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## Comparative Assessment of Radioactive Strontium and Cesium Contents in The Feedstuffs and Dairy Products from Various Regions of Western Siberia.

Olga I. Sebezhko<sup>1</sup>, Valeriy L. Petukhov<sup>1</sup>, Victor A. Sokolov<sup>2</sup>, Olga S. Korotkevich<sup>1</sup>,  
Evgeniy V. Kamaldinov<sup>1\*</sup>, Alexandr I. Syso<sup>1,3</sup>, Tatyana V. Konovalova<sup>1</sup>,  
Nadezhda I. Marmuleva<sup>1</sup>, Kirill N. Narozhnykh<sup>1</sup>, Evgeniy Y. Barinov<sup>1</sup>, and  
Ludmila V. Osadchuk<sup>1,4</sup>

<sup>1</sup>Federal State Budgetary Educational Institution of Higher Education Novosibirsk State Agrarian University, Russia, Novosibirsk, Dobrolubova str. 160

<sup>2</sup>Institute of Molecular and Cellular Biology The Siberian Branch of the RAS, Russia, Novosibirsk, Acad. Lavrentiev Ave. 8/2

<sup>3</sup>Institute of Soil Science and Agrochemistry, The Siberian Branch of the RAS, Russia, Novosibirsk, Acad. Lavrentiev Ave. 8/2

<sup>4</sup>The Federal Research Center Institute of Cytology and Genetics, the Siberian Branch of the RAS, Russia, Novosibirsk, Acad. Lavrentiev Ave. 10

### ABSTRACT

The main radionuclides, which could determine the nature of contamination in the West Siberian region of Russia, are <sup>137</sup>Cs and <sup>90</sup>Sr. In this study, we investigated the accumulation of radioactive strontium and cesium in different types of plant feedstuffs and dairy products from some Siberian regions including the Novosibirsk, Kemerovo and Tomsk regions. It was shown that the content of radionuclides in the five types of plant feedstuffs did not exceed maximum permissible concentration (MPC). At the same time, we found regional differences in content of <sup>90</sup>Sr and <sup>137</sup>Cs in the plant feedstuffs. The content of <sup>90</sup>Sr in the hay from the Kemerovo region was higher as compared with the hay from the Novosibirsk or Tomsk regions, but the content of <sup>90</sup>Sr in concentrated feedstuffs in the Tomsk region was higher than that in Novosibirsk or Kemerovo regions. The concentration of <sup>137</sup>Cs in silage and concentrated feedstuffs from the Tomsk region was higher as compared with other regions, while the content of this radionuclide in hay and silage was lower in Novosibirsk region as compared with Kemerovo or Tomsk regions. The content of both radionuclides in dairy products (whole and dry milk) from Novosibirsk and Tomsk regions was by several times (up to 100) below than the MPC. In conclusion, the territory of Western Siberia is excellent for the production of ecologically safe forage and animal husbandry, and it can be used for the production of dietary and baby foods.

**Keywords:** radioactive strontium and cesium, environmental safety, fodder crops, feedstuffs, dairy products

*\*Corresponding author*

## INTRODUCTION

Organization of the multidisciplinary approach to the production of ecologically safe food products should take into account a natural cycle, which includes the soil, vegetation, animals, produced herbal and animal products, and human beings at the end of the food chain [9, 10, 14]. In this context, one of the most important objects of ecological monitoring should be not only arable lands, grazing and lands used for fodder production, but also various types of vegetable feedstuffs used for livestock and food products [2, 3]. The contamination of plant feedstuffs significantly influences the safety indicators of food products for human consumption [1, 8].

The contamination of soil with radionuclides is of a particular danger to flora and fauna. The main radionuclides, which could determine the nature of contamination in the West Siberian region of Russia, are cesium-137 ( $^{137}\text{Cs}$ ) and strontium-90 ( $^{90}\text{Sr}$ ), which are differently sorted by soil [19]. The basic sorption mechanism of strontium-90 by soil is ion exchange, while for cesium-137 this mechanism includes the ion exchange sorption on the internal surfaces of soil particles. Absorption of strontium-90 by the soil is less pronounced than that of cesium-137 thus it is a movable radionuclide. A significant part of the radionuclides in the soil are localized both on the surface and in the lower layers. At that, their migration depends largely on the type of soil, its granulometric composition, as well as water-physical and agrochemical properties.

