profile structure: Ah (0–4), A (4–18), B_1 (18–40), B_g (40–60), C_g (60–90).

The soil agrochemical properties for experimental period ranged in the following values intervals: pH_{KCl} – 5.2–5.43; hydrolytic acidity – 2.78–2.43 mEq/100g of soil; total absorbed bases – 10.1–12.24 mEq/100g of soil; humus content is 3.11–3.21 % (according to Tyurin); labile phosphorus and exchange potassium content is 121–135 and 50–69 mg/kg, respectively (according to Kirsanov).

The pollution density of the experimental plot averaged 1221–1554 kBq/m^2. The experimental plot flooding during the spring flood lasted 20–22 days.

The researched objects were natural grass stands and seeded bluegrass crop mixture under floodplain treatment by disking and double-depth plowing. The grain grass mixture was of the following composition: *Festuca pratensis*, *Phleum pretense*, *Bromopsis inermis*, *Phalaris arundinacea*, *Alopecurus pratensis*.

Five options of fertilizer application were used on each background of floodplain treatment and natural grass stand: 1 – Control, 2 – $N_{120}P_{90}K_{120}$, 3 – $N_{120}P_{90}K_{240}$, 4 – $N_{180}P_{120}K_{180}$, 5 – $N_{180}P_{120}K_{360}$.

Nitrogen (ammonium nitrate) and potash (potassium chloride) fertilizers were applied at two doses: half of the calculated dose – for the 1st cutting, the second half – for the 2nd cutting. Phosphorus fertilizers (simple granular superphosphate) were applied as a single dose in spring.

The sowing plot area was 63 m^2, the harvesting area – 24 m^2; the experiment was repeated threefold. The perennial grass yield was calculated by yield from each harvest plot and trial sheaf selection. Two cuttings were done per year (the first from June, 1st to 10th, the second from August, 23rd to 30th). The specific ^{137}Cs activity in soil and plant samples was measured on Gamma Plus universal spectrometric complex (Research and Production Enterprise "Dosa", Russia); the measurement error did not exceed 10 %.

In the experimental year (1994) and in 2007 the samples were taken from each soil layer equal to 5 cm up to the depth equal to 60 cm to determine the density of ^{137}Cs contamination (Kharkevich, 2011).

Crop transpiration was calculated by the formula (Penman, 1972), evaporation – by the Ivanov's formula (Ivanov, 1954), ^{137}Cs radioactive decay by the Rutherford's formula (1903). To calculate the biocarrying out of the parameter λ_3 we used an equation derived from field experiment data that appears as follow:

$$\mathbf{Ln(Ak / Ai) = \lambda \Sigma вEт,} \qquad (7)$$

where **Ak**, **Ai** – respectively, the specific ^{137}Cs activity on the control and on the variant I (Pakshina et al., 2017). To calculate the ^{137}Cs mass transfer parameter in the soil under water filtration, the equation (5) was used, where **ЕКО** = 15.3 meq/100g of soil, **S** = 31.3 m2/g, **T** = 279.6 K, **Pe** = 0.4 (natural grass stand), **Pe** = 0.35 (disking), **Pe** = 0.25 (double-depth plowing). Under water filtering both ^{137}Cs forms accessible to the plant root system and the colloidal particles containing ^{137}Cs compounds of low solubility are removed from the soil (Skovorodnikova, 2005). When calculating the ^{137}Cs biocarrying out it was considered that the activity of water-soluble and exchanging ^{137}Cs forms in gleyed soils amounted to 5 % of the total ^{137}Cs activity (Skovorodnikova, 2005).

To calculate volatility, atmospheric moisture deficiency, moisture filtration from the soil layer equal to 60 cm, we used data obtained from the "Krasnaya Gora" meteorological station, the closest to the experiment site.

