REGULARITIES OF ^{137}Cs AND ^{90}Sr TRANSFER THROUGH THE CHAIN "SOIL - PLANT - ANIMAL" ON THE TERRITORY OF UKRAINE, CONTAMINATED AFTER CHERNOBYL NPP ACCIDENT

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As a result of Chernobyl NPP accident, more than 3 mln ha of arable lands and forests were contaminated to the levels requiring radioecological control and countermeasures. The main contributor to the total radioactivity of the soil and to the irradiation dose of human organism is ^{137}Cs; ^{90}Sr contribution is up to 10% of the total irradiation dose of a man. The most important factor of radiation danger on the territories, where the residence is permitted (density of soil contamination is less than 555 GBq/km^2 or 15 Ci/km^2) is inner irradiation resulting from radionuclide intake by the organism with food, mainly with milk and cattle meat.

The present paper is devoted to the examination of the role of basic factors, determining radionuclide transfer through food chain from soil to cow's ration and agricultural products; and to the assessment of possibilities to eliminate radionuclide contamination of these products.

Material and methods of research

Experimental researches, were carried out on the territory of Ukraine, which was more or less contaminated after the Chernobyl NPP accident. The main portion of research was carried out in natural conditions. With this aim samples of soil from all fields separately, plants in the phase of technical ripeness

milk and cow's meat in summer time were collected in 31 collective farm from 1987 annually. Samples of soil and plants after air-drying were milled. Samples of milk, with the volume of 3 l were evaporated up to 0,5 l; sample of meat was collected from 10-15 animals, milled and average sample with 3 kg mass was dried in vacuum-drying box.

^{137}Cs concentration in samples was measured in Marinelly cuvettes on gamma-spectrometer "ADCAM-300" with "Ortec" detector. ^{90}Sr content was measured by radiochemical method after ashing and solving of the sample. Precision of measurements of radiostrontium about $\pm 15\%$; radiocesium - $\pm 10\%$.

Regularities in the radionuclide transfer from soil to plants were also investigated in field experiments on the territory of Chernobyl NPP 30-km zone, Kiev and Rovno regions.

The main agrochemical characteristics of soil were also investigated - pH of salt extract, sum of adsorbed bases, exchangeable Ca and K (mg-eq/100 g soil), humus (%), content of mobile forms of nitrogen and phosphorus.

The farms are situated on the territories of Kiev, Gytomyr, Rovno, Chernigov regions, and the soil and climate conditions range from soddy-podzolic sandy and sandy-loamy acid soils - to leached mid-loamy chernozems with neutral or even slightly alkaline reaction. The brief characteristics of soil features of the region is given in the Table 1.

The sum of positive (higher than +10°C) temperatures and the amount of precipitation during vegetation period was determined according to the data of the nearest meteostations.

Prediction of expected levels of radioactive contamination of milk and meat is determined by prognostication of radionuc-

1. AGROCHEMICAL CHARACTERISTICS OF BASIC SOIL TYPES IN POLESSYE

Humus, %	pH salt	Hydrolitic acidity mg-eq/100 g	Adsorbed Ca mg-eq/100g	K_2O mobile, mg/100 g	Ashes, %	Adsorbed bases, mg-eq/100 g	P_2O_5 mg/100 g
			Soddy-podzolic sandy soil				
0,8	4,6	2,0	I,8	3,I	-	3,2	3,5
			Soddy-podzolic sandy-loam soil				
I,0	5,2	2,6	2,4	4,5	-	3,6	4,3
			Peat-swampy soil				
-	6,0	4,0	I8,6	7,5	30,07	23,I	6,5
			Light-grey podzolled sandy soil				
I,4	5,6	I,7	3,3	4,5	-	5,4	6,3
			Dark-grey podzolled slightly-loamy				
2,0	6,2	3,3	I0,3	5,2	-	I4,3	9,I
			Peaty soil				
-	5,4	0,8	-	5,0	42,90	-	2,6
			Podzolled slightly-loamy chernozems soils				
2,3	6,5	3,2	I4,2	6,4	-	I7,7	I2,3
			Typical chernozem loamy soils				
3,5	7,2	2,8	20,I	8,0	-	25,3	I0,4

lide accumulation in the harvest of forage crops.

