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Monitoring in animal breeding in response to nuclear or radiological emergencies: Chernobyl experience

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ABSTRACT

The main techniques of animal product sampling used in different time periods after the Chernobyl accident are summarised and lessons learned from this analysis are presented. It was shown that simple instruments for measurement γ-radiation in the environment can also be effectively implemented for measurement of γ-emitters in animal products even though these were not originally developed to measure radioactivity in food. The lessons learned related to the major tasks of the monitoring such "what to sample", "where to sample" and "when to sample". The role and example of application of supplementary data on radionuclide of concern properties for sampling planning are also discussed. Based on the statistical analysis of the data obtained in the affected settlement it was shown that radionuclide concentration in the animal products can be fitted by the log-normal distributions whilst the dispersion of the logarithms of the activity ^{137}Cs concentrations in milk is not dependent on the local settlement specific factors. Based on these findings the novel approach for justification of the number of samples that should to be taken to obtain the GM estimate with predefined precision for given variability of the data is suggested.

1. Introduction

Contamination of products derived from animals can constitute the major fraction of the internal dose to the public after radiation accidents. Prister (1990) reported that 70–90% of the intake of ^{137}Cs in the affected areas of Ukraine was attributed to milk and beef for the first 5 years after the accident. Currently, in 25 Ukrainian settlements, average activity concentrations of ^{137}Cs in milk is higher than 100 Bq L^{-1}, reaching 1000 Bq L^{-1} in some samples (Levchuk et al., 2016; Labunska et al., 2018). High values for ^{131}I, ^{134}Cs and ^{137}Cs activity concentrations in animal products were also found in Belarus, Russia and some other countries after the Chernobyl accident (IAEA, 2006; Sanzharova et al., 2016; Institute of Radiology, 2016). Therefore, timely implementation of the radiological monitoring of animal products is one of the key options in the protection of the population in case of radiological or nuclear emergencies (IAEA, 2012; Jacob et al., 2009; Kashparov et al., 2005; Fesenko et al., 2006, 2013; Ulanovsky et al., 2011; Kashparov et al., 2018, Balonov et al., 2018). 35 years of implementation of the monitoring programs following the Chernobyl accident provided valuable experience which should be used for increasing the efficiency of emergency response in case of future radiological or nuclear emergencies.

In Belarus, Russia and Ukraine, radionuclide activity concentrations were measured annually in hundreds of thousands of samples of animal products (milk, meat, etc.) in order to ban or process the contaminated animal products for reducing internal exposure of the population (Fesenko et al., 2007a). In this case, if concentrations in milk exceeded the temporary permissible levels (TPLs), the contaminated milk was withdrawn from the food distribution system and used as a raw material to produce some dairy products such as butter, cheese, cream, casein, in which radiocaesium concentrations were much lower than those in the initial product (IAEA 2009, 2010). Contaminated meat was minced, dried and diluted with non-contaminated dry mince in a proportion to obtain the activity concentration of ^{137}Cs below the required permissible level (State Agro-industrial Committee of the Ukrainian SSR, 1986).

Until 1991, monitoring of radioactive contamination of milk in

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Ukraine was carried out mainly in big collective farms and bulk milk samples were taken. However, this approach was changed in 1991, when the dose criteria were introduced for a classification of the contaminated territories. It became particularly important to identify settlements with an average dose for the population in a range of 1–5.0 mSv y^{-1}, where inhabitants could get a compensation because of restrictions due to the Chernobyl accident.

According to the guide approved by the Ukrainian Ministry of Health, the effective doses in such settlements were calculated based on the average density of contamination (external dose) and average radionuclide concentrations in milk and potatoes produced in the individual households of every affected settlement (internal dose). To comply with Ukrainian guiding documents on the radiation protection introduced in 1991, the effective doses should be assessed for the inhabitants of the affected settlements based on samples of milk that should be taken from individual households (Likhtarov et al., 2012). Therefore, doses were calculated for selected individuals of some households and mean values for the settlements were assessed as mean values for those individuals. The approach based on measurements of radionuclides in bulk samples taken from the entire herd of cows did not fit the purpose because every individual household had a different number of cows of different productivity, which grazed in different pastures. Therefore, the focus of the monitoring was shifted from bulk samples taken from collective farms to individual samples taken from the private households producing milk and potatoes. From five to ten samples taken per year in every affected settlement from 1991 to 2011 were used for internal dose estimating (Likhtarov et al., 2012). Reliable assessment of the mean, geometric mean or median values of ^{137}Cs concentration in milk based on the samples taken from individual households was a very challenging task. This was because the concentrations of radiocaesium in the milk of cows grazing the same pasture could differ by an order of magnitude, even though the milk was sampled in one day (Khomutinin et al., 2001). Additionally, lessons learned from some other Chernobyl related studies demonstrated that the major fraction of overall uncertainty in data used for food safety assessments is directly associated with the sampling of food, whereas the uncertainty of measurements in food samples constitutes less than 10% of the total uncertainty (Mc Gee et al., 1995). Overall, these facts resulted in the need for optimizing the number of samples, the frequency and the sampling period during the year to obtain mean annual concentration of radionuclides in milk with the required accuracy (Khomutinin et al., 2001). Thus, the sampling procedure was important for both estimating the average annual effective internal dose to the population and making decisions on implementation of remedial options (Levchuk et al., 2016; Labunska et al., 2018; UIAR, 2018).

The paper is focused on the key aspects of the implementation of the sampling programs for radionuclides in animal food in areas of the former Soviet Union, which were affected by the Chernobyl accident. Some lessons learned and recommendations for improvement of sampling in case of future emergencies are also considered.

2. General issues

According to the IAEA safety glossary, monitoring represents the measurement of dose or contamination for reasons related to the assessment or control of exposure to radiation or radioactive substances, and the interpretation of the results (IAEA, 2019). For application in case of nuclear or radiological emergency, this term can be defined as the measurement of radionuclides activity concentrations in animal feed and water and animal products, to limit exposure to the public and the staff involved in animal production systems and animal health. The central point of this definition is radiation protection of the public as most of the efforts are applied for this purpose.

2.1. Radionuclides of concern

More than 150 radioactive isotopes can be released into the environment after a reactor accident. The significance of those radionuclides in terms of impact on contamination of animal products can substantially vary depending on the time lapsed after the deposition. Radioiodine (^{131}I, ^{133}I) normally provides the highest radiation risk for the first days or weeks after the accident, whilst radiocaesium (^{134}Cs and ^{137}Cs) and radiostrontium (^{89}Sr and ^{90}Sr) are the main contributors of contamination to animal products during the years and decades after the deposition. Monitoring of animal products should consider specific features of radionuclides of concern both in terms of the identification of the most important exposure pathways (and corresponding products) and capabilities for measurement of the most sensitive radionuclides with the required precision.

