

## Assessment of Factors Influencing Changes in $^{137}\text{Cs}$ Contamination Density of Agricultural Lands<sup>1</sup>

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**Abstract**—A comparative analysis of predictive estimates of the  $^{137}\text{Cs}$  contamination level of agricultural lands with real measurement data is given for the example of southern districts of the Kaluga oblast that suffered from the accident at the Chernobyl nuclear power plant. Predictive calculations based on consideration of only radionuclide decay are sufficiently reliable; however, for greater accuracy it is necessary to take into consideration the amount of agrotechnical protective measures being carried out.

**Keywords:** forecast estimates,  $^{137}\text{Cs}$  contamination, radionuclide decay, protective measures

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After the radiation accident at the Chernobyl nuclear power plant, data on contamination of lands by radionuclides have served as the basis for assessing the radioecological situation in the agroindustrial complex and planning agricultural production for obtaining products meeting the standards [1]. In this case, the results of a radiological survey of agricultural lands became the baseline information when making decision about rehabilitation of contaminated territories [2]. It is necessary to note that measures concerning a large-scale radiological survey of agricultural lands are quite expensive and labor-intensive and they are carried in the same territory, as a rule, in rounds of once in 4–5 years [3]. In recent years, methods of predicting the level of contamination of agricultural lands by radionuclides have been being used when planning agricultural production [4].

In 2009, an Atlas of present and predictive (up to 2056) aspects of the consequences of the Chernobyl accident for Russia and Belarus was published, wherein the principles of predicting  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  contamination levels of territories with consideration of only decay of these radionuclides are substantiated for the example of inhabited localities [5]. This assessment method is sufficient for mapping, but its accuracy when predicting radionuclide contamination of a particular plot of agricultural lands (plowland, meadows, and pastures) is unknown.

As investigations showed, the following factors can affect a reduction of contamination density of agricultural lands in time besides radionuclide decay:

migration of radionuclides in soil [6];

protective agrotechnical measures, for example, plowing of agricultural lands, which leads to redistribution of radionuclides in the soil profile [7];

removal of part of the radionuclides with plant industry and fodder production products over the course of a long time [8].

Estimation of the degree of influence of all these unaccounted factors on the accuracy of predicting the level of radionuclide contamination of agricultural lands is important both for optimizing radiological support and for planning agroindustrial production in contaminated territories.

The purpose of the present work was a comparative analysis of predictive estimates of  $^{137}\text{Cs}$  contamination density of agricultural lands for various periods after the Chernobyl accident with real measurement data and estimation of the role of factors influencing the accuracy of such predictions.

### METHOD

In 2007, during preparation of the Atlas [5] by specialists of the All-Russian Agricultural Radiology and Agroecology Research Institute and Kaluga Agrochemradiology Center, a radiological survey of agricultural lands was carried out at 36 farms in three southern districts of Kaluga oblast affected by the accident. At these farms, 155 plots (135 plowlands and 20 pastures) were monitored. The total area of the survey was 7900 ha (Table 1), the area of the plots varied from 2 to 250 ha; the average was about 52 ha. The region of investigations was selected because each plot

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**Table 1.** Characteristics of the region of investigations in Kaluga oblast

District	Number of farms	Plowland		Haylands and pastures	
		number of plots	area, ha	number of plots	area, ha
Zhizdra	11	41	1933	3	317
Ul'yanovsk	15	53	2320	11	341
Khvastovich	10	41	2753	6	219

was characterized by a sufficiently uniform  $^{137}\text{Cs}$  contamination level.

On each plot we took soil samples in accordance with method [9] and determined the  $^{137}\text{Cs}$  concentration in the samples. On the basis of the data, we estimated the density of contamination of the plots by this radionuclide in 2007 by the formula:

$$Q_i = Ahd \times 10^{-3}, \text{ kBq/m}^2, \quad (1)$$

where  $Q_i$  is the contamination density of soils (stock of radionuclides in the 0–20 cm soil plow layer per 1 m<sup>2</sup> area), kBq/m<sup>2</sup>;  $A$  is the soil radionuclide concentration, Bq/kg;  $h$  is the thickness of the plow horizon, cm;  $d$  is the density of soil, g/cm<sup>3</sup>;  $10^{-3}$  is a coefficient for converting Bq/m<sup>2</sup> to kBq/m<sup>2</sup>.