Each year (from 1994 to 2007) was divided into three periods, including the months: I – IV, V – IX, X – XII. In each period the average monthly precipitation, volatility, and atmospheric moisture deficit were calculated. The atmospheric moisture deficit for 14 years was observed only in the V – IX months, which amounted to 1075 mm of water. The amount of precipitation for 14 years was 7197 mm of water, volatility – 6163 mm, humidity factor (HF) – 1.17, which characterizes flushing type of soil water regime. Flood water filtration according to the data of Bryanskgiprovodkhoz is 26 % of the total annual precipitation amount, which was 134 mm in the considered period (1994–2007). The filtration rate of flood waters was 0.8×10^{-7} m/s, which indicates high water-resistance.

RESULTS AND DISCUSSION

Table 1 presents hay yield data (c/ha) and perennial grasses transpiration (mm) on average for two cuttings and on average for the growing season from 1994 to 2007.

Table 1. The average hay yield and transpiration of perennial grasses for the growing season from 1994 to 2007

Variant	Natural grass stand			Sowed grass stand					
				Disking			Plowing		
	Y	∑Grt	TC	Y	∑Grt	TC	Y	∑Grt	TC
Control	23.9	183	766	31.2	239	766	32.9	252	766
$N_{120}P_{90}K_{120}$	104.3	798	765	128.4	982	765	139.6	1068	765
$N_{120}P_{90}K_{240}$	94.7	724	764	118.0	903	765	120.8	924	765
$N_{180}P_{120}K_{180}$	121.5	929	765	140.0	1071	765	141.4	1082	765
$N_{180}P_{120}K_{360}$	107.0	819	765	121.8	932	765	126.0	964	765

Note. Y – hay yield in total for two cuttings, c/ha; ∑Grt – transpiration during the growing season, mm; TC – transpiration coefficient equal to the consumption of transpiration moisture (t) for 1 ton of air-dry phytomass formation.

It has been proved that when a complete mineral fertilizer is applied, the herbs yield depends both on potassium-nitrogen dose ratio and on phosphorus dose. A twofold increase in the K : N ratio with an equal phosphorus dose causes its deficiency and leads to a decrease in transpiration and yield (Table 1). Transpiration coefficient (TC) does not depend on fertilizer dose and tillage method; it is equal on all experimental variants. The independence of TC value on mineral fertilizer dose had been earlier noted in Shatilov's work (Shatilov, 1978). High TC values are due to the groundwater use during grass vegetation period at their similar level in the central floodplain. Table 2 shows the data on the density of ^{137}Cs soil contamination in root zone profile of perennial grasses in 1994.

Table 2. The distribution of soil contamination (^{137}Cs) density in the root zone profile in 1994 (in the year of trial establishment)

Soil layer, cm / Variant	0–20		21–30		31–40	
	Ai	%	Ai	%	Ai	%
Natural grass stand	1 339.0	98.6	14.8	1.1	4.9	0.3
Sowed grass stand (soil treatment method – disking)	1 311.0	99.0	10.0	0.7	3.7	0.3
Sowed grass stand (soil treatment method – double-depth plowing)	1 323.4	95.4	54.7	4.0	8.9	0.6

Note. Ai is the density of contamination of ^{137}Cs layer, kBq / m^2.

Eight years after Chernobyl accident the density of ^{137}Cs contamination of the alluvial soil layer equal to 0–20 cm on natural grass and under the surface-level floodplain improvement (disking) decreased by only 1 % of the initial pollution. After a radical improvement (double-depth plowing), which led to soil mixing to the depth of 45 cm, the density of ^{137}Cs contamination of 0–20 cm soil layer decreased by 5 %. The ^{137}Cs deposited in 1986 moved to the depth of 40 cm by 1994. Table 3 presents the data on the changes in the ^{137}Cs contamination density in the root zone profile of the alluvial soil in 2007, depending on the experiment variant (the trial experiment in 1994). About 98 % of ^{137}Cs from the layer of 0–60 cm remains at the control in alluvial soil layer equal to 0–20 cm.