Both the extent of radionuclide absorption in the gastrointestinal tract (GIT) of animals, and the concentrations of radionuclides in the animal foodstuffs are significantly dependent on radionuclide properties (Fesenko et al., 2007c). The bioavailability of the radioisotopes of caesium and strontium in the fuel "hot" particle form was two orders of magnitude lower than in the water-soluble form (Kashparov et al., 1997; Yoschenko et al., 2009).

Information on parameters of radionuclide transfer from soil to feeds and further to the animal products can be used for initial identification of areas of high concern, i.e., areas where more detailed monitoring programs should be implemented. Equally important is that such data can be used to identify areas where monitoring in animal production systems can be shortened. Thus, some simple assessments made with the aid of the data on radionuclide transfer along animal food chains allows the optimization of monitoring programme, saving resources and labour required.

Most of the available data on radionuclide transfer factors to animals are for radionuclide transfer to muscle, milk and eggs. However, there are also data on radionuclide transfer to animal tissues other than muscle, which are commonly termed offal and can also form a part of the human diet (liver, kidney, lung etc.) (Fesenko et al., 2018). There are also many processed animal products, including a variety of milk and meat products consumed by the public. Data on food processing retention factors i.e., the fraction of activity of radionuclides that is retained in the food after processing can be easily used for estimating concentrations in the final products. The information on such parameters is given elsewhere (IAEA, 2009; Fesenko et al., 2018).

2.1.1. Radioiodine

^{131}I was the main contributor to thyroid doses through the pasture–cow–milk pathway because of the high activity released and its mobility in the environment (IAEA, 2006; Fesenko et al., 2007a, b; Bazyka et al., 2016). The total ^{131}I activity release was ~ 1760 PBq (IAEA, 2006), whereas the importance of other volatile radionuclides was lower either because of lesser release or because of shorter half-lives (IAEA, 2006; Steinhauser et al., 2014).

After initial contamination, the radionuclides were distributed between soil and plant surfaces. The deposition of radionuclides on soil and plants was a starting point for further radionuclide transfer in the environment, including transfer to the animal food chains.

Interception (i.e. fraction radionuclides initially retained by plants) and the rate of radionuclide loss from plants due to removal by wind, rain, through leaching and by cuticular abrasion are the main processes determining on-going concentrations of radionuclides in the fodder, in animals and animal products shortly after the deposition. Weathering is the parameter describing the rate of radionuclide losses from plants. After the Chernobyl deposition, effective weathering of half-lives ($T_{1/2}^{Eff}$) (i.e. half-life accounting for radioactive decay) ranged between 4 and 7 days for ^{131}I and between 8 and 11 days for caesium radioisotopes (^{134}Cs and ^{137}Cs), respectively (IAEA, 2009, 2010).

Table 1
Recommended values of transfer parameters for domesticated animals after Chernobyl accident (Fesenko et al., 2007b,c; Fesenko et al., 2009a,b, Fesenko et al., 2015; Howard et al., 2009).

	Absorption coefficient in GIT, f_1	Transfer coefficient to milk, F_m, d L^{-1}	Transfer coefficient to meat[a], F_f, d kg^{-1}	Half-lives (milk)				Half-lives (muscle)			
				a^1	$T^1_{1/2}$, d	a^2	$T^2_{1/2}$, d	a^1	$T^1_{1/2}$, d	a^2	$T^2_{1/2}$, d
Cattle											
^{90}Sr	1.6×10^{-1}	2.3×10^{-3}	1.3×10^{-3}	1.0	2.1	–	–	0.42–0.6	3.5–4.0		180–600
^{131}I	1.0	1.0×10^{-2}	6.7×10^{-3}	1.0	1.4	–	–	1.0	7		
^{137}Cs	7.3×10^{-1}	8.1×10^{-3}	2.2×10^{-2}	1.0	1.9	–	–	0.37–0.84	3.7–22.3	0.3–0.5	36–84
Sheep											
^{90}Sr	8.0×10^{-2}	3.0×10^{-2}	1.5×10^{-3}					0.9	3–23.5		40–300
^{131}I	1.0	3.0×10^{-2}	3.0×10^{-2}								
^{137}Cs	8.0×10^{-1}	2.8×10^{-2}	1.9×10^{-1}	1	2.0	–	–				
Goat											
^{90}Sr		3.0×10^{-2}	2.9×10^{-3}								
^{131}I		3.0×10^{-2}									
^{137}Cs		1.4×10^{-1}	3.2×10^{-1}								
Pigs											
^{90}Sr	1.4×10^{-1}	1.7×10^{-2}	2.5×10^{-3}	–	–	–	–	0.9	4–5		450
^{131}I	7.0×10^{-1}	–	4.1×10^{-2}	–	–	–	–				
^{137}Cs	7.3×10^{-1}	–	2.0×10^{-1}	–	–	–	–	0.57–1.0	2.5–15.9		28.6–330
Chicken											
^{90}Sr	6.0×10^{-1}	–	2.0×10^{-2}	–	–	–	–	0.9–0.98	2.7–3.8		16–26
^{131}I		–	8.7×10^{-3}	–	–	–	–				
^{137}Cs	7.0×10^{-1}	–	2.7	–	–	–	–				

[a] Data from IAEA, 2009, 2010.

Thus, during the early phase of the accident, the radionuclide concentrations in feed subjected to the direct contamination were dependent on the deposition density, ability of plants to retain radionuclides, and weathering self-cleaning of vegetation over time after the depositions (IAEA, 2006).

Radioiodine is almost completely absorbed into the gastrointestinal tract of animals (fractional absorption is 0.7–1) (Fesenko et al., 2007b). The transfer coefficient to milk (F_m) or to meat (F_f), defined as the ratio of the activity concentration in animal product or tissue (Bq L^{-1} or Bq kg^{-1}) to the daily radionuclide intake at the equilibrium (Bq day^{-1}), is normally used to quantify transfer to milk and the other animal products (Table 1).

The data in Table 1 are mainly based on a set of the comprehensive reviews of Russian language data specific to areas affected by the radiation accidents in the Chernobyl and Kyshtym accidents (Fesenko et al., 2007b, c; Fesenko et al., 2009a, b, Fesenko et al., 2015; Fesenko et al., 2018). Thus, they can be considered as recommended values for areas most affected after these accidents.