Earlier (1992–2005) a radiological survey was also conducted on these plots, which was accompanied by a study of their soil characteristics (estimation of agrochemical indices and texture) and collection of data on protective and rehabilitation measures on the investigated agricultural lands [10]. On the basis of the information collected in the original radiological survey of the plots, predictive estimates of the level of their  $^{137}\text{Cs}$  contamination ( $Q_r$ ) in 2007 were made. In this case, we used an expression taking into consideration only radionuclide decay:

$$Q_r(t) = Q_i \exp\left(\frac{(-0.693\Delta t)}{T_{1/2}}\right), \text{ kBq/m}^2, \quad (2)$$

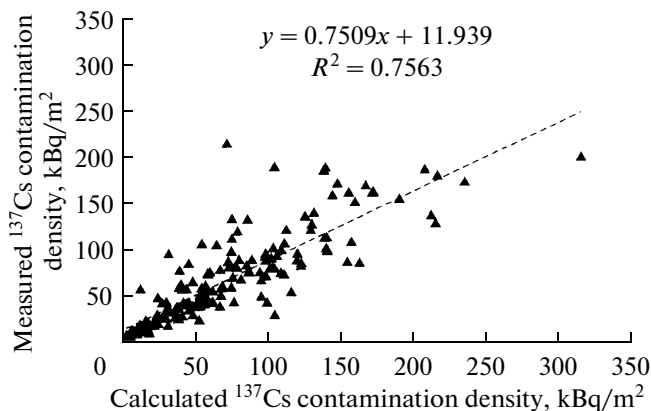
where  $Q_i$  is  $^{137}\text{Cs}$  contamination density of agricultural lands in the year of the first radiological survey, kBq/m<sup>2</sup>;  $\Delta t$  is the number of years since the first survey up to 2007;  $T_{1/2}$  is the half-life of  $^{137}\text{Cs}$ , equal to 30.17 years.

The results obtained allowed comparing the predictive calculations of  $^{137}\text{Cs}$  contamination level of agricultural lands in 2007 with the data of real measurements taken in that year of investigation.

## RESULTS AND DISCUSSION

The predicted  $^{137}\text{Cs}$  contamination density of the investigated plots in 2007 differed from the real (measured) on average (toward the greater side) by 12% (Fig. 1). The correlation between the calculated and real data was quite high ( $r = 0.87$ ). Since the predictive estimates taking into account only  $^{137}\text{Cs}$  decay gave overstated indices of contamination density of the investigated plots, we can assume that, under real conditions, a change in this parameter can be affected by other factors besides radionuclide decay (by  $^{137}\text{Cs}$  migration in soil and food chains, characteristics of conducting agricultural production, and protective measures). A ratio ( $Q_r/Q_i$ ) was found for revealing the degree of their influence between pairs of calculated ( $Q_r$ ) and real ( $Q_i$ ) values of  $^{137}\text{Cs}$  contamination density of the plots in 2007; the results obtained were divided into groups according to each index (factor) under study and samples of the data were treated statistically. Obviously, the closer the ratio ( $Q_r/Q_i$ ) to 1, the more accurate the prediction based on consideration of only  $^{137}\text{Cs}$  decay. If it differs from 1, then another factor besides decay of radionuclides influences a change in  $^{137}\text{Cs}$  contamination density of agricultural lands.

Classification of the data of this ratio according to times of conducting the first radiological survey of the plots allowed identifying two groups of results obtained in different periods after the Chernobyl accident: 1992–1994 and 1999–2005 (Fig. 2). We can assume that, with the course of time, a part of radionuclides is removed from soils with agricultural products. Then the ratio ( $Q_r/Q_i$ ) obtained in 1992–1994 will differ more from 1 compared with the 1999–2005 data. However, as is seen from Fig. 2, the time factor (annual removal of radionuclides with products) essentially didn't affect the accuracy of the predictive calculations. With the exception of the data of the survey in 1992, other results of the comparison were characterized by a high correlation coefficient (0.72–0.95) and small scatter of the values of ( $Q_r/Q_i$ ). The average ratios of the predictive estimates of  $^{137}\text{Cs}$  contamination densities to the real ones (with the exception of 1992 and 2001) were close to 1 and varied within 0.96–1.10. The considerable differences in the average val-



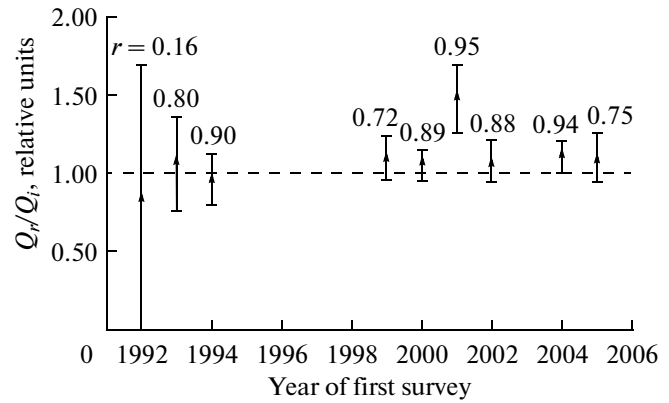
**Fig. 1.** Regression field between calculated and measured  $^{137}\text{Cs}$  contamination density of agricultural lands.