At the time of emission of cesium-137 into the environment, the radionuclide is initially in a soluble state (vapor phase, fine particles, etc.). In this case, plants are easily absorbed cesium-137 from the soil. Further, the radionuclide in the soil can enter into different reactions. Its mobility is reduced, while the fixing strength increases, the radionuclide "ages" entering into the complexes of soil crystallochemical reactions and bonded in the structures of secondary clay minerals [18, 19].

Sorption mechanisms of radioactive isotopes in the soil are quite important, because sorption determines their migration quality, and, consequently, the ability to penetrate into plant roots. Sorption of radionuclides depends on many factors among which the soil mechanical and mineralogical composition is one of the main factors. In heavy soils, in terms of their granulometric composition, the absorbed radionuclides, especially cesium-137, are bonded stronger than in the light soils, and with decrease in the size of mechanical fractions of the soil the bonding strength of strontium-90 and cesium-137 increases, for example, radionuclides are bonded most firmly in the clay fraction of the soil [21].

Retention of radionuclides in the soil is due to the presence in the soil of elements with chemical properties similar to those close to chemical properties of the radionuclides. Thus, calcium is very like close to strontium-90, and thus in particular, liming the soils with high acidity leads to an increase in adsorption capacity of strontium-90 and reduces its migration. Potassium is similar in chemical properties to cesium-137. Potassium, as nonisotopic analogue of cesium is in the soil in macro quantities, while cesium is presented in micro concentrations. Consequently, in the soil solution, trace amounts of cesium-137 are diluted by potassium ions, and while absorbing by plant root systems, they compete for sorption on the root surface. Therefore, penetration of these elements from soil to plants causes antagonism of cesium and potassium ions. Additionally, the effect of radionuclides migration depends on the amount of precipitation [22].

It is revealed that strontium-90, located on the soil surface, is washed away by rain into the lowermost layers. It should be noted that the migration of radionuclides in soil is slow and their main part is in the upper layer from 0 to 5 cm. Accumulation (removal) of radionuclides by agricultural plants depends largely on soil properties and biological peculiarities of plants. In acidic soils, the radionuclides are delivered to plants in large amounts as compared to slightly acidic soils. Reduction of soil acidity usually contributes to reducing the transfer of radionuclides into plants. Thus, depending on the soil properties, the content of strontium-90 and cesium-137 in plants may vary on average by 10-15 times [23].

Contamination of the environment by radionuclides is usually estimated using soil and vegetation sampling, however radionuclides affects productivity, dairy product quality and resistance of cattle to diseases. For example, in the Altai region of Western Siberia, under radionuclide contamination at a level of 10.1-50.0 REM (Roentgen-Equivalent-Man), the proportion of manifestations of leukemia in cattle was 36.9% of the total number of BLV-infected (infected by Bovine Leukemia Virus) animals, whereas in the Kemerovo region at

contamination with radionuclides at a level of 2.1-3.0 REM this proportion was 8.1%. In areas with radionuclide contamination less than 0.64 and 1.11-5.50 Ku/km<sup>2</sup>, the proportion of manifestations of leukemia was 2.0±0.1 and 4.2±0.4 %, respectively. In the farms with the soil contaminated with the <sup>137</sup>Cs at a level of 0.0 and 188 Bq/kg, the proportion of animals suffering from leukemia was 5.1 and 22.3%, respectively. At the same time, the contamination of territory with radionuclides had no effect on the incidence of cattle tuberculosis and brucellosis [7, 15, 17]. Thus, a transfer of radionuclides from contaminated soil and vegetation through water or forage pathway to farm animals is dangerous not only to humans, as radionuclides enter in human through livestock products, but they also worsen the health status of farm animals [6, 11, 12].

Thus, a necessary and promising recent trend of monitoring environmental parameters of farmlands is the assessment of the level of radionuclides (<sup>137</sup>Cs and <sup>90</sup>Sr) in different types of vegetable feedstuffs, and dairy products that is which are the most important indicators for environmental survey of ecological safety of agricultural regions in our country. This study presents the results on the content of radioactive elements in various types of the plant feedstuffs, stored up in different regions of Western Siberia, as well as in some dairy agricultural products (whole and dry milk) produced in the concerned regions.

