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Assessment of countermeasure effects on ^{137}Cs accumulation from soil by farm crops after the accident at the Chernobyl NPP

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Abstract. Using the south-western districts in the Bryansk region affected by the Chernobyl accident as an example, the dynamics has been estimated of ^{137}Cs transfer factors to plants and effects of countermeasures on the accumulation of this radionuclide by farm products. A significant decline with time in TF ^{137}Cs to plants is shown. Over the 20 years after the radioactive fallout this decline varies from 7 to 85 times for different plants, being maximum in the early years. The ^{137}Cs accumulation by plants is greatly influenced by the soil properties. Thus, the maximum transfer of this radionuclide to plants is reported on peaty soils, somewhat lower on sandy soils and minimum on sandy loam soil. A detailed comparative analysis of data on TF ^{137}Cs to plants grown on sandy and sandy loam soils has shown that on the former transfer is higher and varies between 1.2–4.5 times for different crops. An important role of mineral fertilizing and liming of soil has been proved in reducing ^{137}Cs accumulation in plants. On agricultural lands that saw the increased rates of mineral fertilizing, a significant (by an average factor of 3–7) reduction in TF ^{137}Cs was reported compared to the plots with no countermeasures.

1. INTRODUCTION

In studies of the radionuclide behavior in the soil-plant system as the first link of radionuclide uptake to foodstuffs, a focus of special attention is ^{137}Cs, a biologically mobile radionuclide. It is present in the global fallout after nuclear weapons tests and in radioactive releases from the Chernobyl accident, and it is amongst the main dose-forming radionuclides. As evident from the pre-Chernobyl studies, one of the most effective approaches to reduce ^{137}Cs uptake by plants is application of mineral fertilizers and liming of acid soils. In this case ^{137}Cs content in products dropped 2–7 times. The accumulated large actual material on migration of this radionuclide in the soil-plant system, with the account of countermeasures application for more than 20 years, makes it possible to evaluate the dynamics and peculiar features of ^{137}Cs accumulation in plants at different rates of mineral fertilizing and at different times after the accident. These evaluations allow identification of time periods when the application of countermeasures on agricultural lands with various levels of soil fertility is justified.

2. MATERIALS AND METHODS

To study ^{137}Cs transfer to plants, the results from annual monitoring carried out by the Bryansk center "Agrochemradiology" were employed. These data included levels of ^{137}Cs contamination of plant stuffs – grain, potato, vegetables (cabbage, tomatoes, cucumbers) and fodder stuffs (haylage, hay, silage) produced in 143 collective farms of 7 most affected districts of the Bryansk region. In 1986–2006, 1.7 mln tons grain, 1.1 mln tons potato, 0.2 mln tons vegetables, 2.1 mln tons haylage, 1.3 mln tons hay and 2.4 mln tons silage were tested for ^{137}Cs content. For calculations, samples of each product type were used, sample size varying from 4.8 to 21.2 thousand.

The information on ^{137}Cs contamination levels of agricultural land (arable land for plant production and haylands and pastures for fodder production) was based on the results from radiological mapping of soils in the Bryansk region. For each year after the accident, the contamination levels of agricultural lands were estimated considering ^{137}Cs decay:

$$\sigma(t) = \sigma_{86} \cdot \exp^{\frac{(-0{,}693 \cdot \Delta t)}{T_{1/2}}}, \quad \text{kBq/m}^2, \tag{1}$$

where σ_{86} is the ^{137}Cs contamination density of agricultural lands in 1986, kBq/m^2; Δt is the time, number of years elapsed after 1986 till the year of estimation; $T_{1/2}$ is the ^{137}Cs half-life period equal 30.17 years.

An important factor influencing ^{137}Cs accumulation in plants are soil properties. To study the soil properties, soils of all the study agricultural lands were divided into groups based on their granulometric composition in accordance with the radioecological classification used by some international organizations, including the IAEA. By this classification, all soils are divided into four groups by their ability to fix ^{137}Cs which leads to changes in its bioavailability for plant uptake. Mineral soils are classified into three groups based on their granulometric composition: sandy (sandy and sandy loam), loam (medium- and light loam), clay (heavy clay and clay soils). The fourth group includes organic (peaty) soils, which have some peculiar features defining an increased ^{137}Cs transfer to plants. The arable soil of the 7 south-western districts of the Bryansk region, used for plant production, is presented by two groups: sandy and loam. Haylands and pastures, in addition to these two groups, include organic (peat-boggy) soils.