ues of the ratios under study in 1992 and 2001 can be explained by the small size of samples of the data. In those years, a radiological survey of seven plots of agricultural lands out of the number of all those being investigated was carried out, whereas in other years there were from 11 to 35 of them. In 1992, with consideration of the greater scatter of the data of the sample from the average value of the ratio ( $Q_r/Q_i$ ), the methods of taking and measuring soil samples for  $^{137}\text{Cs}$  content were possibly also violated. Thus, we can conclude that  $^{137}\text{Cs}$  is annually removed from soil with phytomass in negligibly small amounts, which essentially doesn't affect the accuracy of predicting the level of contamination of agricultural lands by it over the course of both short and long time intervals after radioactive fallout (15 years and more).

The classification of the plots according to soil texture also didn't allow determining the effect of this factor on the accuracy of predictive estimates (Table 2). The ratio of calculated  $^{137}\text{Cs}$  contamination densities of agricultural lands to the measured differed from 1 by 5–6% for both soil groups characteristic for the region, which fits into the standard system of measurement errors. During the long time after radioactive fallout, the bulk of  $^{137}\text{Cs}$  on sand and loam soils was in the upper 20 cm layer; migration of this radionuclide had a local character and it didn't leave the upper horizon of the soil cover.

Separation of the investigated plots according to types of agricultural lands showed that, on plowland the predictive calculations were more accurate than on haylands and pastures. The ratio of the calculated to the measured data on  $^{137}\text{Cs}$  contamination level of plowland was closest of all to 1 with a small confidence interval (Table 2). It was slightly higher on haylands and pastures, which could be related both to higher removal of  $^{137}\text{Cs}$  with fodder and forage than to products of the plant industry being produced on plowland and to the more active conduction of agromeliorative measures on them.

The data on conducting a set of protective and rehabilitation measures on the plot were used for revealing the degree of influence of the countermeasure factor on the accuracy of predictive calculations of the level of  $^{137}\text{Cs}$  contamination of agricultural lands. From 1992 through 2005, agrochemical and agrotechnical works were carried out on 30% of the plots to increase the productivity of the agricultural lands and reduce the uptake of  $^{137}\text{Cs}$  into plants, which included the application of increased doses of potassium fertilizers, liming, and applying phosphorite. The use of mineral fertilizers was accompanied by reploting the soil deeper than with the usual technology of conducting such works; this affected the redistribution of radionuclides in the upper soil layer. Separation of the plots into two groups with the use or absence of protective measures on them showed that, in the first case, the ratio ( $Q_r/Q_i$ ) was 1.03 and in the second, 1.15.



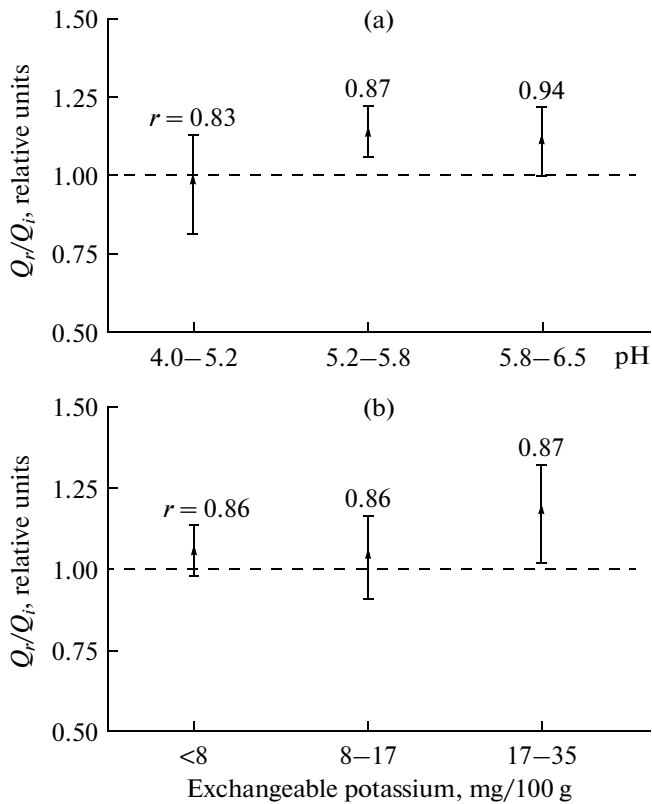
**Fig. 2.** Dependence of the ratio of the calculated to the measured  $^{137}\text{Cs}$  contamination density of agricultural lands on the time of the first survey (the numerical values are the correlation coefficient  $r$ ).

