

Calculation of ^{137}Cs accumulation by phytomass of MOTLEY herbs

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Abstract

Aim: Large-scale development of soils contaminated with artificial radionuclides is accompanied by mandatory determination of ^{137}Cs accumulation factor by crop production. **Materials and Methods:** The paper proposes to replace the experimental determination of the accumulation coefficient by a numerical method, which was developed on the basis of ideas about the bioremoval of ions from the soil. **Results:** The calculated K_H values were estimated experimentally using the correlation-regression analysis, which revealed a close relationship between them. It has been established that the bioyield of ^{137}Cs depends on the relative transpiration of grass crops or the availability of soil moisture by the root system of plants. **Conclusion:** It was revealed that the process of ^{137}Cs adsorption by the root system of grasses is more intensive in the period from the renewal of vegetation to the first cut as compared with the period from the first to the second cut.

Key words: ^{137}Cs , accumulation coefficient, alluvial-meadow soil, bioremoval, correlation-regression analysis, meadow grasses, numerical method of calculating ^{137}Cs accumulation coefficient

INTRODUCTION

In agricultural radiology, they use widely the following empirical parameter - the factor of radionuclide accumulation from plant growing products (K_H). The accumulation coefficient is equal to the ratio of product radionuclide specific activity (Bq/kg) to the specific activity of soil (Bq/kg). K_H characterizes the intensity of radionuclide intake into the product during its root uptake from the soil.

The value of K_H is used to monitor the transition of ^{137}Cs from soil to plant,^[1] to evaluate the effectiveness of new meliorants, their doses,^[1,2] crop cultivation technologies,^[3] and the doses of mineral fertilizers.^[4,5]

To determine K_H , the field or the laboratory experiments are organized, which include the simultaneous of plant and soil sample selection, the measurement of the specific radionuclide activity in them. The sampling is carried out not <10 times in each experiment.

There is a known numerical method to estimate K_H , which makes it possible to replace the laborious experimental determination method.^[6] The numerical method of K_H radionuclide

calculation from crop production was developed taking into account the prevailing ideas about the bioremoval of ions from soil by plants. The process of plant bioremoval by plants includes three stages: The movement of ions to the surface of roots, the movement of ions from the outer surface into a root (primary absorption), and the movement of ions from the root to the stems and leaves (ion transport).^[7]

The process of ion transport in the soil under the influence of gravitational and capillary forces was first described in Volobuev^[8] in the form of the following formula:

$$S_t = S_0 \exp(-\beta t) \quad (1)$$

Where, S_0 and S_t are the initial and time t reserve of salts in a soil layer of a certain thickness, β - the constant for certain conditions of the process. The equation (1) was obtained by experimental data analysis of salt content in the thickness of wash grounds.^[8]

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The theoretical substantiation of salt motion equation in the soil profile was given in Pakshina.^[9,10] For this purpose, they developed the model that included not only the diffusion and convection flows of ions in the soil pores along the solution motion but also the diffusion flux of ions in the electrostatic field around the charged pore walls directed perpendicular to the pore walls, which describes the adsorption of ions on the soil-absorbing complex. During the solution of the equation for the cases of the descending^[9] and ascending^[10] ion fluxes, the following expression was obtained:

$$C_t = C_o \exp(-\lambda vt), \quad (2)$$

Where, C_o and C_t are the initial and final concentrations of ions in soil, v is the flow velocity, t is the time required to reduce the ion content from the value of C_o to C_p and λ is the constant for certain conditions of the process, $\lambda = \beta/v$.

It is important to note that the coefficient λ was expressed in the form of the formula that includes only physical quantities:

$$\lambda = 4\pi Ge Z_o Pe \sqrt{\frac{Z_1 + Z_2}{2}} / SEKT \quad (3)$$

Where, G is the capacity of the cation exchange, e is the electron charge, Z_o - is the charge of the potential-forming ion, $4p$ - the valence of the anion and the cation, respectively, Pe - the Peclet number equal to D/vr , D - the diffusion coefficient of the ion, v is the flow rate of the solution, r - the radius of pores, S - specific surface of soil, E - dielectric constant of solution, K - Boltzmann's constant, and T - absolute temperature.