Following the Chernobyl accident, increased doses of mineral fertilizes were applied to the contaminated agricultural lands to create an agrochemical barrier for ^{137}Cs. These, however, were not always optimal. Soils that saw the largest scales of mineral fertilizing, exhibited decrease in the soil acidity, increase in plant availability of phosphorus and potassium, increase in humus content, which had resulted in reduced ^{137}Cs transfer to plants. Not so pronounced was the influence of mineral fertilizing where it was applied at a limited scale. To determine the influence of countermeasures on ^{137}Cs transfer to plants, all the agricultural lands were divided into three groups based on the scales of agrochemical works and, consequently, achieved level of soil fertility (Table 1).

Table 1. Agrochemical parameters of soils in the Bryansk region in different variants of countermeasure application.

Variant	Humus, %	Acidity, pH	Exchangeable potassium, mg/100 g	Mobile phosphorus mg/100 g	Cultivation level index
1. Active countermeasures	2.2–3.5	5.8–6.5	17–35	15–30	>0.7
2. Mid-countermeasures	1.7–2.2	5.2–5.8	8–17	10–15	0.6–0.7
3. No countermeasures	0.9–1.7	4.0–5.2	<8	<10	0.4–0.6

Based on the samples of data combinations crop-soil-year-countermeasure, a database has been compiled which includes 26718 entries. Using these data, TF ^{137}Cs were computed from various soil groups to farm crops for each year after the accident considering the presence or absence of countermeasures.

3. RESULTS AND CONCLUSIONS

Based on a large body of actual data, a significant decrease with time in TF ^{137}Cs to plants was demonstrated. Over 20 years following the event, this drop amounted to 7–85 times for different plants, being maximal in the early years. ^{137}Cs accumulation by plants is greatly influenced by the soil properties. Thus, the maximal transfer is observed on peaty soils, somewhat less – on sandy soils and minimal – on loams. A more detailed comparative analysis of TF ^{137}Cs data to plants grown on sandy and loam soils has shown that transfer is higher on sandy soils and varies for different crops between 1.2 and 4.5 times.

Analysis of TF ^{137}Cs ratio for the absence of countermeasures and their active application has revealed the effectiveness of countermeasures in plant and fodder production every year after the ChNPP accident (Fig. 1). The figure demonstrates that the maximum countermeasure effectiveness was achieved in the first several years after the accident (1988–1990). In that period the TF ^{137}Cs ratio for the absence and active countermeasures for various crops reached 12 times and more. Such high effectiveness was caused by two reasons. On the one hand, in the early years after the accident agroameliorants were applied to soil on the maximum scales. Thus, in 1986–1990, the Bryansk region alone saw the application of lime, phosphate rocks and potassium to the agricultural land in an area of more than 1.36 mln. ha, and radical improvement of haylands and pastures in 97.6 thousand ha. On the other hand, in the first years after the accident ^{137}Cs in soil was rather mobile and in the absence of increased mineral nutrition was quickly uptaken by plants. Therefore, the difference in the TF ^{137}Cs ratio for the absence and active application of agroameliorants was significant.