Table 2 presents average values of the ^{137}Cs transfer factor from soil to forage plants, averaged for the period from 1987 to 1990. The intensity of transfer of radiocesium to crops have not decreased essentially within 4 years, but there is a notable tendency to it. Averaging error is $\pm 30\%$, which is acceptable for prediction estimates. Rich and concentrated fodder (grain) at equal contamination density have similar values of nuclide accumulation, while concentration in coarse forage is much higher. The nuclide concentration in the biomass of the most nutricious leguminous plants decreases in following order: vetch, clover, lupin, lucerne; differences between extreme members of this sequence are 5-folds. Radionuclide soil-to-plant transfer factor depends on the concentration of potassium and calcium.

Experiment data correspond to the Cadarash researchers conclusion that the prediction of TF of ^{134}Cs is possible, using K^+ and pH as predictors.

As it is shown above, specific differences essentially influence radionuclides accumulation in crops.

Differences, bound up with this factor may reach two orders of value.

Grasses, especially on natural meadows and pastures accumulate essentially more radionuclides. Differences of ^{137}Cs transfer factor to fodder crops reach 40-folds on acid and slightly acid soddy-podzolic soils, and 30-folds on neutral mid-loamy chernozem.

Annualy more than 30 values of transfer factor for each in-

2. Cs-137 TRANSFER COEFFICIENT FROM SOIL TO FORAGE CROPS

(average for 1987-1990), $\frac{\text{Bq/kg product}}{\text{kBq/m}^2 \text{ soil}}$

Crop	Soil type, pH salt extract			Differences, time
	soddy-podzolic sandy: 4,5-5,5	grey forest 5,6-6,5	chernosems 6,6-7,2	
Hay of natural grasses	10,00	4,00	1,80	5,5
Hay of sown grasses	4,00	3,00	1,60	2,5
Vetch	2,70	0,45	0,20	13,0
Clover	1,80	0,30	0,30	6,0
Lupin	1,50	0,40	0,15	10,0
Lucerne	0,80	0,40	0,20	4,0
Maize for silage	0,40	0,20	0,08	5,0
Root beet	0,50	0,35	0,20	2,5
Potatoes	0,25	0,13	0,045	5,5
Winter grain crops	0,50	0,20	0,05	10,0
Rye	0,40	0,10	0,04	10,0
Barley	0,30	0,10	0,06	5,0
Differences, time	40	40	30	

vestigated crop were measured' and this allowed to create data bank and to carry out statistical recessing of the results. Close connection of TC value with all explored soil characteristics was observed. However, these relations have exponen - tial character for all the parameteres, besides pH, and are convenient for predicting only the narrow sphere of soil charac - teristic values. It is important, that functional connection between TC and pH is observed, and TF values are equally sensible to pH value in wide range - from 4,6 to 7,6.

Distinctions in the capability to accumulate radionuclides, observed among basic forage crops, determine the influence of ration composition on radioactive cesium uptake by organism of animals. Differences between ^{137}Cs concentration in hay and biomass of forage crops are so considerable, that coarse forage cultivated on the territory with equal density of contamination make principal contribution to radiocesium content in the daily ration (Table 3): 43-68% when the cattle is kept stalled and 94-89% when pastured. Due to this ^{137}Cs absolute content in the ration of cattle grased on the cultivated pasture is approximately 2-4-folds more than when forage is produced on ploughed field. When cattle is grased or hay is stocked on natural pastures and hayfields these differences are more than 4-fold.

Such a great role of coarse forage in the formation of daily radionuclide ingress to the organism of cattle limits the possibilities of reducing radiocesium content in the ration by proper selection of root crops, tuber crops, concentrates and their sorts, as its portion in ^{137}Cs content in the ration is 2-14%. When cattle is kept stalled, considerable part of radio-

3. CS-137 CONTENT IN THE RATION OF CATTLE; FORAGE IS PRODUCED ON SODDY-PODZOLIC SOIL

(pH=4,5-5,5; density of contamination 1 kBq/m^2)

