Fluxes of Radionuclides in Agricultural Environments: Main Results and still unsolved Problems

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Abstract Agricultural products originating from the areas subjected to high radioactive deposit after the Chernobyl accident are a main contributor to the radiological dose to local populations. The transfer fluxes of radionuclides through agricultural food chains, to food products consumed by humans, depend on the characteristics of the ecosystems considered (first of all, on the soil type), on the type of agricultural product of concern, as well as on the physico-chemical properties of the radioactive element and its speciation in the released. The parameters describing the fluxes through the compartments of the agricultural ecosystem are dynamic; they change with the time after the accident an are strongly influenced by agricultural practice, including the application of countermeasures. The influence of these factors on the ^{137}Cs fluxes in the main agricultural ecosystems of the Chernobyl accident zone are quantitatively determined and the main topics, where further investigation is needed, are identified.

1. Introduction

Agricultural products originating from those regions in Belarus, Ukraine and Russia heavily contaminated after the Chernobyl accident in 1986 are an important source of additional radiological exposure of populations. On soils with light mechanical composition - soddy podzols and peaty soils - which are the most representative in the affected area, foodstuffs (principally agricultural produces) and water can contribute up to 90% of the total irradiation dose [1]. The remedial actions applied to control the fluxes in the various agricultural systems, as well as in natural and semi-natural biocenoses, allowed to limit the radionuclides transfer to humans, and hence the internal radiological dose delivered to individuals. These actions aiming at reducing the radionuclide fluxes trough food chains constitute the most effective way to reduce the total radiological dose to exposed populations; methods based on the reduction of the external dose exposure are substantially more difficult to apply and very expensive.

The intensity of the radionuclide fluxes trough the food chains depends on different factors. Among those, the most important ones are:

1. the type of (agricultural, semi-natural or natural) ecosystem considered,
2. the physico-chemical properties of the radionuclides, their speciation in the fallout, and
3. the change of these characteristics with time due to environmental factors (weathering, aging) and human interventions (agricultural practices and application of countermeasures) [2].

2. Influence of the type of ecosystem

The intensity of the radionuclide transfer through food chains, which governs the rate of radionuclides incorporation into human body, is mainly influenced by the agricultural land use pattern (meadows, pastures, arable soils etc.), the nature of the plant and/or animal productions on these lands (plant products -cereals, leafy vegetables, tubers -, fodder, milk, meat, ...) and specific biogeochemical parameters of the ecosystem considered.
The intensity of radionuclide fluxes in agroecosystems depends on whether or not, mechanical treatments (ploughing, discing, ...) are performed immediately after the fallout; in other words, the fluxes intensity on arable soils differs substantially from those on soils not disturbed (meadows, pastures) after the deposit. The mat present in meadows and pastures, where accumulated radionuclides remain weakly fixed and hence available for root uptake, is an important factor explaining the higher content of ^{137}Cs in meadows and pastures grass than in temporary pasture grass installed on arable soils. Moreover, the decrease of ^{137}Cs availability for plants (binding to the clay fraction) as a result of aging proceeds more slowly in meadows and permanent pastures than in arable soils, with the consequence that the ecological half-time in pasture and meadow plants is longer than that in plants grown on arable lands. A quasi-equilibrium situation considering the ^{137}Cs transfer from soil to plant in meadows and pastures in the Chernobyl zone was established approximately 5 to 6 years after the deposit whereas, for arable soils, a relative constant accumulation rate for ^{137}Cs in plants had already be reached earlier. These respective delays correspond to the time required to come to an equilibrium between the different water-soluble, exchangeable, acid soluble fractions of ^{137}Cs in the soil [3].