The transfer coefficient values for ^{131}I transfer to cow's milk based on the world data presented in the IAEA TRS 472 vary from 4.0×10^{-4} to 2.5×10^{-2} (d L^{-1}), with the geometric mean of 5.4×10^{-3} d L^{-1} and geometric standard deviation around that mean of 2.4 (IAEA, 2009, 2010). A single loss exponential model well describe data from single ^{131}I administration for 12–14 d after ingestion with the biological half-life of 1.4 d (Table 1). Overall, these data are also important for sampling justification depending on time after the accidental release of ^{131}I.

2.1.2. Radiocaesium

A few months after the fallout, contamination of fodder crops with radioisotopes of strontium and caesium occurs, mainly due to root uptake. The rate of the radionuclide root intake may vary by several orders of magnitude, depending on the environmental conditions (humidity and property of soil). The highest radiocaesium transfers were found to be in the wet peat soils (Histosols), specific to some districts of the north part of the Rivne region of Ukraine. High availability of radionuclides for root uptake was observed also in the normal peat soil, followed by sand, loam and finally clay. The lowest bioavailability was observed to be in the soils of the Voronezh and Tula regions of Russia and in the soils of the south of the Kiev region of Ukraine. The soil in all these regions is largely presented by chernozem (Phaeozems) soils (Fesenko et al., 1997; Levchuk et al., 2016; Maloshtan et al., 2017).

High values of ^{137}Cs transfer factors were also due to specific species' composition of natural plant cultures and soil properties (peat-bog soils on sands with very high organic matter content (>70%) and low clay minerals, low productivity and quality of grass, with extremely low content mobile phosphorus, exchangeable calcium and poor potassium supply) can also greatly contribute to high ^{137}Cs transfer from the soil to the vegetation (Marey et al., 1974; Bondar and Udinzeva, 1984). As a result, concentrations of radiocaesium in the milk of animals grazing on waterlogged peatlands was more than 100 times higher than those observed in areas with highly fertile, heavy soils, such as chernozem (IAEA, 2006). The ecological half-life of the radiocaesium concentrations in pasture plants in such environments may exceed 20 years in the late phase of accidents (Labunska et al., 2018). Therefore, the high radiocaesium activity concentrations in animal products can persist in such areas over a long time.

The transfer factor to fodder plants was found to increase in the following order: fodder beet, maize, perennial grasses, and finally natural grasses. It decreased with increasing clay content in the soil. Effective half-lives of ^{137}Cs transfer factors to the fodder between one and three years were estimated for the period up to 1990. After that time, the data shows no consistent trend and somewhat longer half-lives were suggested (IAEA, 2006).

For the initial years after the accident, the effective half-lives in milk were generally between one and four years (Fesenko et al., 1995; Shutov et al., 1993 Sanzharova et al., 1994; Ivanov, 2016).

Like iodine, radiocaesium is almost completely absorbed into the gastrointestinal tract of animals (fractional absorption coefficient is 0.67–0.93). Radiocaesium is an analogue of potassium and is, therefore, found in all soft tissues, particularly muscle. The geometric mean of ^{137}Cs transfer factors to muscles of agricultural animals amounted to 2×10^{-1} d kg^{-1} for sheep and pigs and up to 2.7 d kg^{-1} for chickens (Table 1) (Fesenko et al., 2009a;b; 2018). Biological half-life of radiocaesium in milk of 1.9 d is longer than that of ^{131}I.

The geometric mean of radiocaesium transfer coefficient to cow's milk at the equilibrium is 4.6×10^{-3} d L^{-1} and to beef - 2.2×10^{-2} d kg^{-1} (Table 1). Worldwide investigations have shown the variation in F_m to cow's milk values ranging from 0.5 to 1.3×10^{-2} d L^{-1}. Nevertheless, mean F_m values were less than 1.0×10^{-2} d L^{-1}, with GM = 8.1×10^{-3} d

Table 2
Temporary permissible levels (TPLs) for radionuclides in foodstuff after the Chernobyl accident in the USSR and permissible levels (PL) in Belarus, Russia and Ukraine now (IAEA, 2006; Balonov et al., 2018), Bq kg^{-1} or Bq L^{-1}.

Foodstuff	EU 1986	the USSR					Belarus		Russia		Ukraine	
		May 06, 1986		May 30, 1986	Dec 15, 1987	Jan 22, 1991	1999		2001		1997	
		^{131}I	Total β activity	$^{134+137}$Cs	$^{134+137}$Cs	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr
Milk	370	3700	370[a]	370	370	37	100	3.7	100	25	100	20
Infant food		370	370	370	185	3.7	37	1.85	40	25	40	5
Dairy products	600	18,500–74000	3700–18,500	370–1850	370–1850	37–185	50–200	–	100–500	25–200	100–300	20–100
Meat and meat products		–	3700	1850–3000	740	–	180–500	–	200	–	200	20
Eggs		–	1850 Bq/egg	1850	740	–	370	–	–	–	100	30

[a] Introduced from August 01, 1986.

L^{-1} – Table 1 (Fesenko et al., 2007c).

2.1.3. Radiostrontium

Radiostrontium transfer factor values from mineral soil to plants are normally higher than transfer factors observed for radiocaesium. The soil organic matter may strongly adsorb ^{90}Sr, and its accumulation by plants is inversely proportional to the content in the soil of the chemical macro analogues, namely, exchangeable calcium. Strontium transfer to plants from different soils increases in the following order: organic soil, clay soil, loam soil, sand soil (IAEA 2009, 2010). At the same time, transfer factors of ^{90}Sr and ^{137}Cs are rather comparable for plants growing on peat-bog organic soils.

The ecological half-lives of the ^{90}Sr concentration in plants in areas affected by the Chernobyl depositions ranged from two (sandy soil) to nine (clay soil) years. The transfer factors of ^{90}Sr from soil to plants on sands with a low content of organic matter were as much as 40 times higher than that observed from clay soils (Ivanov, 2016).

The ^{90}Sr apparent absorption coefficient values vary from 0.06 to 0.27, depending on animal species and age. Therefore, the transfer coefficients from feed to adult animals for strontium are lower compared to radioiodine and radiocaesium (IAEA, 2010), reflecting the requirements of animals in essential elements (Fesenko et al., 2007c). The behaviour of strontium is strongly influenced by that of its analogue, calcium, which is a homeostatically controlled essential element. The extent of calcium absorption in the gastrointestinal tract is governed by the animal's calcium requirement which depends on factors such as age, growth rate and milk yield (IAEA, 2009). Beresford et al., (2000) have suggested an inverse relationship between the ratio between the calcium intake and requirement and the true absorption of radiostrontium in ruminants. There is a considerable amount of data in the literature documenting changes in gastrointestinal absorption with age (IAEA, 2009).