Expressing the physical constants e , E , and K through their numerical values, the equation (3) is reduced to the following form:

$$\lambda = 1.8 \times 10^3 GPe \sqrt{\frac{Z_1 + Z_2}{2}} / ST, \text{m}^{-1} \quad (4)$$

The ions that move in the soil to the roots of plants with filtration, capillary, or film moisture, before reaching the surface of the root hair, overcome two electrostatic fields of different intensity by diffusion at the boundaries of the soil solution and root solution.

To describe the movement of an ion in a thin film of the solution located between the surface of the root hair and the soil, the equation has been solved that includes the diffusion of ion in the electrostatic field, in which the double electrical layers are closed and directed perpendicular to the surface of the root and soil. The solution of this equation has the following form:

$$C_k = C_p \exp[-(\phi_k - \phi_n)] \quad (5)$$

Where, C_k is the ion concentration on the root surface; C_p - the ion concentration in soil solution; and ϕ_k , ϕ_n - interfacial potentials

at the root solution and soil solution interface, respectively. Expressing the interphase potentials through the density of surface charges, the equation (5) acquires the following form:

$$C_k = C_p \exp[-(\lambda_k - \lambda_n)] \quad (6)$$

It follows from equation (6) that the bioremoval of ions from the soil is determined by the difference of the following constants: λ_k and λ_n .^[11]

Assuming that the migration losses of ^{137}Cs take place in the root zone of the soil, the activity of which during the growing season decreases by 0.988 times from the initial contamination due to radioactive decay, the total activity of ^{137}Cs carried out with the transpiration moisture of the crops from the soil made:

$$A_t = A_o \exp[-(\lambda_k - \lambda_n) \sum_B E_T] \quad (7)$$

Where, A_o , A_t - the activity of the soil root layer at the beginning and the end of vegetation, respectively, $\sum_B E_T$ - the transpiration during the vegetation season.

Then, the expression for the accumulation coefficient will take the following form:

$$K_H = \frac{A_o \exp \left[-(\lambda_k - \lambda_n) \sum_e E_T \right]}{A_o - A_o \exp \left[-(\lambda_k - \lambda_n) \sum_e E_T \right]} = 1 / \left[\frac{\exp(\lambda_k - \lambda_n)}{\sum_e E_T - 1} \right] \quad (8)$$

To calculate the value of λ equal to $\lambda_k - \lambda_n$, one can use experimentally obtained values of λ and calculated from the equation (4). The calculation of the experimental values of λ is carried out according to the formula derived from the experimental data, which has the following form:

$$\ln \frac{A_k}{A_i} = \lambda \sum_B E_T \quad (9)$$

Where, $\frac{A_k}{A_i}$ - the ratio of ^{137}Cs activity decrease in crop production (A_i) as compared to the control (A_k).^[12]

The purpose of this paper is to evaluate the numerical method to calculate the coefficient of ^{137}Cs accumulation and the possible ways of its use.

MATERIALS AND METHODS

The reliability of the numerical method for KH calculation was carried out on single-species crops of meadow grasses: *Dactylis glomerata* L., *Festuca pratensis* Huds., and *Phalaroides arundinacea* L. The experience was laid in

the central floodplain of the Iput River of Novozybkovsky District, Bryansk Region.

The soil of the experimental site is represented by alluvial meadow, low potent, medium humus, and sandy on sandy loamy alluvium and has the following structure of the profile: Ad (0–4), A1 (4–18), B1 (18–40), Bg (40–60), and Cg (60–90). The contamination density of the experimental section of ¹³⁷Cs during the study period was 493–872 kBq/m².

The soil was characterized by the following agrochemical properties: pHKCl - 5.3, hydrolytic acidity - 2.7 mg-eq. per 100 g of soil, the sum of the absorbed bases is 12.2 mg-eq. per 100 g of soil, the capacity of cation exchange is 14.4 mg-eq. per 100 g of soil, the degree of saturation with bases was 81.5%, humus content - 3.2% (according to Tyurin), mobile phosphorus – 620–840 mg/kg, and exchangeable potassium - 133–180 mg/kg (according to Kirsanov).