Type of keeping and feeding	Forage	Mass, kg	Cs-137		
			concentration, Bq/kg	content, Bq	% of the ration
Stalled; milk cattle average productivity	Hay	4	4,00	16,0	68,0
	Corn silage	15	0,40	6,0	26,0
	Beet, potatoes	2	0,35	0,7	2,9
	Concentrates	2	0,40	0,8	3,1
	Ration	23	-	23,5	100,0
Stalled; meat cattle	Hay	2	4,00	8,0	43,0
	Corn silage	20	0,40	8,0	43,0
	Beet, potatoes	5	0,35	1,7	9,2
	Concentrates	2	0,40	0,8	4,8
	Ration	29	-	18,5	100,0
Cultivated pasture	Hay	8	4,00	32,0	94,0
	Beet, potatoes	3	0,35	1,0	3,2
	Concentrates	2	0,40	0,8	2,8
	Ration	13	-	33,8	100,0
Natural pasture	Hay	8	10,00	80,0	98,0
	Beet, potatoes	3	0,35	1,0	1,2
	Concentrates	2	0,40	0,8	0,8
	Ration	13	-	81,8	100,0

nuclide enters the organism with corn silage - from 26 to 43% due to great mass of this kind of forage.

On the whole, regularities observed for ^{137}Cs accumulation by forage crops remain the same for ^{90}Sr, but the differences between rich forage and sown grasses are less (Table 4). On chernozem soils factor of nuclide transfer to plants decreased 1,6-2,3 times for 4 years. On sandy soils of Polessye, which are characterized with extremely low content of clay and mica minerals regularities in decreasing of ^{137}Cs ingress into fodder crops was not observed.

Thus, when fallout deposition is evenly distributed on the territory, level of ration contamination is determined by its composition, i.e., proper selection of crops and their sorts. This allows to decrease considerably radionuclide content in the ration. Fast-ripening (early) sorts of leguminous plants, grain-crops, potatoes with lower accumulation of K and Ca, accumulate 2-4,5 times less concentrations of ^{137}Cs.

Contamination of milk, when cattle is pastured, is determined not only by the type of soil, but also by pasture productivity. Observations, conducted in Gitomyr region showed that ^{137}Cs soil-to-milk transfer factor on low-productive slightly sodded pastures was 2,5-3,4 times more than on highly productive, where fertilizers are regularly applied (Table 5).

It should be noted, that all produced estimates concern only the case of even contamination of soil for all plants. The experience of elimination (liquidation) of Kyshtym and Chernobyl NPP accidents effects demonstrates that levels of farmland contamination considerably differ even within one farm, average

4. AVERAGE Sr-90 TRANSFER COEFFICIENT FROM SOIL PRODUCE IN 1987-1990

$$\left(\frac{\text{Bq/kg of produce in natural humidity}}{\text{kBq/m}^2 \text{ of soil}} \right)$$

Produce	Soddy-podzolic sandy soils; pH salt 4,5-5,5				Average for 4 years	Leached chernozems pH salt 6,5-7,2				Average for 4 years
	1987	1988	1989	1990		1987	1988	1989	1990	
Hay of cereals	110	96	130	130	116,5	20	18	16	12	16,5
Lucerne	80	70	69	75	72,5	20	16	12	9	14,3
Maize for silage	41	50	56	41	47,0	8	7	4	5	6,0
Root beet	10	16	18	15	14,8	7	5	3	3	4,5

5. Cs-137 SOIL-TO-MILK TRANSFER COEFFICIENT WHEN COWS ARE PASTURED (soddy-podzolic sandy soil, Gitomyr region; $Bq \cdot l^{-1}/kBq \cdot m^{-2}$)

Type of pasture	:	Transfer coefficient
Slightly sodded low-productive		$3,7 \pm 0,5$
Well sodded high-productive (NPK fertilizing 15 years)		$1,2 \pm 0,4$
Pasture after cultivation		$0,7 \pm 0,2$

area of which in the Soviet Union is 2500-3000 ha. These differences are determined both by existance of regular gradients of contamination level and by statistic micro- and mezo-heterogeneities the distribution of fallout deposition on the territories. Therefore different levels of soil contamination on separate fields will be observed within the territory of one farm. Within small farms these differences would be less, but neighbouring farms may differ considerably in density of radioactive contamination.

In case of existance of radioactive soil contamination gradients, proper organization of forage crop rotations is determining: coarse forage must be produced on fields or pastures with minimal contamination level and high level of fertility in order to eliminate (reduce) the quantity of radionuclide transfer to plants.