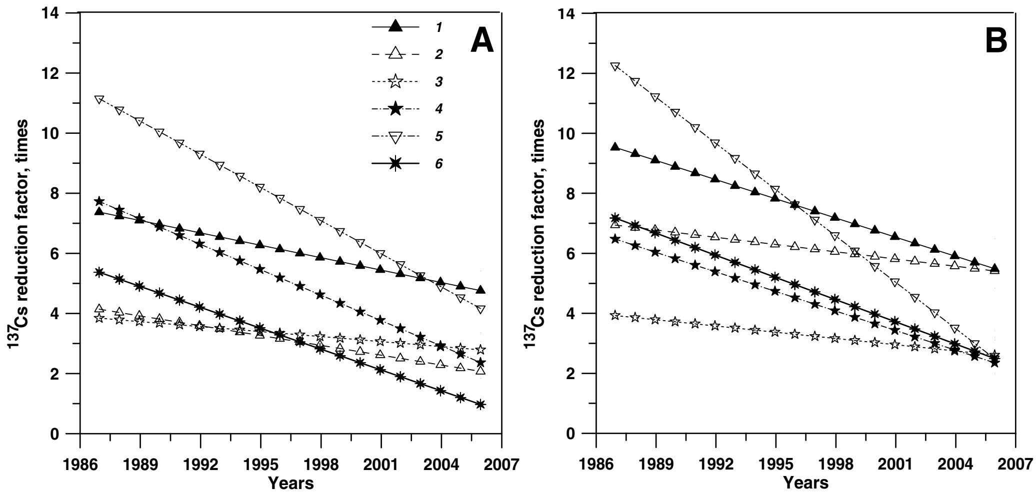

Figure 1. Dynamics of TF ^{137}Cs to farm crops in the absence of countermeasures and their active application on sandy – (A) and loam – (B) soils (1 – grain, 2 – potatoes, 3 – vegetables, 4 – haylage, 5 – hay, 6 – silage).

In the later years, ^{137}Cs was fixed by soil becoming less available for plants, and effects from mineral fertilizing started to decrease. However, up to now, TF^{137}Cs to plants, should mineral fertilizing is sufficient compared to the control, was at the level of 1.8–6.6 times for different crops. At the same time, it should be noted that such a significant difference in TF ^{137}Cs (particularly in the early years after the accident) was only reported for the agricultural land with intensive countermeasures and in their absence (i.e. in extreme cases). However, in practice, more often were the situations of different agroameliorant treatments at moderate rates. For these conditions the difference in TF ^{137}Cs to various farm plants from different soil types varied within 1.1–3.0 times. The investigations have also demonstrated that in the same conditions ^{137}Cs to a larger extent is transferred to fodder products and to a lesser extent – to plant products, which is necessary to be taken into account when planning crop rotation schemes in radioactively contaminated areas.

Table 2. Model parameters describing the dynamics of TF ^{137}Cs to plants.