The scheme of the experiment included the following options: 1. Control - without the introduction of fertilizers; 2. N₉₀P₆₀K₉₀; 3. N₉₀P₆₀K₁₂₀; 4. N₉₀P₆₀K₁₅₀; 5. N₁₂₀P₆₀K₁₂₀; 6. N₁₂₀P₆₀K₁₅₀; and 7. N₁₂₀P₆₀K₁₈₀.

Ammonium nitrate, simple granulated superphosphate, and potassium chloride were used as fertilizers. Fertilizers were introduced annually: Nitrogen and potassium in two doses, half of the estimated dose for the first slope, the second half for the second slope, and phosphorus by the full dose at 1 time under the first slope.

The area of the seed plot is 63 m², the harvesting area is 24 m², and the repetition of the experiments is 3-fold.

The yield of perennial meadow grasses was taken into account by the method of continuous area harvesting and the selection of a test sheaf. Two cuttings were carried out per year (the first cut from June 1 to 10, the second one from August 23 to 30).

To determine the accumulation coefficient of ¹³⁷Cs by aboveground phytomass, the conjugated samples of plants and soils were selected from 1 m² in 3-fold repeatability on each variant. The specific activity of ¹³⁷Cs in the samples of soil and plants was measured on the universal spectrometric complex Gamma Plus (NPP “Doza,” Russia), the measurement error did not exceed 30%.

The transpiration of cultures during the growing season was calculated according to the following formula:

$$\sum_B E_T = \frac{0.4 K \phi_{ap} \sum_E Q_c}{L} \quad (10)$$

Where, $\sum_B Q_c$ - the sum of radiation balance daily values during the growing season, MJ/m²; $K\phi_{ap}$ - the coefficient

of photosynthetically active radiation (FAR) use, %; and L - specific heat of vaporization at air temperature during vegetation period, J/m³.^[13]

FAR use factor was calculated by the following formula:

$$K_{qpb} = \frac{Yq100}{\sum_e Q_{qb}} \quad (11)$$

Where, Y - the productivity of air-dry grass mass, kg/ha; q - the caloric content of herbs, J/kg; and $\sum_B Q_{qb}$ - the sum of the average daily FAR values during the vegetation season, J/kg.

The following equation was used to calculate FAR during the vegetative period:

$$\sum_B Q_{\phi} = 0.43 \sum_B S + 0.57 \sum_B D \quad (12)$$

Where, $\sum_B S$ is the sum of the direct radiation coming to the horizontal surface and $\sum_B D$ - the sum of scattered radiation during the vegetation season.

Positive daily sums of radiation balance ($\sum_B Q_k$) were found by the following formula:

$$\sum_B Q_k = \sum_B (S+D) - \sum_B R_k \quad (13)$$

Where, $\sum_B R_k$ is the sum of the reflected radiation during the vegetation period.^[14]

To calculate the daily sums of the radiation balance, the empirical coefficients were used calculated according to the data of Abakumova *et al.*^[15] The coefficients for April, May, June, July, and August were 0.78, 0.87, 0.90, 0.90, and 0.86, respectively.

The experimental values of K_H were compared with those calculated ones using the formula (8), the value of the bioremoval parameter (λ) obtained in the experiment and calculated by the formula (4).

RESULTS AND DISCUSSION

To determine ¹³⁷Cs accumulation coefficient by plant growing production (KH), it is required to measure the specific activity of soil and plant samples in a sufficient number of replicates of each sample measurement on the instruments with a certain error. For the numerical calculation of KH, it is necessary to have the value of ¹³⁷Cs bioremoval parameter (λ) from the soil and transpiration during the vegetation season ($\sum_B E_T$). If there are the results of experiments with a given culture, the calculation of λ is carried out according to the formula (9). For the calculation of $\sum_w E_T$ one can use formula (10) or the transpiration coefficient of culture during the cultivation on a given soil.

Table 1 shows the data $\sum_B E_T$ calculated by the formula (10).

As follows from Table 1, the transpiration of the meadow grasses cultivated on a specific soil depends on the type of culture and climatic conditions.

Figure 1 shows the correlation coefficients between the values of K_H ¹³⁷Cs calculated by formula (8) and the experimental values of K_H ¹³⁷Cs of air-dry overground mass of Pooideae herbs in different years. The bioremoval parameter was calculated using the formula (9).