Characteristic feature of Ukrainian Polessye is prevalence of peaty, peat-swampy and meadow-swampy soils, where the transfer factor of cesium radionuclides is about 30-80, and for some lands-even 189 Bq/kg^{-1} of biomass; soil contamination is 1 $kBq \cdot m^{-2}$ for ^{137}Cs. Availability of pastures with such soils caused producing of milk in separate private farms with concentration much higher than the EEC (European Economic Community) standard (360 $Bq \cdot l^{-1}$), even when the density of soil contamination in rather low- 10-20 $GBq \cdot km^{-2}$. Consumption of milk in such regions may lead toexceeding inner irradiation dose 5 mSv on the territory with density of contamination 37 $GBq \cdot km^{-2}$. As the described landscapes make about 30% of the farmland in Polessye, they can be certainly called critical.

Prediction of radionuclide concentration in milk can be made on the basis of data on its content in daily ration. Accuracy on the prediction is determined to great extent by taking into account factors, influencing the transfer from gastrointestinal tract to blood, and then - to milk. Apparently, physiological characteristic features of an animal and the content of mineral elements in the ration play the most important role in the prediction of forage-to-milk TF value.

Table 6 contains radionuclide transfer factors from daily intake by kg/l of the product, averaged to within 30%. Using these data one has to make correction on animal productive milk cows have TF about 2-folds less than low productive ones. In the region, which was the most contaminated after Chernobyl NPP accident ^{137}Cs forage-to-milk transfer factor ranges from 0,5 to 1,7%.

As it is seen from the Table 7 differencies in the value of radionuclide transfer to cows milk depend to great extent on different content of K and Ca in the ration: in districts where K and Ca intake was about 61-84 and 37-65 g per day respectively, transfer to milk was on the average 2-folds less than in the districts where K and Ca intake reached 130-153 and 88-112 g per day.

Polessye is geochemical province with endemically low confent of microelements in soil and biote. Adding the principal microelements to the animal ration within physiological norm led to the double decrease of radiocesium transfer to milk and meat of cows.

Experimental data of radionuclide factor of transfer to milk and meat obtained during 4-year period in the observation region

5. Cs-137 SOIL-TO-MILK TRANSFER COEFFICIENT WHEN COWS ARE PASTURED (soddy-podzolic sandy soil, Gitomyr region; $Bq \cdot l^{-1}/kBq \cdot m^{-2}$)

Type of pasture	:	Transfer coefficient
Slightly sodded low-productive		3,7 ± 0,5
Well sodded high-productive (NPK fertilizing 15 years)		1,2 ± 0,4
Pasture after cultivation		0,7 ± 0,2

6. CS-137 AND SR-90 TRANSFER FROM THE RATION TO MILK AND MEAT

(% by 1 kg /l/ of daily intake with the ration)

Product	Transfer coefficient	
	Cs-137	Sr-90
Cows milk	1,0	0,10
Beef	4,0	0,06
Pork	15,0	0,30
Mutton	8,0	0,10
Chicken-meat	50,0	40,00

are presented in Table 8. Transfer factor values for each soil group, which differ by pH of saltextract are averaged for 6-10 farms, located in the appropriate zone at factor' calculations average values of contamination density of the farm as a whole was used.

On the whole, differences in intensity of radionuclide transfer to milk reflect regularities of strontium and cesium transfer from soil to plants. Some decrease of transfer factor in time is observed, it is more strongly marked on heavy soils with high capacity of cation adsorption. As a result, differences in radionuclide content in milk between districts with different features increase in time after the accident.

Influence of Prussian blue on ^{137}Cs secretion with cows milk was explored during the experiment with 2 analogous groups, 10 head each, with by the beginning of the experiment were on 4-5 month of 3-4 lactation. Nuclide content in the ration was controlled during the experiment; average content was 17,4 $kBq \cdot day^{-1}$. Feeding Prussian blue to cows by 10 g per day with mixed forage led to essential, up to 5 folds decrease of nuclide concentration in milk by the 5-th day of experiment and remaine on the same level (Table 9). When Prussian blue feeding stopped, cesium secretion with milk preserved its value for three days, then began to increase, and by 7-th day reached initial level.