The F_f values for adult cattle vary in a range from 2.0×10^{-4} d kg^{-1} to 6.2×10^{-3} d kg^{-1}, with an arithmetic mean and the standard deviation around that mean of $(2.1 \pm 1.8) \times 10^{-3}$ d kg^{-1} (Fesenko et al., 2009; Howard et al., 2009). The F_f values for sheep and pigs are higher than for cattle (Table 1). The transfer coefficient values for ^{90}Sr transfer to cow milk vary from 0.5×10^{-3} to 18×10^{-3} d L^{-1} (IAEA 2009, 2010; Fesenko et al., 2007b) with the geometric mean about 2.3×10^{-3} d L^{-1} (Table 1). It is also worth mentioning that there were no statistically significant differences between data on ^{90}Sr transfer to animals derived from the Kyshtym and Chernobyl accidents (Fesenko et al., 2007b).

2.2. Radiological criteria

Monitoring in animal production systems is a continuous process of sampling, measurements, assessments and recording the radionuclide concentrations in animals, animal feeds and animal products in respect of some radiological criteria. Such criteria are normally derived from the annual dose limits, which are considered as acceptable in some exposure situations. In general, the annual dose limit of 5 mSv was assumed as an acceptable level of human exposure under pre-Chernobyl conditions. However, no acceptable dose limits for radiological emergencies were specifically recommended at that time in the documents published by the ICRP and the IAEA (ICRP, 1977).

The distinguishing feature of the regulation implemented in the USSR after the Chernobyl accident was establishment of temporary dose limits of the average equivalent whole-body dose, which was achievable in the situation formed after the accident. These limits were intended to encourage implementation of the actions to reduce the dose levels to the public and to decrease the radionuclide activity concentrations in the food products.

The first temporary effective dose limit of 100 mSv was set for the first year (from 26 April 1986 until 26 April 1987). The next temporary annual dose limit of 30 mSv was accepted for the second year and the third limit of 25 mSv was suggested for the years 1988 and 1989. The derived criterion of the internal dose of the population from the radionuclide intake of contaminated food was set at 50 mSv for the first year after the accident (excepting the thyroid dose from ^{131}I), 8 mSv in 1988 and 5 mSv in 1991 (Balonov et al., 2018).

The annual dose limit of 1 mSv, that was five times lower than the previous one, was set after the disintegration of the Soviet Union in 1991 by the relevant national authorities. It is worth noting that this annual dose limit was considered as a long-term goal (IAEA, 2006) as it was also done after the Fukushima Daiichi accident (IAEA, 2014).

The annual dose limit of 1 mSv was based on the ICRP Publication 60 (1990) where the annual effective dose of 1 mSv was considered as acceptable for the members of the public in exposure situations termed as "a practice" (ICRP, 1991). Therefore, this level was also considered as a safe level in the three most affected countries in post-emergency conditions for the long term. The above levels were considered as a realistic goal and suggested based on expert assessment of accessibility to these levels with the assumption that most of the possible countermeasure options were implemented. Thus, an approach based on setting new annual temporary reference levels, served as a driving tool for remediation based on the principle of optimization. Considering this Chernobyl experience, the ICRP suggested a similar approach, naming a concept of reference levels that was implemented after the Fukushima accident.

Based on these dose criteria, for each year after the accident the TPLs for radionuclide activity concentrations in food were suggested (Table 2). A gradual reduction of these TPLs has been used as an instrument to oblige producers to apply technologies limiting internal exposure to the public (IAEA, 2006; Balonov et al., 2018).

The first TPLs approved by the USSR Ministry of Health on May 6, 1986 was concerned with the ^{131}I activity concentrations in some foodstuffs (Table 2). The permissible level for ^{131}I in milk was set as 3700 Bq L^{-1}, whereas the permissible level for ^{131}I in baby food,

including milk, cheese, cottage cheese and sour cream was set about 10-fold lower than those for similar adult food items.[1] Milk or sour cream was diluted with lower contaminated products if the concentration of ^{131}I in milk was above the TPLs. The TPLs concentrations of ^{131}I in food and drinking water fit the monthly dose limit of 0.3 Gy for thyroid, excluding the contributions of other iodine isotopes.

The TPLs approved on May 30, 1986 complied to an internal dose of 50 mSv or lower, with equivalent values for the TPL in 1988 of 8 mSv, and for a TPL in 1991 of 5 mSv (Balonov et al., 2018). A month after the Chernobyl accident, due to the radioactive decay of the short-lived ^{131}I (half-life of 8 days), the activity of this radionuclide in food items became lower than that of ^{134,137}Cs. The TPLs adopted on May 30, 1986 concerned the total beta activity in food but was primarily focused on radiocaesium[2] (Table 2). The latter TPLs introduced since 1988 and 1991 directly referred to the sum of ^{134}Cs and ^{137}Cs concentrations. The TPLs for both caesium radionuclides, and for ^{90}Sr, were set in 1991 (Table 2).

In 1997 - 11 years after the Chernobyl accident - Ukraine introduced post-accidental food safety standards for ^{90}Sr and ^{134,137}Cs. The list of food items that were under regulation was expanded in 2006 – Table 2 (MHPU, 2006). In Belarus (MHRB, 1999) and Russia (CSI, 2001), post-accidental food safety standards adopted for the whole territory of the Russian Federation were introduced in 1999 and 2001, respectively (Table 2).

Current national TPLs for food products, drinking water and wood in the three countries are comparable and all of them are substantially lower than the EU permissible levels for import (Table 2) (Balonov et al., 2018).

Japan introduced post-accidental food safety standards for radiocaesium in food items already one year after the accident. These post-accidental food safety criteria amounted to 50 Bq L^{-1} for milk and infant food and 100 Bq kg^{-1} for all other food (IAEA, 2015).

The Chernobyl-related regulation was also introduced in European countries. Council Regulation (EC) No 733/2008 of July 15, 2008 established the permissible levels of 370 Bq L^{-1} for milk and milk products and 600 Bq kg^{-1} for all other food[3] for activity concentrations of ^{134}Cs and ^{137}Cs in the products imported into European Union countries. In 2009, the document was extended to March 31, 2020 (Council Regulation (EC) No. 1048/2009 of October 23, 2009). Since 2012, the European Commission has been applying the same restrictions on food items imported from Japan. The permissible levels for import concerning ^{134}Cs and ^{137}Cs concentrations in agricultural products were taken as the same as the food safety standard for the products consumed in Japan (Commission Implementing Regulation (EU) No. 996/2012 of October 26, 2012).