It follows from Figure 1 that the accuracy of the prediction with the use of the numerical calculation method for the first and second slopes makes 91–97%, thus that the parameter λ , calculated from the experimental data (formula 9), reliably reflects the process of bioremoval of ions from the soil. According to formulas (3) and (4), the bioremoval parameter also includes the following indicators: The cation exchange capacity and the specific surface of the roots and soil, the temperature, the ion valence, and the Peclet number, which characterizes the relationship between diffusion and convective ion transfer in the solution stream.

Figure 2 demonstrates the graphs of the bioremoval parameter (λ) dependence, calculated by the formula (9), on the relative transpiration.

It follows from Figure 2 that a linear direct-proportional relationship between the parameter λ and relative transpiration is observed for the three species of grasses. It follows from the correlation equation that in the absence of soil moisture

cocksfoot, Randall, and lady's-laces has the following values of λ_0 : 15.22, 16.85, and 12.76 m^{-1} . These dependencies make it possible to calculate the Peclet number (Pe) for each culture.

Figure 3 shows the correlation between the number of Pe and the relative transpiration of Pooideae grasses.

As follows from Figure 3, with the increase of soil moisture availability in the root system of plants, determined by relative transpiration of grass sowing or soil moisture, the convective ion flux increases and the Pe number decreases. With the decrease of moisture availability to the root systems of grass crops, the diffusion flux of ions with the liquid flow increases and the number Pe increases.

At the same time, the performed studies of moisture reserves in 1 m soil layer at the end of vegetation period and relative transpiration during the vegetation period of various types of Pooideae cultures made it possible to establish that the following values of $\sum_B E_T / \sum_B E_0$: 0.7–0.85, 0.52–0.67, and 0.35–0.42 correspond to the soil hydrological constants PPV, VPK, and VZ.^[16] According to the correlation equations shown in Figure 3, these values of $\sum_B E_T / \sum_B E_0$, regardless of the type of culture, are equal to the following values of Pe number: 0.81–0.77, 0.86–0.82, and 0.91–0.89.

Thus, regardless of culture type, the soil-hydrological constants are characterized by definite values of the Pe number. Consequently, it became possible to use the formula (4) to forecast the contamination of crop production during the cultivation of new crops or varieties on radionuclide-contaminated soils.

Table 1: Transpiration of the Pooideae grasses (mm)

Variant	2009			2010			2011		
	1	2	3	1	2	3	1	2	3
Vegetation period until the first slope									
Control	50	53	56	122	117	124	90	98	96
$N_{45}P_{60}K_{45}$	179	193	198	447	446	466	401	438	452
$N_{45}P_{60}K_{60}$	180	201	204	474	421	478	405	442	456
$N_{45}P_{60}K_{75}$	194	201	212	481	486	488	415	477	479
$N_{60}P_{60}K_{60}$	206	244	221	466	469	471	442	458	470
$N_{60}P_{60}K_{75}$	227	236	253	479	483	486	443	470	477
$N_{60}P_{60}K_{90}$	229	248	277	491	498	512	479	480	486
Vegetation period until the second slope									
Control	29	30	31	56	60	64	43	41	45
$N_{45}K_{45}$	92	96	99	154	156	158	164	160	180
$N_{45}K_{60}$	97	99	103	166	167	169	171	167	185
$N_{45}K_{75}$	102	110	105	173	172	174	175	173	188
$N_{60}K_{60}$	105	112	115	170	175	180	187	188	200
$N_{60}K_{75}$	122	120	120	178	180	183	194	195	211
$N_{60}K_{90}$	124	122	182	187	188	189	197	207	231