### MATERIALS AND METHODS

Studies on accumulation of strontium-90 and cesium-137 by plants were carried out at Veterinary laboratories located in the territories of Novosibirsk, Kemerovo and Tomsk regions. Samples of feedstuffs were collected in different agricultural farms. Preparation of samples in terms of content of radionuclides was carried out in accordance with the requirements of SanPiN 1.3.2.560-96 (Russian sanitary regulations and norms) and MUK 2.6.1.1194-03 (Russian procedural guidelines). In total, we investigated 179 samples of hay from natural acreages, 85 haylage samples, and 147 samples of silage, 117 samples of grass from natural acreages, and 186 samples of concentrated feedstuffs.

Samples of herbage were taken before animals grazing or cutting for forage. Composite samples were formed from the three portions, taken on the areas located within the triangle and spaced from each other at a distance of 50-100 m. The height of cut was 3-5 cm from the ground. Distance from highways was more than 200 m. The samples' mass varied within the range from 4 to 5 kg. Hay samples were collected in 4-8 points at the height of 1- 1.5 m from the ground surface from all available sides of haystacks at a depth of at least 0.5 m. Composite sample with the weight of 2-3 kg was prepared combining individual samples. Analytical feed sample weighing 1-2 kg was prepared by mixing 3-5 portions taken from different bags. Simultaneously with the sampling, natural background radiation rate was measured using radiometers IMD-5 and RKS-107.

The activity of <sup>137</sup>Cs and <sup>90</sup>Sr was measured employing a universal spectrometric complex (USC) "Gamma+". The device includes two detection units. To assess the content of <sup>137</sup>Cs we used the method of native samples. The measurements were performed using a semiconductor gamma-spectrometer with the detector unit located in a lead shield ( $\gamma$ -path). The same device was employed to determine the activity of <sup>90</sup>Sr using a beta spectrometer ( $\beta$ -tract). Hygienic assessment of feedstuff in terms of radiation safety criteria was performed using the current regulatory documents.

Sampling of milk and milk products as well as their preparation for analysis was conducted in accordance with the requirements of GOST 26809-86 "Milk and dairy products. Acceptance rules, sampling and preparation methods for analysis" (All-Union State Standard, Russia). In total, 290 samples of whole milk and 98 samples of dry milk taken in Novosibirsk and Tomsk region were studied. Measurement of <sup>137</sup>Cs and <sup>90</sup>Sr activity was performed employing a universal spectrometric complex (USC) "Gamma+" using the "Progress" software. Preparation of samples in terms of radionuclides content was carried out in accordance with the requirements of SanPiN 1.3.2.560-96 and MU-13-7-2/1056. It included samples grinding and dry ashing in a furnace at controlled temperature regime. For measuring <sup>137</sup>Cs, we used a semiconductor gamma-spectrometer with the detector unit located in a lead shield ( $\gamma$ -path). When measuring <sup>137</sup>Cs, we employed a beta spectrometer ( $\beta$ -tract).

The data are presented as mean  $\pm$  standard error. The data were analyzed by 2-way ANOVA using the package «STATISTICA 8.0». All data were tested for normality before statistical analysis by the Shapiro-Wilk criterion. In the absence of the normal distribution, we used methods of nonparametric statistics: Kendall

correlation coefficient ( $\tau$ ) instead of the Pearson correlation coefficient ( $r$ ), and the Wald-Wolfowitz run test, which is an analog of Student's t-test.

### RESULTS AND DISCUSSION

The South of Western Siberia is the territory most developed in terms of industry and agriculture as well as most complicated by its biogeochemical conditions. Here, natural and anthropogenic factors cause a significant negative change in soil quality and forage crops in agricultural lands, both arable and natural. The industrialized Kuzbass (Kuznetsk basin, Kemerovo region in Russia) is characterized by catastrophic technogenic environmental pollution, destruction of soil and vegetation cover, which is caused by coal mining. In this regard, we can recall that coals were the first commercial sources of uranium in our country [13]. Hence, it is clear that their open mining and burning at thermal power plants is the strongest source of environmental pollution with radionuclides. According to some estimates, in the areas of coal-fired plants, the degree of contamination with radionuclides can exceed the pollution generated by nuclear plants with equal capacity. The toxic effects of many emissions of thermal power plants are multiplied at their integrated effects in case of penetration into living organisms [20].