3. Implementation of monitoring programs in areas affected by the Chernobyl accident

3.1. Sampling programs in the acute phase of the accident

3.1.1. Monitoring after the Chernobyl accident

Activity concentrations of ^{131}I and ^{137}Cs in cow's and goat's milk achieved maximum values after 1–2 days after the depositions. The increase of radionuclide concentrations in the muscles of animals, poultry or fish was slowly compared to that in milk because biological half-lives for muscles were much longer compared to those in milk (see Table 1). Therefore, bearing in mind rather high consumption rates specific to that product, milk was identified as a critical animal product that has required rapid implementation of the monitoring program.

In late April/early May 1986, dairy cows were already grazing outdoors in the areas surrounding the Chernobyl NPP and high concentrations of ^{131}I in cow's milk were achieved very rapidly (within about 1 day) after deposition. Concentrations of ^{131}I in cow's milk in the affected regions of Ukraine, Belarus and Russia reached hundreds of kBq L^{-1} in early May of 1986 (IAEA, 2006).

Radiation monitoring of milk and other dairy products was introduced only 1–2 weeks after the Chernobyl accident and only at the large milk processing plants. Simultaneously, measurements of radionuclide activity in milk were organized at the collective farms and were performed with the aid of the SRP-68-01 (Electron, Ukraine) dosimeters originally intended for use in geological surveys adopted for such measurements. Within the measurements, the scintillation detection units (NaI(Tl) 30 × 25 mm) wrapped with plastic bags were submerged into large tanks (cisterns, barrels, cans) containing contaminated milk. Activity concentrations of ^{131}I in milk at the TPL level, i.e. of 3700 Bq L^{-1}, complied to the increase of the dose rate level measured by that device of 50 μR h^{-1} (0.5 μSv h^{-1}).

During the first weeks after the accident, the main purpose of the application of countermeasures in the USSR was to decrease the ^{131}I activity concentrations in milk or to prevent contaminated milk from entering the food chain. To achieve that, the following measures were implemented (IAEA, 2006):

- radiation monitoring and subsequent rejection of collective milk at processing plants in which ^{131}I activity concentrations were above TPLs (3700 Bq L^{-1}).
- processing rejected milk (mainly converting milk to storable products such as condensed or dried milk, cheese or butter) in order to decrease the ^{131}I activity due to its radioactive decay.

In the early stages after the Chernobyl accident, it was not possible to measure in vivo the radionuclide concentration in animals because of the lack of the relevant techniques. A technique for in vivo measurements of ^{137}Cs in animals (live monitoring of animals) that allowed a reduction of the contaminated meat production was developed in 1987. In the absence of this method and in the face of a lack of clean forage for the animals, which had to be evacuated from the 30-km zone, 95,500 cattle and 23,000 pigs were slaughtered. Many corpses were buried and some corpses were stored in refrigerators that created great hygienic and economic problems. The issues faced following the Chernobyl accident provide some important lessons learned which are discussed in depth in the next section.

3.1.2. Animal monitoring: when to sample?

The Chernobyl experience allows for a conclusion that at an early stage of the radiation accident, milk sampling should be conducted starting from the first information on potential contamination at least once a day. Two stage sampling was organized; both at the farm, directly after milking, and at the milk processing plants at the time of receiving the milk for processing. Repeated (periodical) sampling of milk was carried out every 3–5 days for the period of high iodine contamination to determine the ecological half-lives of radionuclides in milk or pasture grass.

In vivo measurement of radiocaesium in animal tissue was also performed during animal fattening on the farms and before slaughtering at the slaughterhouse. Animal product sampling (meat, bones, forages, milk, eggs, wool, etc.) was carried out at the animal processing plants before shipping those products for sale in accordance with the established procedure. The sampling of milk and grass for radioactivity was conducted over two - three weeks following a change in the animal feedstuffs composition.

3.1.3. Animal monitoring: where to sample?

Exceeding the TPLs for radionuclides in animal products (Table 2) is

[1] After the Fukushima Daiichi accident, permissible levels for ^{131}I in dairy produce in Japan was set as 300 Bq L^{-1} (IAEA, 2015).

[2] After the Fukushima Daiichi accident, permissible levels for ^{134,137}Cs in dairy produce was set as 200 Bq L^{-1} (IAEA, 2015).

[3] Including food foraged from the forest.

likely to be observed if the daily radionuclide intake (Bq d^{-1}) is above $TPL/F_{m,f}$. For instance, the TPL of 100 Bq L^{-1} of ^{131}I and ^{137}Cs in cow's milk (Table 2) is likely to be exceeded for daily radionuclide intake above:

$$\frac{TPL}{F_{m,f}} = \frac{100\frac{Bq}{L}}{0.01\frac{d}{Bq}} = 10000\frac{Bq}{d} \quad (1)$$

A dairy cow with an average weight of about 500 kg normally consumes approximately 15–20 kg of feedstuffs a day (DW). Therefore, the activity of ^{131}I and ^{137}Cs is below TPLs of 100 Bq L^{-1} if the activity of ^{131}I and ^{137}Cs in the feed is below 500 Bq kg^{-1} (DW). In a similar way, it is possible to estimate the acceptable level of feed contamination for other radionuclides and products presented in Table 2.

Concentrations of radionuclides in the feed of dairy cows in the winter period when animals are kept indoors are normally lower than in the summer, when animals are mainly grazing outdoors. If the accident happens in late spring or summer, when animals are grazing on the pasture, the radionuclide concentrations in milk depends on their concentrations in pasture grass, which in turn depends on interception of radionuclides by plants.

Deposition density, type of deposition (wet or dry), meteorological conditions at the time of depositions and biomass of the plants should be listed among factors governing radionuclide interception by plants. The mass interception fraction (f, m^2 kg^{-1}), defined as the ratio of the radionuclide activity concentration, which is initially intercepted by the plant after the deposition event (A_v, Bq kg^{-1}, DW) to the total activity deposited, (A_s, Bq m^{-2}) (Pröhl and Hoffman, 1996; Karunakara, et al., 2018) can be calculated based on the following equation:

$$f = \frac{A_v}{A_s} \quad (2)$$

The mass interception fractions of radioiodine and radiocaesium by the grass measured after the Chernobyl accident varied in a range from 0.7 to 5.5 m^2 kg^{-1} for the wet deposition and 0.4–3 m^2 kg^{-1} for the dry deposition (Pröhl and Hoffman, 1996).

Thus, the radionuclide activity concentration in grass (A_v) is likely to exceed 500–700 Bq kg^{-1} in grass with the mass interception fraction of 3–5 m^2 kg^{-1} for the deposition density of ^{131}I and ^{137}Cs above 100–300 Bq m^{-2}.