1 - cocksfoot, 2 - Randall, 3 - lady's-laces

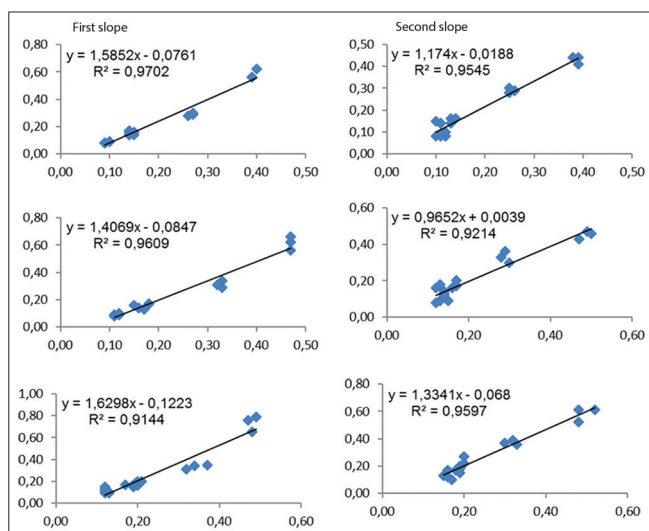


Figure 1: Correlation coefficients between the calculated values of ^{137}Cs accumulation coefficient and the experimental values of ^{137}Cs accumulation coefficient of air-dry overground mass of Pooideae grasses in different years (2009–2011): Bioremoval parameter $\lambda = \lambda_k - \lambda_n$ calculated by the formula (9), a - cocksfoot, b - Randall, and b - lady's-laces

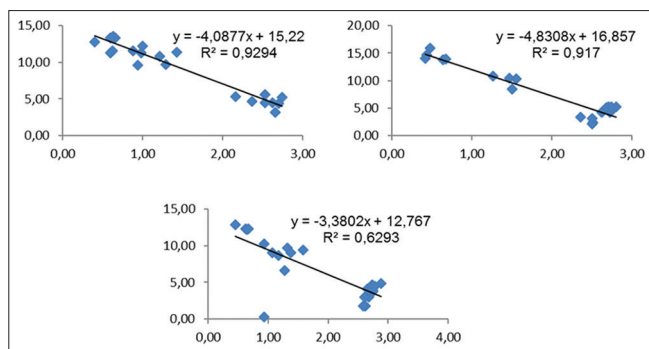


Figure 2: Correlation coefficients between the bioremoval parameter λ calculated by formula (9) and relative transpiration: a - Cocksfoot, b - Randall, and b - lady's-laces

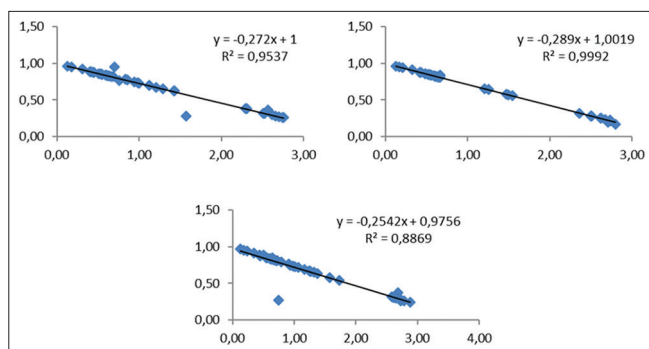


Figure 3: Correlation relationship between the number of Pe flows of soil moisture and the relative transpiration of Pooideae grasses: a - Cocksfoot, b - Randall, and c - lady's-laces

Table 2 shows the values of the parameters λ calculated by formula (4) and characterizing the soils and the root system. To calculate the specific surface of soil, the fractional

analysis data were used by the method given in Pakshina and Skovorodnikova.^[17] In order to calculate the parameter λ_k the capacity of the cation exchange of the root system was taken from.^[18-20]

The specific activity of the root systems was calculated by the formula (4), assuming that $\lambda_k = \lambda_0 + \lambda_n$, where λ_0 and λ_n are known values [Figure 2 and Table 2].

The specific surface of the root systems of cocksfoot, Randall, and lady's-laces was 7.47, 8.24, and 10,18 m^2/g of air-dry roots.

Figure 4 shows the correlation between the values of K_H ^{137}Cs of Pooideae grass air-dry mass calculated according to formula (8) and the calculation of the parameter according to the formula (4), and the experimental values of K_H .

It follows from Figure 4 that there is a close relationship between the calculated and experimental values of K_H .

Thus, in the absence of experimental data for the calculation of the parameter λ by the formula (9), the formula (4) can be used to estimate the K_H ^{137}Cs of crop production.