Radionuclide contents in different plant feedstuffs from three regions of West Siberia are shown in Table 1. It was revealed that the maximum content of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  was detected in the natural grass hay, regardless of region. At that, the average concentration of  $^{137}\text{Cs}$  in Kemerovo region is much higher than that in other regions. This is probably due to anthropogenic pollution of environmental objects and orographic specifics of the Kemerovo region, which is located mostly in the Kuznetsk basin. The content of  $^{137}\text{Cs}$  in the silage and compound feedstuffs from Tomsk region was significantly higher ( $p < 0.001$ ) than that in samples from the Novosibirsk and Kemerovo regions. The highest phenotypic variation in radionuclide accumulation was revealed in silage from the Tomsk region, as well as compound feedstuffs and natural grass hay of from the Kemerovo region (Table 1). There was a notable variability of the  $^{137}\text{Cs}$  content in animal feedstuffs. Thus, in some samples, the content of  $^{137}\text{Cs}$  was 35 times higher than that in other samples. Nevertheless, none of the studied samples contained radionuclides at amounts exceeding the permissible levels (VP 13.5.13/06-01). The degree of radionuclides accumulation depended not only on type of feedstuffs, but also on region. For example, the concentration of  $^{137}\text{Cs}$  in the natural grass hay in the Kemerovo region was significantly higher than that in Tomsk and Novosibirsk regions (3.1 and 2.4-fold, Table. 1). In the samples of natural grass hay in Kemerovo region, the concentration of  $^{90}\text{Sr}$  was 2.0 and 1.3 times higher than in the samples from Novosibirsk and Tomsk regions ( $p < 0.001$ ). The highest variability in the accumulation of this radionuclide was noted in natural grass hay from Tomsk and Kemerovo regions, as well as silage from Tomsk region. In some feedstuff samples the concentration of  $^{90}\text{Sr}$  was at 56.8% higher than in the other samples, though, neither of the samples contained strontium in concentrations exceeding permissible and reference levels (SanPiN 2.3.2.1078-01. 13.5.13/06 – 01).

**Table 1. The content of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  (nKu/kg) in various types of feedstuffs from different regions of Western Siberia**

Type of feedstuffs	Kemerovo region		Novosibirsk region		Tomsk region	
	$^{90}\text{Sr}$	$^{137}\text{Cs}$	$^{90}\text{Sr}$	$^{137}\text{Cs}$	$^{90}\text{Sr}$	$^{137}\text{Cs}$
Grass of natural pastures	38.0±2.0	31.0±1.0	32.0±2.0	16.0±2.0	39.0±2.0	25.0±1.0
Natural grass hay	92.0±8.0	85.0±6.0	46.0±2.0	46.0±2.0	54.0±3.0	72.0±2.0
Haylage	30.0±2.0	25.0±1.0	23.0±1.0	13.0±0.9	11.8±1.0	59.4±6.3
Silage	25.0±1.0	24.0±1.0	21.0±2.0	13.0±1.0	20.0±1.0	29.0±1.0
Concentrated feedstuffs	21.0±1.0	22.0±2.0	16.0±1.0	9.0±1.0	36.0±2.0	58.0±2.0

Table 1 shows the impact of ecological and climate factors on the accumulation of radioactive  $^{137}\text{Cs}$  and the influence of technogenic pollution on its accumulation in by various types of feedstuffs. The content of this radionuclide in concentrated feedstuffs and haylage depends to the greatest extent on ecological zone ( $p < 0.001$ ). It may be supposed that this is, partly, due to peaks in its concentration in the feedstuffs of the

Tomsk region. We determined the differences in the accumulation degree of <sup>90</sup>Sr and <sup>137</sup>Cs in forage, depending on the region that was 1.6 times lower for the first radionuclide than for the second one. Influence of ecological-climatic zone on the content of both radionuclides in the silage was consistently high (for <sup>90</sup>Sr  $r_w = 0.58$ , for <sup>137</sup>Cs –  $r_w = 0.52$ ). A sharp decline in the influence of environmental factors on the concentration of <sup>90</sup>Sr was observed in the grass of natural pastures and silage, which respectively was 9 and 3 times lower than that for <sup>137</sup>Cs. The influence of foodstuff type on the level of <sup>90</sup>Sr in the Novosibirsk region was the highest ( $r_w = 0.70$ ), 1.7 and 1.3 times higher than that in Tomsk and Kemerovo regions, respectively.

Thus, monitoring of the radionuclides concentration (<sup>90</sup>Sr and <sup>137</sup>Cs) in feedstuffs of plant origin allowed estimating the role of ecological and climatic factors, as well as the type of plant feedstuff in the radionuclides accumulation. The minimum content of radionuclides in most types of feedstuffs was observed in the Novosibirsk region in comparison to the Tomsk and Kemerovo regions; however, their levels did not exceed the MPC.