The type of soil and their physico-chemical characteristics are other important parameters ruling the radionuclide fluxes in agrocenoses. For instance, when ^{137}Cs TF's to different types of agricultural products grown on 4 different soil types representative of the Chernobyl zone (i.e. peat, soddy podzolic sandy soils, soddy podzolic loamy soils and chernozem) are compared, the highest values are obtained on peaty soil and lowest, on chernozem. The mean ratios of the transfer values for ^{137}Cs between these two extremes soil types amount to 10.0 and 26.6 for hay (respectively on amended and non-amended meadows), 8.3 for potato tubers, 32.5 for cereal grains. These ratios calculated for animal products are equal to 1 5.5 and 15.0 for beef and pork, and 10.0 and 26.6 for milk from collective farms and the private sector, respectively.

In the late phase after the accident, when production of foodstuffs with a radionuclide concentration exceeding DIL is practically excluded special attention should be paid to the restriction of the collective doses to the population, which are consuming contaminated food. In that case it is desirable to estimate the radionuclide fluxes in agricultural land per unit area, i.e. taking into account these products that are representative for main diet. These parameters may be used for planning and implementation of methods of agricultural practice in such a way that proposes the minimal collective doses and minimal risks of irradiation [4, 5].

The relative intensity of the radionuclide annual fluxes from the soil to the main foodstuffs produced in the areas of Ukraine and Belarus affected by the accident at the ChNPP are presented in Table 1. The ^{137}Cs fluxes to plant products (grain and potato) are, in general, higher than those to animal products (milk and meat). On both amended and non-amended lands (natural pastures and meadows), the ^{137}Cs fluxes to milk and meat are lower than the fluxes to potato, by a factor ranging form 2 to 24 and from 6 to 42, respectively; compared to the fluxes to cereal grains, respective differences by a factor up to 7 and 12, are observed. On another hand, the radiocaesium fluxes to meat and milk are similar on amended pastures and meadows (e.g. for lands where countermeasures were implemented to decrease the ^{137}Cs transfer to agricultural products), and non-amended lands, in the likeness of the fluxes to fodder (hay): a higher transfer on non-amended lands being compensated by a proportionally lower overall productivity.

Table 1. Annual ^{137}Cs fluxes in different agroecosystems in the Chernobyl accident area in 1994 (relative to a ^{137}Cs contamination density equal to 1 Bq/m^2)

Agricultural product	BELARUS (Data from the Byelorussian Institute of Agricultural Radiology)			UKRAINE (Data from the Ukrainian Institute of Agricultural Radiology)		
	productivity ($kg.m^{-2}.y^{-1}$)	TF (10^{-3} $m^2.kg^{-1}$)	flux (y^{-1})	productivity ($kg.m^{-2}.y^{-1}$)	TF (10^{-3} $m^2.kg^{-1}$)	flux (y^{-1})
Grains of cereals	0. 35	0.1	$3.5\ 10^{-5}$	0.45	0.3	$1.3\ 10^{-4}$
Potato	2.0	0.05	$1.0\ 10^{-4}$	2.5	0.18	$4.6\ 10^{-4}$
(non-amended lands)	0.1	6.4	$6.4\ 10^{-4}$	0.1	2.2	$2.2\ 10^{-5}$
Hay	0.083	0.64	$5.1\ 10^{-5}$	0.083	0.22	$1.9\ 10^{-5}$
Milk	0.006	2.6	$1.6\ 10^{-5}$	0.0125	0.88	$1.1\ 10^{-5}$
Meat						
(amended lands)						
Hay	0.3	2.2	$6.6\ 10^{-4}$	0.3	0.74	$2.2\ 10^{-4}$
Milk	0.25	0.22	$5.5\ 10^{-5}$	0.25	0.074	$1.9\ 10^{-5}$
Meat	0.018	0.88	$1.6\ 10^{-5}$	0.0375	0.29	$1.1\ 10^{-5}$

Table 2. Annual ^{137}Cs fluxes in different agroecosystems of the Bryansk region averaged over the period 1992-1994 (relative to a ^{137}Cs contamination density equal to 1 Bq/m^2)