It follows from the equation (5) and (6) that the main role in the bioremoval of ions from the soil belongs to the absorption (adsorption) capacity of the root systems. The equations describe the process of ion adsorption by root globules, outgrowths formed by the cells of the surface layer. The ratio of the radionuclide amount, absorbed by 1 g of the adsorbent, to its content in 1 cm^3 of the equilibrium soil solution, was named the “distribution coefficient” (K_d) and serves to characterize the adsorption of capillary-porous systems.^[21]

According to the formula (6), they calculated the K_d values of single-species grass crops, which were compared with the experimental values of K_H . As can be seen from Figure 5, there is a very close correlation between the adsorption of ^{137}Cs by the root system and the K_H of the aboveground phytomass of grasses.

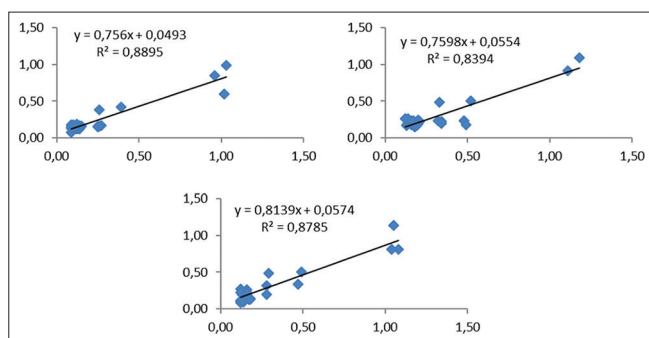
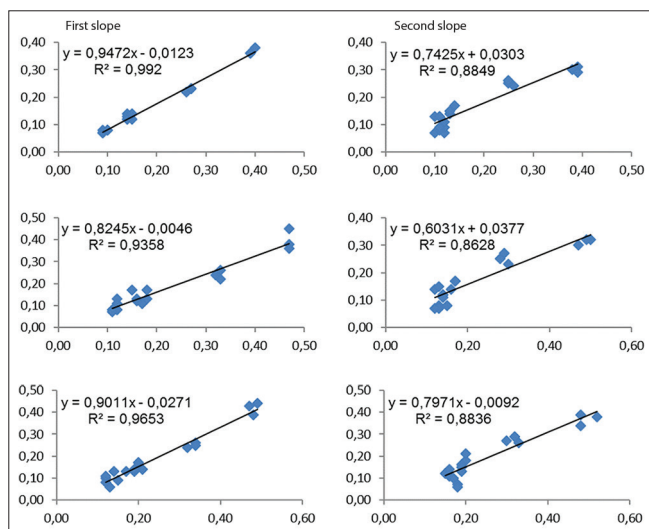
According to the correlation equations on Figure 5, it follows that when the values of K_H change by one, the K_d values of cocksfoot, Randall, and lady's-laces make 0.947, 0.824, and 0.901 for the first slope and 0.742, 0.603, and 0.797 for the second slope. Thus, during vegetation, a more active absorption of ^{137}Cs from soil by the root systems is observed before the first slope, then from the first to the second slope.

Figure 6 shows the dependence of ^{137}Cs distribution coefficient by the root system of grasses on relative transpiration. As follows from the figure, there is a close relationship between the values of K_d and $\sum_B E_T / \sum_B E_0$ during the vegetation period.

Table 2: The indicators of soil and root system of plants used to calculate the bioremoval parameter of ^{137}Cs of Pooideae grasses and ^{137}Cs accumulation coefficient of aboveground phytomass

Year	Air temperature, °K	Soil, λ_n	Root system, λ_k			Bioremoval parameter at $\sum_B E_T / \sum_B E_0 = 0, \lambda_0$		
			1	2	3	1	2	3
First slope								
2009	286.9	6.27	21.44	22.89	19.00	15.17	16.62	12.73
2010	289.2	6.22	21.27	22.70	18.85	15.05	16.48	12.63
2011	289.3	6.22	21.27	22.70	18.85	15.05	16.48	12.63
Second slope								
2009	294.4	6.16	21.07	22.48	18.66	14.91	16.32	12.50
2010	297.0	6.06	20.72	22.72	18.36	14.66	16.66	12.30
2011	294.1	6.12	20.93	22.34	18.21	14.81	16.22	12.12