Table 3. The distribution of soil contamination (^{137}Cs) density in the root zone profile in various treatments (2007)

Soil layer, cm Variant	0–20		21–30		31–40		41–50		51–60	
	Ai	%	Ai	%	Ai	%	Ai	%	Ai	%
Natural grass stand										
Control	788.8	97.5	18.4	2.1	2.5	0.3	0.5	0.07	0.32	0.06
$N_{120}P_{90}K_{120}$	784.3	92.8	55.2	6.2	6.8	0.7	1.3	0.14	0.48	0.06
$N_{120}P_{90}K_{240}$	805.9	91.3	70.9	7.5	7.6	0.8	2.9	0.3	0.72	0.07
$N_{180}P_{120}K_{180}$	790.4	91.7	56.5	6.2	14.7	1.6	3.9	0.4	0.92	0.1
$N_{180}P_{120}K_{360}$	719.6	86.8	89.7	10.2	20.3	2.1	6.4	0.7	1.52	0.16
Sowed grass stand (soil treatment – disking)										
Control	803.2	96.6	24.0	3.0	1.6	0.2	1.0	0.14	0.48	0.06
$N_{120}P_{90}K_{120}$	666.9	93.1	46.7	6.2	4.2	0.5	1.1	0.15	0.41	0.05
$N_{120}P_{90}K_{240}$	762.6	94.7	33.6	3.9	9.0	1.0	9.2	0.35	0.56	0.05
$N_{180}P_{120}K_{180}$	777.0	97.8	16.9	2.0	1.1	0.1	0.4	0.05	0.32	0.04
$N_{180}P_{120}K_{360}$	631.0	91.5	50.9	6.9	8.1	1.1	2.4	0.3	0.96	0.12
Sowed grass stand (soil treatment – double-depth plowing)										
Control	414.5	61.7	255.8	36.2	15.1	2.0	0.7	0.07	0.46	0.05
$N_{120}P_{90}K_{120}$	311.1	49.6	278.0	42.3	56.1	7.9	0.9	0.14	0.32	0.06
$N_{120}P_{90}K_{240}$	380.4	60.3	258.6	39.1	2.8	0.4	1.0	0.13	0.56	0.07
$N_{180}P_{120}K_{180}$	492.1	68.6	223.6	29.6	13.4	1.7	0.8	0.08	0.32	0.02
$N_{180}P_{120}K_{360}$	427.3	68.6	200.9	20.8	3.3	0.4	1.0	0.14	0.36	0.06

Note. Ai is the density of ^{137}Cs contamination layer of 0–60 cm (kBq/m^2).

Application of mineral fertilizer accelerated ^{137}Cs desorption process from the turf and its removal from the layer equal to 0–20 cm. In this layer ^{137}Cs activity averaged 91 % of the layer 0–60 cm activity.

Turf grinding at disking and its leaving on the surface did not have a significant effect on ^{137}Cs removal compared to natural grass stand. ^{137}Cs activity in the soil layer of 0–20 cm during the control and with the addition of NPK was correspondently 97 % and 94 % of 0–60 cm layer activity.

Turf grinding and mixing its residues with other horizons under two-depth plowing toward the depth of 45 cm increased ^{137}Cs removal from 0–20 cm layer during the control and the variants with NPK application. ^{137}Cs activity in this layer averaged 62 % of ^{137}Cs activity in 0–60 cm layer. In all variants with different tillages ^{137}Cs presents in the layer of 50–60 cm adjoining with the gleyed parent material Cg (60–90).

Table 4 presents a comparative assessment of At values (t = 14 years) calculated by equation (6) with the experimental values. The experimental values included a pollution density of 0–60 cm soil layer in 1994 and 2007, a decrease in the pollution density over 14 years and experimental values of herbs biomass biocarrying out. ^{137}Cs biocarrying out from the soil was determined by multiplying hay yield for 14 years by its ^{137}Cs specific activity. Calculating ^{137}Cs biocarrying out from the soil by herbs biomass, we used the value $\lambda_3\Sigma\text{вЕт}$ obtained by formula (7).