Agricultural product	RUSSIA (Data from the Russian Institute of Agricultural Radiology and Radioecology)		
	productivity ($kg.m^{-2}.y^{-1}$)	TF (10^{-3} $m^2.kg^{-1}$)	flux (y^{-1})
Grains of cereals	0.3	0.2-1.3	0.6-3.9 10^{-4}
Potato	2.0	0.03-0.05	0.6-1.0 10^{-4}
Hay (non-amended lands)	nd	1.2-6.4	na
Hay (amended lands)	nd	0.47-1.4	na
Milk (collective farms)	0.04-0.06	0.047-0.14	1.9-8.4 10^{-6}
Meat (collective farms)	0.006-0.01	0.33	2.0-3.3 10^{-6}

Comparable data (annual productivity, transfer factors and estimated radionuclide fluxes) gathered in one of the most heavily contaminated area in Russia (Bryansk region, Novozybkovsky district) are reported in table 2. In this case, the ^{137}Cs relative fluxes to plant products are substantially higher (up to a factor 10 to 20 in average) than those to meat and milk. The ^{137}Cs fluxes to plant foodstuffs (potatoes and cereal grains) are in the same order of magnitude; those to animal products (milk and meat) are also similar. The differences in ^{137}Cs flux intensities observed for the same agricultural products (plants as well as animal products) between the three republics (tables 1 and 2) are essentially explained by differences in soil type, crop yield and animal productivity.

This approach, considering fluxes instead of transfer factors, has the main advantage that it reflects the most important characteristics of the contaminated areas, including the influence of soil type and the specific productivity of the foodstuffs. However, it has the weakness that it does not take into account the real land use nor the agricultural management structure on the contaminated territories. Therefore, in order to overcome this drawback and to obtain a more realistic evaluation of the ^{137}Cs fluxes to agricultural products and of their change with time, we suggest to calculate the fluxes on actual foodstuffs production in the region considered, weighted by the radionuclide concentrations in each specific food product (table 3). The results provided by this method allow to draw two main conclusions. First of all, they show that in real situation the milk and meat pathways contribute the most to the radionuclide ingestion by the populations living in contaminated areas. Secondly, they demonstrate that the relative contribution of the different pathways varies depending on time evolved since the accident. This can be explained by the considerable changes of the ^{137}Cs concentration in different crops induced by the implementation of countermeasures. Moreover, the yields in the contaminated areas can also vary with time, due to restrictions imposed to the production of certain crops, as well as, to economical and social reasons.

Table 3. Annual ^{137}Cs fluxes in agriculture of Novozybkovsky district of Bryansk region in 1987 and 1992

Agricultural products	Total quantity produced in the district (10^3 t/y)	Average ^{137}Cs concentration in the product ($Bq.kg^{-1}$)	Total flux ($Bq.y^{-1}$)	Flux per unit of area ($Bq.\ y^{-1}.m^{-2}$)	Flux relative to the flux in cereal grains
1987					
Grain of cereals	65.0	187	12.2 10^9	4.1 10^{-5}	1.00
Potato	140.0	108	15.1 10^9	5.1 10^{-5}	1.25
Vegetables	13.0	112	1.5 10^9	5.0 10^{-6}	0.25
Milk	515	1820	1.5 10^{11}	5.8 10^{-4}	15.0
Meat	11.7	6300	7.4 10^{10}	2.9 10^{-4}	7.50
1992					
Grain of cereals	53.0	29	1.5 10^9	5.3 10^{-6}	1.0
Potato	129.0	26	3.4 10^9	1.2 10^{-5}	2.3
Vegetables	11.2	22	0.3 10^9	0.9 10^{-6}	0.1
Milk	38.0	220	8.3 10^9	4.4 10^{-5}	7.6
Meat	8.1	590	4.8 10^9	1.9 10^{-5}	3.3

The differences in intensity between the ^{137}Cs fluxes for different foodstuffs make clear which one is critical in terms of a source of irradiation of population. In the Chernobyl region, milk (and meat to a lesser extent) is undoubtedly the main contributor to the internal dose; the relative contribution of other foodstuffs like cereals (bread) and potatoes is less important. For this reason, restriction on milk (and possibly meat) production in zones where the radionuclide concentration is expected to exceed the DIL appears as the first action to be taken in the immediate aftermath of an accident. But, to decrease the radiological exposure (collective doses) in the longer term, the fact that, in some agricultural area, the ^{137}Cs fluxes to grain and potato can be higher than those to milk and meat must be kept in mind.