1 - cocksfoot, 2 - Randall, 3 - lady's-laces

**Figure 4:** Correlation coefficient between the values of ^{137}Cs accumulation coefficient calculated by the formula (8) and the experimental values of ^{137}Cs accumulation coefficient of air-dry overground mass of Pooideae grasses (2009–2011): (a) Cocksfoot, (b) Randall, (c) Lady's-laces**Figure 5:** Correlation coefficient between the values of the ^{137}Cs distribution coefficient of the root system calculated from formula (6) and the experimental values of the ^{137}Cs accumulation coefficient of air-dry by overground mass of the Pooideae grasses in different years (2009–2011): (a) Cocksfoot, (b) Randall, (c) Lady's-laces

The coefficients of determination for cocksfoot, Randall, and lady's-laces were 0.85, 0.89, and 0.88, respectively. This means that 85, 89, and 88% of the changes in ^{137}Cs adsorption by the root system of these herbs are conditioned by relative transpiration or accessibility of soil moisture to the root system of plants.

According to the correlation equations on Figure 6, it follows that if the relative transpiration is equal, and consequently, an equal concentration of the soil solution, the adsorption of ^{137}Cs by the surface of cocksfoot, Randall, and lady's-laces roots at $\sum_B E_T / \sum_B E_0 = 1$ makes 0.24, 0.25, and 0.27 Bq/g of air-dry roots. According to the works,^[18-20] the capacity of absorption by the roots of cocksfoot, Randall, and lady's-laces makes 25.6, 30.4, and 30.8 mg-equiv. per 100 g of air-dry roots.

The sequence of ^{137}Cs adsorption values coincides with the values of λ and KH cultures. From this, it follows that the formula (8) reflects the role of the root system reliably in the bioremoval of ^{137}Cs from the soil by different types of crops.

CONCLUSION

The results of KH, Pe, and Kd calculations and the parameters of ^{137}Cs bioremoval from the soil by the phytomass of Pooideae grasses are presented in this article and allow us to draw the following conclusions.

The numerical method of ^{137}Cs accumulation coefficient calculation of herbal phytomass can be used in two cases: In the presence of experimental data of the bioremoval parameter λ calculated according to formula (9), and at their absence, by calculation using the formula (4).

It was established that the number Pe entering into the formula (4) depends on the relative transpiration of grass

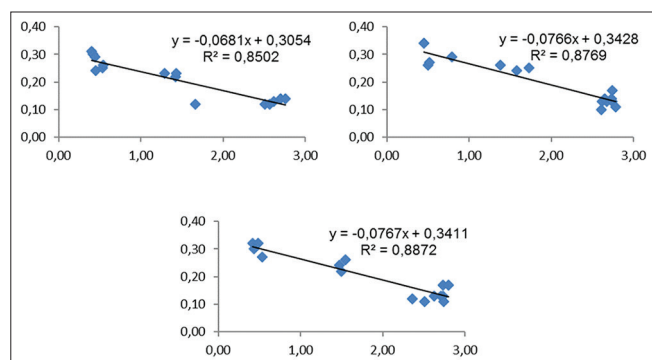


Figure 6: Correlation coefficients between the distribution coefficient of ^{137}Cs of the root system and relative transpiration of Poideae grasses: (a) Cocksfoot, (b) Randall, (c) Lady's-laces

crops or the availability of soil moisture to the root system of plants. Soil-hydrological constants are characterized by definite values of the number P_e . These data make it possible to calculate KH without a preliminary experimental determination of the bioremoval parameter, directly according to the formula (4).

The parameter of ^{137}Cs bioremoval from the soil by a certain type of culture (λ) is equal to the difference of the root system parameters λ_k and the soil parameter λ_n . These parameters depend on the following indicators: Cation exchange capacity, specific surface area, P_e number, cation and salt anion temperature, and valence. The leading role in the bioremoval of ^{137}Cs from the soil belongs to the root system, which, as compared with the soil, is characterized by a large absorption capacity and an incomparably small specific surface area.

Root hairs, forming the water and ion absorption zone at the root are characterized by a large adsorption capacity. It was revealed that the ^{137}Cs adsorption process by the root system of grasses is more intensive during the period from the renewal of vegetation to the first slope, then in the period from the first slope to the second one.

The absorptive capacity of the root systems of plants depends on the type of culture. With equal values of relative transpiration, the rooting system of lady's-laces has the largest absorbing capacity, and cocksfoot has the least absorbing capacity. This sequence of herb species is observed for the value of the bioremoval parameter and ^{137}Cs accumulation coefficient of grass biomass.

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