The soil agrochemical indices changing upon introducing countermeasures can give a clearer idea about the effect of protective measures on the accuracy of predicting the level of  $^{137}\text{Cs}$  contamination of agricultural lands. Thus, the average soil pH index of plots located in southern districts of Kaluga oblast without protective measures was 4.4–5.1 and the weighted average of mobile potassium was 7.6–9.3 mg/100 g soil [10]. After taking agrochemical measures, soil acidity decreased and the content of mobile potassium in soil increased. Since the application of mineral fertilizers on agricultural lands was accompanied by conducting agrotechnical works, the dynamics of soil agrochemical indices can be used as an indicator of the extent and effectiveness of agromeliorative protective measures.

**Table 2.** Dependence of the ratio of the calculated to the measured  $^{137}\text{Cs}$  contamination density of plots on soil properties and type of lands

Classifier	Geometric mean of $Q_r/Q_i$	95% confidence interval	Correlation coefficient	Number of plots
Soil texture*				
Sand, loamy sand	1.06	0.15	0.90	46
Sandy–sand clay loam	1.05	0.07	0.85	108
Type of agricultural lands				
Plowland	1.06	0.06	0.87	135
Haylands and pastures	1.08	0.31	0.85	20

\* One plot of the 155 that was located on a peat bog was not taken into consideration in the calculations.



**Fig. 3.** Dependence of the ratio of the calculated to the measured  $^{137}\text{Cs}$  contamination density of agricultural lands on soil agrochemical characteristics: (a) acidity; (b) content of exchangeable potassium (the numerical values are the correlation coefficient  $r$ ).

All plots were divided into three groups according to soil agrochemical indices, which are affected by the intensity of the measures being taken. Examining only soil acidity, we can note that, without protective measures (pH 4.0–5.2), the prediction accuracy was the highest ( $Q_r/Q_i = 0.97$ ). With a decrease of soil acidity due to applying lime to the soil, the reliability of the prediction decreased, since the value of ( $Q_r/Q_i$ ) increased to 1.11–1.14 (Fig. 3a).

The same is noted also for the index characterizing the soil content of mobile potassium. With application of increased potassium fertilizer doses, leading to an increase of the proportion of mobile potassium to 17–35 mg/100 g soil, the accuracy of predictive calculations decreased 17% on average (Fig. 3b). Naturally, the accuracy of the calculations was affected not by the use of mineral fertilizers but by the intensity of accompanying agrotechnical works (deep replowing), which led to movement of part of the radionuclides from the upper to deeper soil layers (25–40 cm), which promoted a reduction of  $^{137}\text{Cs}$  contamination density of the plots.

It is necessary to point out that the introduction of protective measures on agricultural lands in southern districts of Kaluga oblast after the Chernobyl accident

was accomplished on a vastly smaller scale than in the Bryansk oblast [6, 10]. It is not ruled out that, with a greater amount of such works, the noted regularities of a decrease in the accuracy of predicting the  $^{137}\text{Cs}$  contamination level could have had a more pronounced character.

Thus, a long-range prediction of the level of  $^{137}\text{Cs}$  contamination of agricultural lands can be made quite accurately by taking into consideration only decay of the given radionuclide. In this case, the error in the calculations will not exceed 5–8%. Despite the fact that, on haylands and pastures, removal of  $^{137}\text{Cs}$  with fodder production products is somewhat higher than with plant industry products being produced on plowland; the removal of this radionuclide with phytomass doesn't significantly affect the reliability of predictive calculations. With the course of time after radioactive fallout,  $^{137}\text{Cs}$  is not only not removed to a significant degree from agricultural products but also doesn't migrate into deeper soil layers. Its bulk is localized in the upper 20 cm soil layer regardless of texture. At the same time, the predictive estimates of contamination density are always higher than the data of real measurements. When predicting the level of  $^{137}\text{Cs}$  contamination of agricultural lands, in addition to radionuclide decay it is necessary to take into consideration the factor of taking protective and rehabilitation measures. On plots with a set of agrotechnical works, a part of the radionuclides is transported into deeper soil layers, which leads to a reduction of contamination density of agricultural lands. Therefore, on such plots it is necessary to reduce by 15–20% the results of predictive calculations of the  $^{137}\text{Cs}$  contamination level depending on the intensity of agrotechnical measures.

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