To calculate ^{137}Cs removal from the soil during floods, we found $\lambda 2$ values by formula (5) and Pe value for different soil treatment methods. The research (Pakshina et al., 2018) shows that there is a linear inverse proportion between Fe values and relative transpiration. The figure shows the dependence of Pe values on the relative transpiration of three bluegrass crops. As the figure represents, the Pe number decreases with an increase in soil water losses.

With relative transpiration values equal to 2 and 2.5, Pe number is 0.4 and 0.25 respectively. The evaporation in April during floods averaged 53.9 mm over the course of 14 years. Hence, $Q/\Sigma E_0$ = 2.5, here Q = 134 mm. At maximum water losses for filtration Pe number is 0.25, and at minimum ones – 0.4.

Table 4. Quantitative assessment of ^{137}Cs removal from 0–60 cm soil layer for the period from 1994 to 2007

Variant	1994	2007	^{137}Cs removal Ao-Af		^{137} Cs removal process, experimental and calculated values					
	Ao	Af	Exp.	Calc.	Radioactive decay		Biocarrying out		Water filtration	
					A_1	Ao-At	Exp.	Calc.	Exp.	Calc.
Natural grass stand										
Control	1 359	810	549		985	374	13.2		161	
$N_{120}P_{90}K_{120}$		848	511	526			22.1	26	115	128
$N_{120}P_{90}K_{240}$		888	471				5.6	7.2	91	
$N_{180}P_{120}K_{180}$		856	503				16.8	17	112	
$N_{180}P_{120}K_{360}$		847	512				3.5	4	134	
Sowed grass stand (soil treatment – disking)										
Control	1 325	830	495		961	364	13.7		117	
$N_{120}P_{90}K_{120}$		719	606	511			20.3	23.8	221	169
$N_{120}P_{90}K_{240}$		809	516				4.2	5.6	147	
$N_{180}P_{120}K_{180}$		796	529				9.1	9.8	155	
$N_{180}P_{120}K_{360}$		693	632				2.5	3.1	265	
Sowed grass stand (soil treatment – double-depth plowing)										
Control	1 387	687	700		1006	381	9.1		310	
$N_{120}P_{90}K_{120}$		646	741	724			13.2	23.6	347	319
$N_{120}P_{90}K_{240}$		643	744				2.9	6.2	360	
$N_{180}P_{120}K_{180}$		730	357				7.4	13.2	269	
$N_{180}P_{120}K_{360}$		633	754				1.7	3.4	371	

Note. A0, Af (exp.), Af (calc.) – respectively, the initial (1994) and final experimental and calculated according to the formula (6) for the layer of 0–60 cm soil pollution density kBq/m^2.