3. Influence of the physico-chemical properties of radionuclides, including their forms in fallout

Two main forms of ^{90}Sr and ^{137}Cs were released by the Chernobyl accident: fuel particles from the destroyed reactor core (mainly deposited in near zone , in a radius of 30 km from the ChNPP) and condensed forms (which traveled longer distances). This resulted in the fact that the fluxes of these two radionuclides in agroecosystems were dependent from the ChNPP distance and also exhibited a different dynamic pattern in course of time after the deposit. Indeed, the intensity of the radionuclide fluxes through food chains depends not only on their physico-chemical properties, but also on their speciation in the deposit. If the influence of the physico-chemical characteristics of the radioelement has been largely addressed in the past in agricultural radioecology, that of their speciation in the fallout is much less documented and constitutes a rather novel item. Most of the information regarding the behaviour and accumulation in plants of ^{90}Sr and ^{137}Cs deposited as fuel particles released from an exploded reactor core were obtained from the numerous investigations carried out in the near zone of the ChNPP (until 30 km from reactor). These studies have demonstrated that the ^{90}Sr and ^{137}Cs mobility in soil and availability for plant uptake depend on the interaction of two opposite processes:

1. on one hand, the weathering of the fuel particles leading to the subsequent release of the radionuclides from their matrices, making them available for root uptake and,
2. on the other hand, their concomitant fixation on the soil solid phase (exchange complex), which reduces their availability for plant assimilation.

In the areas where condensed form dominated, ^{90}Sr and ^{137}Cs were mobile and, subsequently, highly available for plant uptake. The condensed form of the Chernobyl ^{137}Cs was even more mobile than that from the global fallout. On the contrary, ^{90}Sr and ^{137}Cs released into the environment as fuel particles was less available for root uptake until the radioelements were leached from their matrix.

In the Bryansk region where the contamination was due to condensed forms, ^{90}Sr was present in soil, during the period 1986-1987, mainly in exchangeable forms, easily available forms for plant assimilation. On the contrary, in the near zone of the ChNPP, the ^{90}Sr bio-availability was very low, so that their soil-to-plant concentration ratios were, in the first years after the accident, 2 to 4 times lower than those calculated for ^{137}Cs. A lower ^{90}Sr availability compared to that of ^{137}Cs does not correspond to common knowledge regarding these radionuclides as learned from their behaviour in the global fallout, the data from the Kyshtym accident (1957) or research carried out on experimental systems artificially contaminated with ionic forms. In areas contaminated by fuel particles, the root uptake of ^{137}Cs decreased with time, due to irreversible fixation of this radionuclide on clay minerals (aging), whereas ^{90}Sr accumulation, on the contrary, progressively increased as a result of fuel particles weathering and ^{90}Sr redistribution within the soil profile. The exchangeable ^{90}Sr fraction increased rapidly from 1987 until 1990 and remained practically constant thereafter [6, 7]. ^{137}Cs aging in the soil, as the result of its gradual fixation on poorly reversible binding sites on clay minerals (namely *illite* type clays), is one of the most important factors governing its availability for plant uptake.