Milk samples should be collected firstly in the regions where there is a probability of exceeding the above-mentioned levels of the areal contamination density of ^{131}I and $^{134,137}Cs$. At a contamination density of ^{131}I, ^{134}Cs and ^{137}Cs at the level of 200 Bq m^{-2}, the background radiation dose rate of about 0.1 μSv h^{-1} will be increased only by 3.0×10^{-4} μSv h^{-1}, 1.0×10^{-3} μSv h^{-1} and 4.0×10^{-4} μSv h^{-1} (Eckerman and Ryman, 1993), respectively. Such low dose rate increments cannot be identified based on measurements that are made by any modern dosimeters.

A reliable judgment on the increase of the external dose rate above the natural background by 0.1 μSv h^{-1} can be made at a density of surface contamination with ^{131}I of 75 kBq m^{-2}, ^{134}Cs of 20 kBq m^{-2} and ^{137}Cs of 50 kBq m^{-2} (Eckerman and Ryman, 1993). For such values of the air dose rate, the activity concentration of ^{131}I, ^{134}Cs and ^{137}Cs in the milk of cows may significantly exceed the TPLs for these radionuclides in milk and reach 38 kBq L^{-1}, 10 kBq L^{-1} and 25 kBq L^{-1}, respectively. Therefore, justification of the areas where the monitoring in animal breading is required in the early phase of the accident should be done combining simple estimates and data on the expected density of contamination of the territory by radionuclides or data of field in-situ gamma spectrometric measurements.

Table 3
Soil-specific ^{90}Sr and ^{137}Cs mean aggregated transfer factors (T_{ag}, Bq kg^{-1}fresh mass per kBq m^{-2}) from soil to animal products (Ulanovsky et al., 2011).

Product	Soil group	T_{ag}, Bq kg^{-1}fresh mass per kBq m^{-2}	
		^{134}Cs, ^{137}Cs	^{90}Sr
Milk	Sand	0.2	0.3
	Loam	0.07	0.2
	Clay	0.003	0.1
	Organic	0.6	0.06
	Wet peat	5	–
Beef	Sand	0.6	0.3
	Loam	0.25	0.2
	Clay	0.1	0.1
	Organic	2	0.06
	Wet peat	10	–

Table 4
The requirements to the eggs sampling in areas affected after the Chernobyl accident.

Number of sampled eggs	Number of sampled standard packages with eggs	Number of packages with eggs in the lot
20	1	<10
30	3	10–50
50	5	51–100
100	15	101–500
150	20	>500

3.2. Sampling programs in the mid and long term after the Chernobyl accident

3.2.1. Monitoring after the Chernobyl accident

In the mid and long term after the depositions, radionuclide activity concentrations in feedstuffs depend on both contamination density and the transfer factors of radionuclides to plants used for feeding animals. In turn, transfer factors from soil to plants depend on soil and plant properties and weather conditions. Depending upon the environmental factors mentioned above, mean values of the soil to plant transfer factors of radiocaesium to natural grasses can vary up to 10 times or more. (Ulanovsky et al., 2011). This results in high variations of radionuclide activity concentrations in animal products even for the same ^{90}Sr, ^{134}Cs and ^{137}Cs activity concentrations in soil (Ulanovsky et al., 2011). Therefore, current permissible levels (PLs) for radiocaesium in milk (100 Bq L^{-1}) and meat (200 Bq kg^{-1}) presented in Table 3 are likely to be exceeded at rather low soil contamination levels (Table 4).

A simple estimate assuming that the aggregated transfer factor from the soil to milk is 0.3 for sand soil (Table 3) shows that the PL for ^{90}Sr in milk of 20 Bq L^{-1} is likely to be exceeded in areas with contamination with ^{90}Sr at a level of around 70 kBq m^{-2} because:

$$\frac{20\ [Bq\ L^{-1}]}{0.3\ \left[\frac{Bq\ L^{-1}}{kBq\ m^{-2}}\right]} = 67\ [kBq\ m^{-2}] \quad (3)$$

Therefore, identification of areas where monitoring of animal production systems is greatly required should be based on both data on the contamination density and soil specific transfer factor values (Table 4). In doing that, special attention should be given to peat-bog soils for ^{137}Cs and to sandy soils with low organic matter content for ^{90}Sr (Maloshtan et al., 2017). Such an approach based on an application of the aggregated soil-to-animal product transfer factors was widely used for the monitoring of animal products in areas affected by the Chernobyl accident and was implemented in national food safety standards and guiding documents on environmental monitoring (SOU, 2006a).

3.2.2. Sampling of fodder

Sampling of fodder was carried out in places of cultivation,

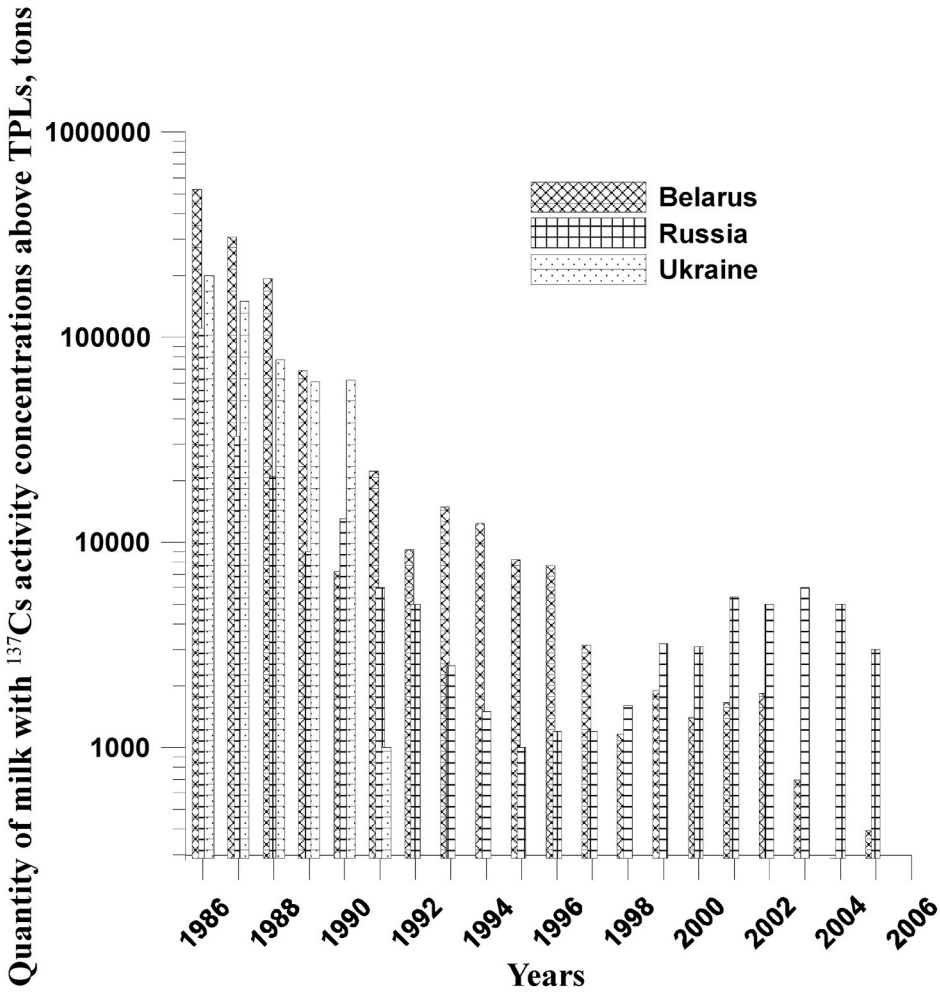

Fig. 1. Dynamics of milk quantities with ^{137}Cs activity concentrations above TPLs, produced in Belarus, Russia and Ukraine during first 20 years after the Chernobyl accident (Fesenko et al., 2007a).