Soil group	Countermeasure option	First period, 1987–1991			Second period, 1992–1997			Third period, 1998–2006		
		Tf_1	λ_1	R^2	Tf_2	λ_2	R^2	Tf_3	λ_3	R^2
Grain										
Sandy	No counter.	3.19	0.43	0.90	0.20	0.08	0.86	0.13	0.02	0.97
	Mid-counter.	2.04	0.51	0.94	0.10	0.06	0.97	0.07	0.04	0.94
	Active counter.	0.45	0.52	0.94	0.03	0.05	0.94	0.03	0.04	0.83
Loam	No counter.	2.17	0.46	0.90	0.13	0.07	0.90	0.10	0.03	0.89
	Mid-counter.	1.11	0.45	0.90	0.06	0.06	0.88	0.05	0.03	0.86
	Active counter.	0.20	0.54	0.99	0.02	0.10	0.98	0.02	0.03	0.86
Potato										
Sandy	No counter.	0.87	0.47	0.98	0.11	0.20	0.80	0.10	0.09	0.87
	Mid-counter.	0.68	0.62	0.99	0.07	0.10	(0.21)	0.05	0.07	0.63
	Active counter.	0.19	0.48	0.91	0.04	0.09	0.59	0.03	0.07	0.54
Loam	No counter.	0.55	0.61	0.97	0.09	0.15	0.91	0.08	0.11	0.80
	Mid-counter.	0.41	0.78	0.92	0.05	0.09	0.78	0.03	0.03	0.66
	Active counter.	0.15	1.35	0.97	0.01	0.60	0.72	0.01	0.05	0.77
Vegetables										
Sandy	No counter.	0.67	0.36	0.99	0.13	0.20	0.98	0.06	0.06	0.95
	Mid-counter.	0.56	0.51	0.99	0.07	0.15	0.85	0.04	0.08	0.97
	Active counter.	0.21	0.38	0.95	0.03	0.14	0.98	0.02	0.07	0.97
Loam	No counter.	0.32	0.71	0.96	0.05	0.20	0.91	0.04	0.08	0.96
	Mid-counter.	0.28	0.80	0.96	0.04	0.15	0.74	0.03	0.09	0.92
	Active counter.	0.05	0.30	0.99	0.01	0.11	(0.19)	0.01	0.06	0.99
Haylage										
Sandy	No counter.	13.71	0.59	0.95	0.74	0.12	0.91	0.41	0.10	0.90
	Mid-counter.	10.22	0.71	0.96	0.48	0.12	0.96	0.22	0.03	0.59
	Active counter.	2.67	0.64	0.87	0.12	0.10	0.94	0.09	0.02	0.81
Loam	No counter.	10.95	0.81	0.99	0.55	0.25	0.82	0.22	0.05	0.96
	Mid-counter.	8.12	0.90	0.99	0.24	0.11	0.78	0.17	0.05	0.97
	Active counter.	2.03	0.79	0.97	0.08	0.10	0.64	0.06	0.03	0.87
Hay										
Sandy	No counter.	19.00	0.33	0.94	3.61	0.25	0.89	1.34	0.17	0.95
	Mid-counter.	12.05	0.48	0.98	1.36	0.12	0.87	0.82	0.19	0.87
	Active counter.	3.03	0.51	0.87	0.36	0.14	0.98	0.17	0.08	0.84
Loam	No counter.	15.45	0.36	0.91	2.30	0.23	0.97	0.75	0.18	0.91
	Mid-counter.	11.71	0.55	0.98	1.48	0.20	0.99	0.52	0.21	0.96
	Active counter.	2.37	0.59	0.95	0.19	0.09	0.96	0.12	0.07	0.93
Peaty	No counter.	35.20	0.52	0.99	4.76	0.19	0.97	1.74	0.10	0.93
	Mid-counter.	16.38	0.47	0.98	2.06	0.13	0.92	1.01	0.06	0.74
	Active counter.	4.76	0.75	0.98	0.37	0.10	0.73	0.34	0.05	0.93
Silage										
Sandy	No counter.	3.49	0.45	0.98	0.29	0.07	0.89	0.19	0.04	0.97
	Mid-counter.	2.57	0.68	0.96	0.20	0.04	0.82	0.16	0.04	0.96
	Active counter.	0.78	0.62	0.99	0.10	0.06	0.82	0.09	0.02	0.91
Loam	No counter.	2.75	0.52	0.99	0.23	0.08	0.94	0.16	0.05	0.94
	Mid-counter.	1.20	0.56	0.99	0.16	0.05	0.97	0.12	0.04	0.91
	Active counter.	0.43	0.74	0.96	0.05	0.05	0.94	0.04	0.03	0.82

* - In brackets are values for which correlation is not significant.

Analysis of TF ^{137}Cs data has demonstrated that reduction in ^{137}Cs accumulation by plants varied significantly in different times after the ChNPP accident. Thus, in the first 4–5 years, TF ^{137}Cs reduction was most intensive. Within 6–10 years after radioactive contamination it became slower, and after 1998 decrease in TF ^{137}Cs was minimal. For the correct description of these processes a method is being

Table 3. Effective half-lives of ^{137}Cs in plants, years.