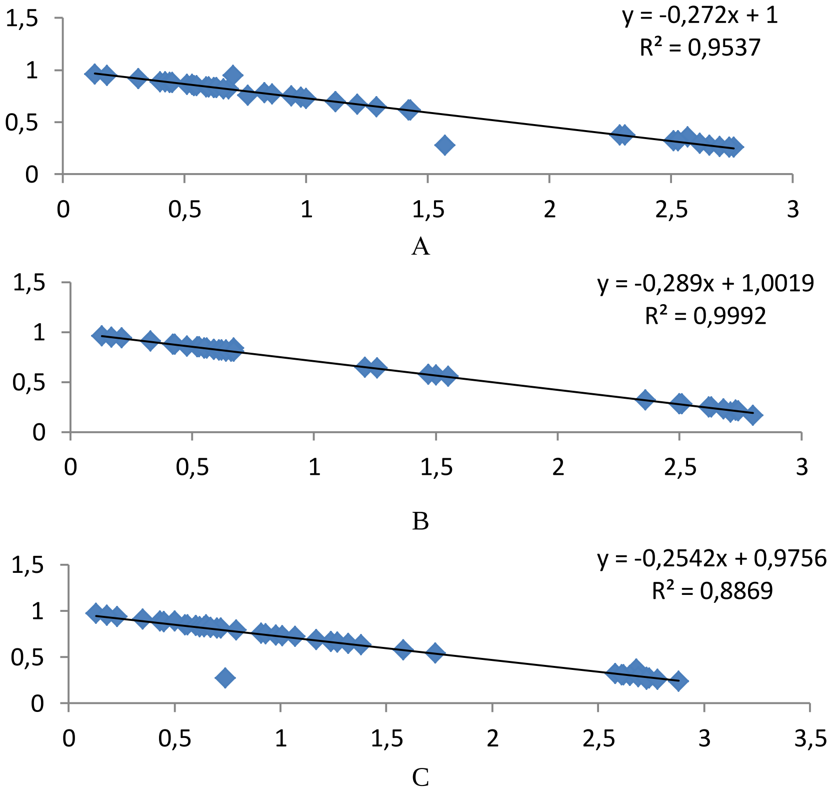

Figure. Correlation communication between Peclet's number of streams of soil moisture and a relative transpiration of crops of bluegrass herbs: A – *Dactylis glomerata*, B – *Festuca pratensis*, C – *Phalaris arundinacea*.

The figure shows the correlation communication between Peclet's number and a relative transpiration of crops of bluegrass herbs.

As it follows from the figure, with an increase in soil moisture consumption for grass crop transpiration or by soil moisture, the convective ion flow increases and Pe number decreases. With a decrease in moisture consumption by grass crops for transpiration the diffusion ion flow with the fluid flow increases altogether with Pe number.

As it follows from Table 4 the experimental values of ^{137}Cs biocarrying out from soil by grass phytomass and ^{137}Cs removal during flood water filtration through the soil layer of 0–60 cm coincided.

Quantitative assessment of ^{137}Cs removal from the root zone grass profile for the period of 1994–2007 revealed that biocarrying out contribution to the total ^{137}Cs removal is not significant and does not exceed 4 % at all variants. Mineral fertilizers have an important effect on ^{137}Cs biocarrying out from the soil. Particularly low ^{137}Cs biocarrying out is observed when NPK is applied at doses of K : N = 2.

^{137}Cs removal at water filtration through the soil layer is several times higher than biocarrying out. A particularly high ^{137}Cs filtration removal is observed at double-depth floodplain plowing.

Double-depth plowing, increasing the porosity, soil pore size, moisture flow rate and decreasing Pe number, increases ^{137}Cs removal into lower soil layers during flooding by 1.9 and 2.5 times, compared to disking and natural grass stand.

CONCLUSIONS

The analysis of the field-experimental data on the change in ^{137}Cs contamination density in the root zone soil profile of perennial grasses gradually allows us to identify the following:

– Decreasing ^{137}Cs activity density of the root zone profile depends on each process contribution. The contribution of radioactive decay, water infiltration, ^{137}Cs biocarrying out from the soil over 14 years, respectively, amounted to: 50–79 %; 20–50 %; 0.3–2.2 %.

– Double-depth plowing increases the ^{137}Cs removal from the upper soil layer, equal to 0–60 cm in comparison with disking and the natural grass stand during flooding by 1.9 and 2.5 times correspondently.

– The experimentally confirmed formula linking ^{137}Cs activity in root zone profile to the time, which has the following form: $\mathbf{At = A0(e\text{-}\lambda 1,\ t \times e\text{-}\lambda 2Q \times ne\text{-}\lambda 3\Sigma \text{в}E\text{т})}$, where the first, the second, the third terms belong respectively, to radioactive decay, water infiltration and ^{137}Cs biocarrying out from the soil.

– It is proved that the reason for ^{137}Cs increased removal from alluvial soils, compared to other soils of Bryansk Region, is the intensive ^{137} Cs migration during the period of annual floodings.

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