production, storage and feeding of the animals. The mass of the average sample feed was determined from the calculation of one composite sample from every 5 tons (fresh weight) of fodder, unless otherwise recommended for the specific type of forage.

Grass and fresh mass of plants. Ten individual samples of grass weighing 0.5 kg each from pastures or hayfields were taken along a diagonal of a square with sides of around 100 m. From the fresh mass of plants supplied for feeding to animals or for the preparation of haylage or silage, at least 10–15 samples from different places were taken from dried plants. The weight of each such individual sample was also 0.5 kg as for pasture grass. Individual samples were mixed, spread out in an even layer and a composite sample was selected from the pooled sample.

Coarse forages (dry hay and straw). Samples of non-pressed coarse forages with a weight of 0.2–0.3 kg were taken from 15 to 20 batch locations at a depth of 50 cm or more. When samples were taken from a stack, the sampling points were being selected at equal distances along the perimeter at a height of 1–1.5 m. Three percent of the total amount of hay bales were taken if the weight of the lot of forage-laden feedstuffs was lower than 15 tons and 1% if the weight of the lot was above 15 tons. Individual samples with a mass of 0.2–0.3 kg were taken from each bale selected for examination. The feeds were sampled from different bale layers, after placing the hay on a tarpaulin and mixing. Special care was taken for preventing the breakage of plants and the formation of debris. Afterwards, based on such a procedure a composite sample was compiled.

Haylage and silage. The samples of haylage and silage were taken around 4 weeks after stowage or earlier. Samples were being taken from the stowage trenches or tower to a depth of 2 m or more. Point samples were taken in the centre and along the edges of the trenches or tower at a distance of 0.5 m from the walls, mixed thoroughly and a composite sample was formed from the pooled sample.

Root tubers (beets, potatoes, carrots, etc.) sampling was conducted directly in the fields, from the storage facilities and during loading for transportation. The samples, which were packed in bags, boxes, pallets and other containers, included 2–5% of packages (but not less than 3), from each lot. Three – five individual samples, i.e. whole root crops, thoroughly cleansed from the earth were taken from different layers (top, middle, and bottom) of the pack units. Ten - fifteen individual samples were taken from the piles of beetroots in the fields in the case of field sampling. Individual samples were combined to form a composite sample.

Concentrates (feed, grain, cake, meal, etc.). From a lot of concentrates, stored in bulk, individual samples were selected by a bucket or a cone sampler from the different pack layers (from the upper, the middle and lower layer. For sampling of concentrates packed in the bags, the sample included 2–5% of the pack-forging bags (but not less than 3 samples). Selection of individual samples was performed on each such pack-forging bag selected for sampling at 3 points (at the top, middle and bottom of the bag). The individual samples were combined,

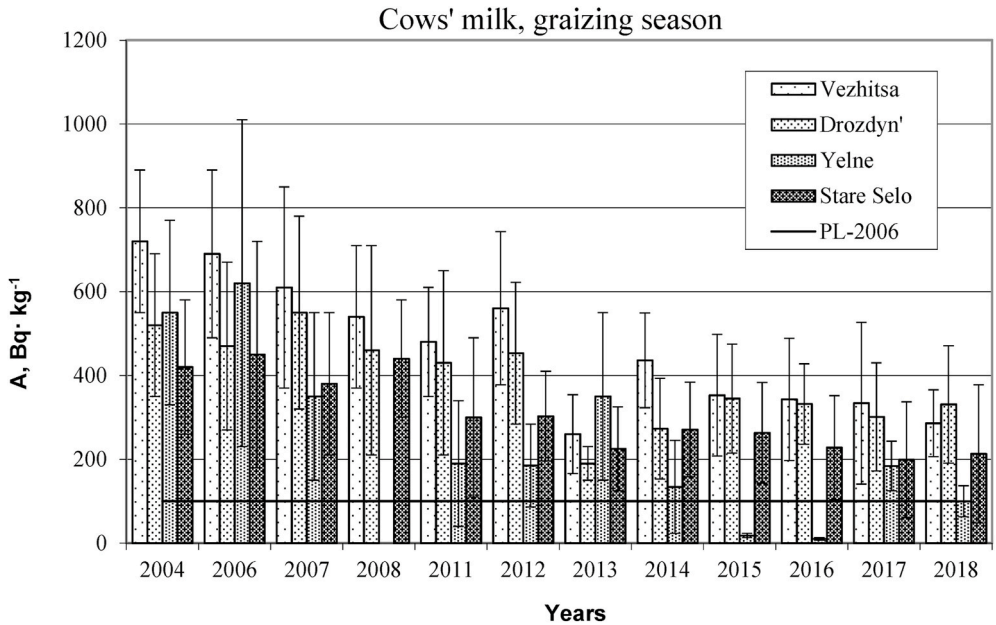

Fig. 2. Mean values (n > 10) of ^{137}Cs activity concentrations in cows' milk from private smallholdings in the "critical" settlements of the Rivne region (Ukraine). The error bars are standard deviations.

thoroughly mixed and the pooled sample was taken by the quartering method.[4] No loose concentrates (such as oil cake, etc.) were milled before sampling.

3.2.3. *Sampling of milk*

Samples of milk were taken from all species of dairy animal: cows, goats, sheep, horses, camels etc. Milk samples were taken in the places of production, sale and processing of dairy products or milk for dairy food production. For the prompt decision on the scale and spatial resolution of monitoring in case the levels of radioactive contamination were still not fully recognized, the monitoring of animal products started with points of the milk reception at the milk processing plants. This made possible quick identification of the areas with high animal products contamination and to make prompt decisions for protecting the public.