Influence of forage addition "Humolite" on ^{137}Cs secretion with cows milk was investigated in the experiment with 2 analogous group, 10 head each, which were on the 4-5 month of lactation. The preparation was devised by Hungarian firm "Catalyst" on the basis of natural organic-mineral sorbents and is intended

7. INFLUENCE OF CA AND K CONTENT IN THE COWS ' RATION ON CS-137 AND SR-90 TRANSFER TO MILK

(% per 1 of daily intake with the ration)

District, region	Time of observation (year)	Average content in the ration, $g \cdot day^{-1}$		Transfer to milk	
		Ca	K	Cs-137	Sr-90
Rovno region; Rakitnoye district	1987	43 ± 4	73 ± 5	1,6	0,30
	1990	37 ± 4	61 ± 3	1,6	0,30
Gitomir region; Narodichi district	1987	59 ± 4	84 ± 7	1,3	0,25
	1990	102 ± 9	140 ± 11	0,6	0,10
Chernigov region; Repkino district	1987	60 ± 7	74 ± 7	1,4	0,20
	1990	100 ± 11	141 ± 13	1,3	0,10
Kiev region; Polesskoye district	1987	65 ± 6	74 ± 7	1,4	0,25
	1990	112 ± 11	141 ± 13	0,7	0,10

8. COEFFICIENT OF Cs-137 AND Sr-90 TRANSFER TO MILK AND MEAT OF CATTLE

$\left(\frac{\text{Bq/kg}}{\text{kBq/m}^2 \text{ soil}}\right)$

Product	Soil pH salt	Cs-137				Sr-90			
		1987	1988	1989	1990	1987	1988	1989	1990
Milk	4,5-5,5	1,5	1,2	1,2	0,9	3,0	2,5	3,0	3,4
	5,6-6,5	0,5	0,6	0,3	0,2	1,9	1,4	1,0	1,2
	6,6-7,2	0,2	0,2	0,1	0,1	0,9	0,5	0,4	0,2
Meat	4,5-5,5	2,0	2,4	2,0	1,7				
	5,6-6,5	0,6	0,7	0,6	0,7				
	6,6-7,2	0,5	0,3	0,2	0,2				

9. CHANGES OF Cs-137 SECRETION WITH MILK OF COWS UNDER THE INFLUENCE OF PRUSSIAN BLUE, 10 g daily with mixed forage

Group	Period after the beginning of the experiment, days						
	1	2	3	4	5	6	7
Control	1,4±0,1	1,3±0,05	1,3±0,1	1,4± 0,1	1,25±0,1	1,3±0,1	1,3±0,1
Experimental	1,4±0,1	1,2±0,1	0,89±0,07	0,41±0,05	0,26±0,01	0,33±0,03	0,26±0,05
Concentration decrease, times	1,00	1,08	1,46	3,41	4,81	3,94	5,00

10. CHANGES OF CS-137 CONCENTRATION IN COWS MILK AFTER THE FORAGE ADDITION "HUMOLYTE" INTRODUCING AND AFTER DISCONTINUANCE OF IT, Bq/l^{-1}

Group of animals	Period of introducing, days									Time after discontinuance of introducing				
	0	1	3	6	11	19	25	32	43	1	2	3	5	7
Control	204±12	203±12	200±15	204±15	191±14	188±12	189±18	197±19	192±11	199±12	197±12	193±11	190±14	196±14
Experimental	207±14	195±14	129±11	112±10	60±7	61±7	75±8	62±6	63±5	67±5	70±6	81±7	175±11	198±20
Concentration decrease; times	1,00	1,04	1,55	1,82	3,18	3,08	2,52	3,18	3,05	2,97	2,81	2,38	1,09	0,99

for long-time usage as a zootechnical forage addition in order to maintain animals productivity provide there needs for vitamins, microelements, and to introduce medicine, if necessary. The addition was introduced to the ration with concentrated forage at 7,5% of its weight (300 g daily per head).

On the third day after beginning of feeding the preparation, decrease of radionuclide concentration in milk is to be observed. On the eleventh day of the experiment, preparation ' effectiveness reaches its maximum - concentration decrease is 3,2 times and further it remains on the same level. After the introducing of preparation to the animals ' ration is stopped, its effect preserves for about 3 days, and then cesium concentration in milk goes back to the initial value.

The preparation did not influence the content of Ca, K and P macroelements in cows' blood, but it led to decrease of heavey metals Pb and Cd secretion with milk; under the influence of "Humolyte" secretion of Co from the microelements with milk decreased up to 1,6 times. Deviations in the biochemical status of animals resulting from preparation effect were not observed. Evidently, it is advisable to continue the exploration of this preparation effectiveness in the conditions of territory contamination with radionuclides and heavy metals.