4. Role of countermeasures in agricultural practice

The experience gathered with the mitigation of the consequences of the Kyshtym accident in the South Urals (1957) and the Chernobyl accident (1986) has taught that the introduction of countermeasures in the agricultural practice is an effective way to decrease of the intensity of radionuclide fluxes through the soil-plant-animal-human food chains. The main attention is usually paid to soil-plant steps of the food chains, taking into account that soils represents the most important reservoir of radionuclides in a contaminated terrestrial environment.
On meadows, among the possible agrotechnical treatments, rotating ploughing and disking decrease the ^{137}Cs transfer to plants by a factor 1.2 to 1.8, ploughing results in reduction factor ranging from 1.8 to 3.3 times, ploughing with deep-placement of upper contaminated layer (to the depth up 0.5 m) reduces the plant contamination by a factor 8 to 16. A surface amendment of meadows promotes a Cs transfer decrease from 1.3 to 3.1 times, and a radical amendment, from 2.3 to 11.2 times.

On arable soils the most important countermeasures are liming and fertilization. Lime application leads to a 1.6 to 2.3 times decrease of the ^{137}Cs transfer to plants, addition of phosphorus and potassium fertilizers induces a 1.2 to 2.2 times reduction and manure, a 1.3 to 1.6 times decrease. Combined application of lime, manure and high P and K doses can be even more effective, with reduction factors ranging from 2.5 to 3.5.
For ^{90}Sr, agrotechnical and agrochemical countermeasures have similar efficiencies: deep-ploughing (up to 50 cm depth) decreases of the ^{90}Sr concentration in plants by 1.3 to 2.3 times, deep-placement of the upper layer allows a 1.6 times reduction, liming lowers the transfer by 1.1 to 2.7 times and application of mineral fertilizers induces a 1.1 to 1.9 times decrease.
On soils with light mechanical composition, the application of alumino-silicates can be a useful way to decrease of radionuclide uptake by plants The application of zeolites leads however to contradictory results, but, in the most favorable situation, this countermeasure can decrease the ^{137}Cs concentration in plants up to 15 times.
The application of high N-fertilizer doses must be avoided as it leads to an increase the ^{90}Sr and ^{137}Cs concentration in plants.
The efficiency of agricultural countermeasures depends on the time of their implementation: the earliest they are applied, the most effective they are.
In the period 1993-1995, the difficult economical situation in Russia caused a reduction of the restoration effort in the Chernobyl zone. As a consequence, the addition of fertilizers (mineral as well as manure) were diminished and, subsequently, the ^{137}Cs concentration in agricultural products increased. According to data from the Ministry of Agriculture and Food of Russia, the ^{137}Cs concentration in cereal grains and potatoes in the Bryansk region was increased, in 1993, in comparison with the 1991 situation by a factor about 3 (from 70 Bq/kg in 1991 to 26 Bq/kg in 1993 for grains and from 107 to 37 Bq/kg for potatoes). Between 1991 and 1993 the K_2O fertilization was decreased from 81 to 18 kg/ha.

5. Dynamics of the radionuclide fluxes in agroecosystems

The intensity of the radionuclide flux tends to decrease with time after the deposit. Two different ecological half-times (T_c), corresponding to periods of time, can be identified: the decrease is more rapid in the first years after the accident and slows down thereafter until a steady state is reached (Tables 4 and 5). The analysis of the results presented in table 4 shows that the ecological half-time for ^{137}Cs in milk, averaged over the whole observation period (1987-1992) amounts to 1.6 to 2.3 years, and 2.3 to 4.8 years, for Bryansk and Kaluga regions, respectively. The different half-times between the two regions can be principally attributed to differences in the extent of applied countermeasures. When the three first year only are taken into account, the T_c^1 for ^{137}Cs in milk in the districts of Bryansk and Kaluga are rather similar (from 0.8 to 1.8 y); lower values are obtained in areas like the Krasnogorsky (1.0 y) and Novozybkovsky (0.8 y) districts, in the Bryansk region, where countermeasures were started earlier and carried out to a greater degree. Considering the later period (1990-1992), T_c^2 for ^{137}Cs concentration in milk estimated for different districts in the Bryansk region vary from 2.6 to 4.7 y, and in the Kaluga region, between 5.7 and 11.7 y. This discrepancy between the two region is again in agreement with the scale of countermeasures carried out in each respective region. Moreover, in areas where early countermeasures were taken, and completed before

1990, the ratio between the half-times of the first and second periods is larger (3.5 at Krasnogorsk to 4.5 at Novozybkov) than at other locations in the same region, because the countermeasures influence on the changes in ^{137}Cs levels in milk during the second period is still appreciable.