We have also assessed the content of <sup>137</sup>Cs and <sup>90</sup>Sr in whole and dry milk produced at the enterprises of Novosibirsk and Tomsk regions (Table 2). It was found that their concentration was many times lower than MPC [5]. Thus, concentrations of <sup>90</sup>Sr and <sup>137</sup>Cs in the whole milk from the Novosibirsk region were respectively 18 and 62 times below the MPC. Similar data were obtained with regard to concentrations of radionuclides in dry milk produced in Novosibirsk and Tomsk regions (Table 2). Dry milk was characterized by greater accumulation of <sup>137</sup>Cs as compared to whole milk, whereas such dependence was not noted for <sup>90</sup>Sr.

**Table 2. The content of <sup>90</sup>Sr and <sup>137</sup>Cs in whole and dry milk (Bq/kg) from Novosibirsk and Tomsk regions.**

Product		Novosibirsk region		Tomsk region	
		<sup>90</sup> Sr	<sup>137</sup> Cs	<sup>90</sup> Sr	<sup>137</sup> Cs
Whole milk		0.80±0.15	2.70±0.20	0.80±0.02	0.24±0.03*
	lim	0-18.0	0-22.0	0-2.8	0-2.8
	MPC	25	50	25	50
Dry milk		0.80±0.40	7.2±0.50	0.48±0.03*	0.20±0.03*
	lim	0-24.0	0-32.5	0-0.60	0.17-0.40
	MPC	200	360	200	360

\* - significant differences between the regions ( $p < 0.001$ ).

As known, there are significant interspecies differences in agricultural crops in terms of the ability to accumulate radionuclides. For example, legumes absorb strontium-90 and cesium-137 by 2-6 times more intensively than cereals. Intake of strontium-90 and cesium-137 by grass stand in the meadows and pastures is determined by the nature of their distribution in the soil crossover. The accumulation of radionuclides in the herbaceous plants depends on the characteristics of the sod layer structure. In the meadow grass with strong dense sod layer, the content of cesium-137 in the phytomass is 3-4 times higher than that in the forb meadow with a loose thin sod layer [19].

Intake of cesium-137 in plants depends on soil type. In terms of cesium accumulation, the soils can be ranged in the following order: soddy-podzolic loamy sandy soils, soddy-podzolic loamy soils, gray forest soil, chernozems, etc. The accumulation of radionuclides in plants depends not only on soil type, but also on the biological characteristics of plants. It is noted that the calcium-demanding plants usually absorb more strontium-90 than plants poor in calcium. Strontium-90 is accumulated most of all by legume crops, in a less degree – by root crops and tubers, and even less – by cereals. Cultures with low content of potassium accumulate less amount of cesium. Grasses accumulate less amount of cesium in comparison with legumes. Though plants are relatively resistant to radioactive exposure, they can accumulate such amount of radionuclides that they become not suitable for use as animal feedstuff or food production.

The accumulation of radionuclides in plants depends on the content of nutrients in the soil. Thus, it is recognized that mineral fertilizer applied in doses of N 90, P 90, increase the concentration of cesium-137 [21,

22]. All the activities carried out currently to improve the soil fertility will help to reduce the intensity of the radionuclides transfer in plants at pollution of agricultural land with radioactive fallout. The simplest and cheapest method of reducing the content of radionuclides in agricultural products is the selection of crops and varieties, characterized by their ability to accumulate the minimum amount of strontium-90 and cesium-137. As a rule, these are varieties with a low content of potassium and calcium. Effective techniques include also plowing the contaminated topsoil, liming of acid soils and applying mineral and organic fertilizers. The correct choice of soil tillage depth and methods can significantly reduce the intake of radionuclides into the plants several fold [21].

### CONCLUSION

The findings of the current study allow concluding that the agricultural lands in Novosibirsk, Tomsk and Kemerovo regions are suitable for the production of ecologically safe forage and, consequently, crop production and animal husbandry, especially for the production of dietary and functional products as well as baby foods. However, it becomes clear that the radionuclide monitoring strategy should include food chain continuous monitoring of radionuclide content not only in the soil and in feedstuffs as the most popular monitor system, but also all kinds of agricultural foodstuff produced in Western Siberia.

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