Soil group	Countermeasure option	First period, 1987–1991	Second period, 1992–1997	Third period, 1998–2006
Grain				
Sandy	No counter.	1.62	8.70	28.88
	Mid-counter.	1.36	12.37	16.86
	Active counter.	1.32	14.40	17.29
Loam	No counter.	1.52	10.08	24.53
	Mid-counter.	1.53	12.18	23.10
	Active counter.	1.28	7.10	20.29
Potato				
Sandy	No counter.	1.46	3.47	7.68
	Mid-counter.	1.12	6.93	9.25
	Active counter.	1.45	7.70	10.51
Loam	No counter.	1.14	4.62	6.10
	Mid-counter.	0.89	7.70	19.90
	Active counter.	0.51	1.16	14.61
Vegetables				
Sandy	No counter.	1.91	3.53	11.31
	Mid-counter.	1.35	4.62	9.16
	Active counter.	1.80	4.79	9.75
Loam	No counter.	0.97	3.47	8.85
	Mid-counter.	0.87	4.62	7.33
	Active counter.	2.34	6.30	10.80
Haylage				
Sandy	No counter.	1.18	5.69	6.92
	Mid-counter.	0.97	5.64	27.36
	Active counter.	1.08	6.93	29.28
Loam	No counter.	0.86	2.77	12.78
	Mid-counter.	0.77	6.56	13.07
	Active counter.	0.88	7.01	20.33
Hay				
Sandy	No counter.	2.12	2.73	4.05
	Mid-counter.	1.43	5.69	3.65
	Active counter.	1.35	4.96	9.08
Loam	No counter.	1.90	3.08	3.83
	Mid-counter.	1.26	3.52	3.27
	Active counter.	1.17	7.70	10.60
Peaty	No counter.	1.33	3.70	7.15
	Mid-counter.	1.49	5.27	11.24
	Active counter.	0.92	6.93	13.49
Silage				
Sandy	No counter.	1.53	9.59	16.71
	Mid-counter.	1.01	15.52	16.72
	Active counter.	1.11	11.55	30.12
Loam	No counter.	1.34	8.74	13.26
	Mid-counter.	1.24	14.40	19.28
	Active counter.	0.94	13.75	19.91

effectively used in recent years based on the estimation of the effective half-lives (T_{eff}), i.e. time during which the radionuclide content in different components of the trophic chain drops by half under the influence of various factors. Considering that the pattern of TF ^{137}Cs to farm crops varied significantly in different periods after the accident, T_{eff} TF ^{137}Cs for the three time periods: 1987–1991, 1992–1997 and 1998–2006 were calculated. The reduction dynamics of TF ^{137}Cs was described as a sum of three exponential functions:

$$Tf(t) = \begin{cases} Tf_1(0) \cdot \exp^{-\lambda_1 t}, \partial\pi R\ t_1 \leq t < t_2 \\ Tf_2(0) \cdot \exp^{-\lambda_2 t}, \partial\pi R\ t_2 \leq t < t_3 \\ Tf_3(0) \cdot \exp^{-\lambda_3 t}, \partial\pi R\ t \geq t_3 \end{cases} \quad (2)$$

where $Tf(t)$ is the TF ^{137}Cs from soil to plants, (Bq/kg)/(kBq/m^2); t is the time after radioactive fallout, years; λ_1, λ_2, λ_3 are the constants of TF ^{137}Cs reduction rate, year^{-1}; Tf_1, Tf_2, Tf_3 are the model parameters, (Bq/kg)/(kBq/m^2).

In this case T_{eff} may be presented as follows:

$$T_{eff} = \frac{\ln(2)}{\lambda} \quad (3)$$

The model parameters were computed in MS Excel by a decision function. The derived parameters for different soil groups, plants and countermeasure options are presented in Table 2.

In most cases the correlation coefficients (R^2), which reflect the level of agreement between the model and experimental data, are fairly high and range between 0.54 and 0.99. The T_{eff} ^{137}Cs in plants for various times after radioactive fallout, soil groups and countermeasure scales are given in Table 3.

The data analysis shows that for the first period after the event T_{eff} ranges between 0.5–2.3 year, the parameter being higher for the same crops on the plots with no countermeasures and minimal on lands with active application of countermeasures. For the second period T_{eff} varies in a wider range: 1.2 to 15.5 years. The maximal T_{eff} are reported in cereals (7.1–14.4 years) and maize for silage (8.7–15.5 years). The third period after the Chernobyl accident is characterized by the maximal T_{eff} values (3.3–30.1 years). The T_{eff} values for this period have maximal scattering. It is, however, seen that for some crops decrease in ^{137}Cs content in products is mainly dictated by the radionuclide decay.

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