Table 4. Half-time periods of ^{137}Cs concentration decrease in milk, T_c (y) [8]

Types of production	T_c (y) Whole period (1987-1992)	T_c^1 (y) First period (1987-1989)	T_c^2 (y) Second period (1990-1992)
Bryansk region			
Gordeyevsky	1.7	1.2	2.6
Klimovsky	1.8	nd	4.7
Klintsovsky	2.3	1.3	2.6
Krasnorsky	1.8	1.0	3.5
Novozybkovsky	1.6	0.8	4.2
Kaluga region			
Khvastovichsky	2.3	1.5	6.3
Ulyanovsky	4.8	1.8	11.7
Zhizdrinsky	2.9	1.1	5.7

Table 5. Half-time periods of ^{137}Cs concentration decrease in plant products, T_c (y) [8]

Types of production	T_c (y) Whole period (1987-1992)	T_c^1 (y) First period (1987-1989)	T_c^2 (y) Second period (1990-1992)
Bryansk region			
Grain	1.4 - 1.9	1.2 - 2.1	1.7 - 4.3
Potato & root crops	1.5 - 2.8	1.4 - 1.9	2.2 - 7.7
Hay	1.5 - 1.9	0.9 - 1.4	1.7 - 7.3
Kaluga region			
Grain	3.1 - 4.9	nd	nd
Potato & root crops	3.2 - 5.6	2.9	5.8 - 14.1
Hay	2.0 - 3.0	1.4 - 2.3	3.3 - 4.8

Similar conclusions are also valid concerning the decrease of the ^{137}Cs concentrations in plant products. In most cases, the T_c calculated for the main plant products are in the same order of magnitude than those for milk. In the Bryansk region, the ecological half-times range from 0.9 to 2.1 y and from 1.7 to 7.7 y for T_c^1 and T_c^2, respectively. The corresponding values estimated for the districts of Kaluga region vary from 1.4 to 2.9 and from 3.3 to 14 y. Thus, the overall T_c, estimated over the whole period 1987-1992, for all types of plant products in the districts of the Bryansk region are considerably less (1.4 to 2.8 y), than in the districts of the Kaluga region (2.0 to 5.6 y), in agreement with the pattern of countermeasures implementation in both regions. Half-times (from 0.7 to 1.5 y) quite similar to those reported here regarding the decontamination of agricultural products have been mentioned earlier [9].

As a general conclusion, the results presented here above demonstrate that the changes with time of the intensities of the radionuclide fluxes in agroecosystems is ruled by three main factors:

1. radioactive decay,
2. changes in the biological availability of radionuclides under the influence of biogeochemical processes and
3. application of countermeasures.

For contaminated areas in Russia, in regions where countermeasures were intensively applied, the relative contribution of these three processes to the decrease of ^{137}Cs content in agricultural products were estimated to 0.06-0.07, 0.33-0.36 and 0.57-0.61, respectively.

6. Some actual problems

The intensity of radionuclides fluxes in different types of agricultural ecosystem, which determine the internal irradiation dose, can be changed in the time course after the accidental contamination. Therefore the study of the main factors governing the radionuclides transfer processes is extremely important. The investigation of the long-term consequences of countermeasures implementation on the fluxes of radionuclides is also essential. Such information must be used for planning the agricultural practice and the countermeasure strategy on contaminated lands, in order to mitigate the negative consequences of a radioactive contamination.

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