Sampling performed in the field was carried out together with measurements of the air dose rates in the various locations of the settlement, including the dairy farms. If GPS devices were available, the geographical coordinates of the sampling sites were determined and recorded in the sampling card attached to each sample.

All sampling teams had similar instructions with respect to sampling methods, methods of preservation, labelling and description of samples. Supplementary information that might be useful for further interpretation of the results (for example, information on precipitation during the passage of a radioactive cloud, conditions of livestock maintenance (pasture or indoor, feed used, date of grazing, duration of grazing and place of grazing dairy cattle, etc.) was also recorded as a comment in the sampling card. Sampling of dairy products was conducted in all populated areas, where the measured external dose rates were statistically significantly higher than that of the natural radiation background.

The sampling protocol used at that time, requested to select 1–3 individual samples at every collective milk farm, considering that one sample should be representative of 500 L of milk. Milk sampling from the households, with one or two cows per household, was carried out by taking a mixed (pooled) sample at the settlement milk collection centres during radiological monitoring in Russia. It was required to take a mixed sample from each individual herd if there were several herds feeding on different pastures. Between 3 and 5 milk samples were taken from the farming households at different locations of the settlement, if selection of mixed samples was not technically feasible or estimation of uncertainty of the mean values of the radionuclide concentrations in milk in a settlement was required.

Milk sampling was performed after mixing it in a collecting container, since radionuclide concentration in the cream differs from that in milk. The volume of the milk sample was dependent on the minimum detectable activity (MDA) of the measuring equipment. As a rule, 0.1–0.5 L was enough to determine the activity of radionuclides in milk with activity concentrations corresponding to the permissible level for radionuclides in food products.

The samples were placed in the refrigerator - if the samples were going to be sent to the laboratory on the same day. Otherwise, a special preservative, preventing milk decomposition, was added to the milk sample for better storage. Samples were labelled and registered, indicating the date and time of sampling, the date and time of milking, the method for keeping cows (indoor or outdoor), the amount and type of preservative used etc.

The express methods such as dose rate measurements on the surface of the milk tank were widely used for preliminary (rough) estimation of milk contamination during sorting and banning of milk.

To prevent use of dairy products with high activity concentrations of ^{137}Cs, the intensive monitoring program described earlier was implemented in the collective farms of the affected regions. Some summary results of the milk monitoring programs implemented in 1986–2006 are given in Fig. 1. High quantities of milk were banned in 1986–1990, when the amount of milk with ^{137}Cs concentrations above the action level reached several hundred thousand tons each year. However, due to the effective implementation of remedial actions, the amount of milk with high ^{137}Cs concentrations decreased rather quickly. At the same time, production of the milk with ^{137}Cs concentrations above TPLs is observed in some private farms. Exceptionally high levels of ^{137}Cs in animal products were found in most contaminated areas of the Rovno regions with a high share of wet peat soils known as soils of highest availability of radiocaesium for plants (Fig. 2).

During the period from 2006 to 2014, 99.6% of the total amount of milk produced in Belarus had 137Cs activity concentrations lower than

[4] Reduction in size into quarters of a crushed or powdered sample. Two opposite quarters are combined, while the other two quarters are discarded. The process is repeated as many times as necessary to obtain the quantity desired for some final use (e.g. as the laboratory sample or as the test sample).

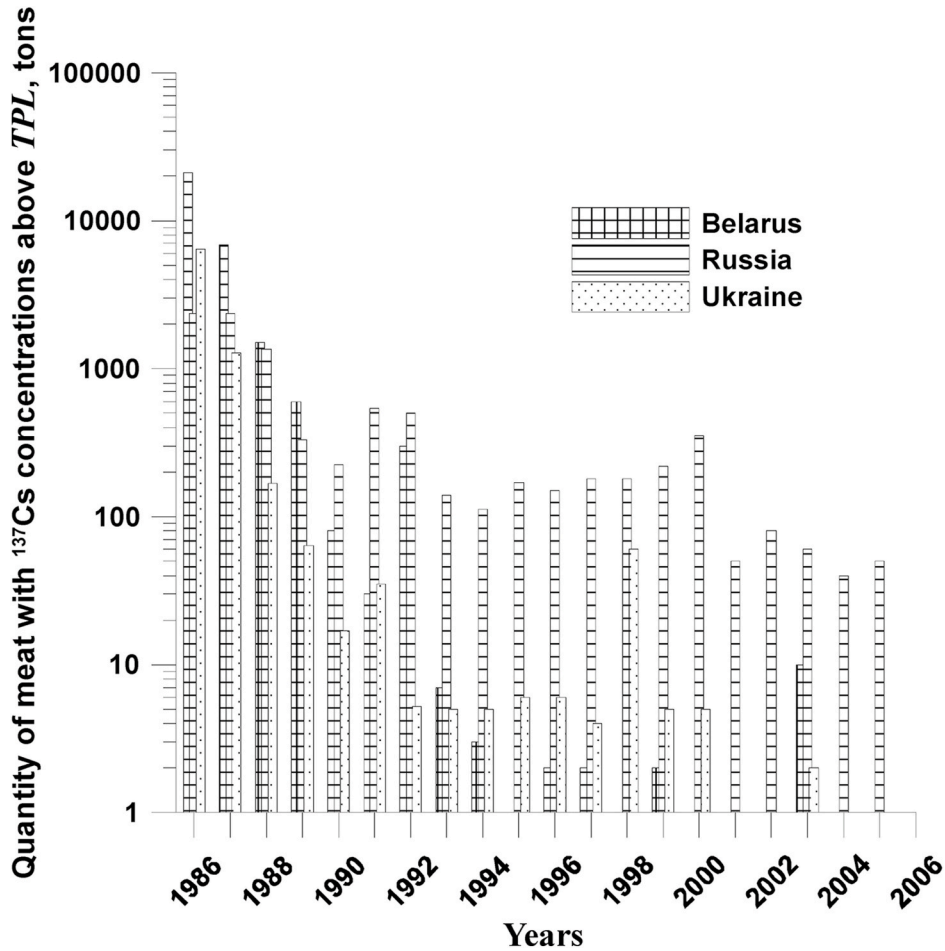

Fig. 3. Dynamics of meat quantities with ^{137}Cs activity concentrations above TPLs, produced in Belarus, Russia and Ukraine during first 20 years after the Chernobyl accident (Fesenko et al., 2007a).

37 Bq L-1, meeting the safety radiation standard for baby food (Tsybulka, 2015). In the Russian Federation, there are still 11 collective farms and more than 100 settlements where activity concentrations of ^{137}Cs in milk are above the permissible level of 100 Bq L^{-1}. In Ukraine, only 6% of milk measured annually in Ukraine in 2010–2017 had the concentration of ^{137}Cs exceeding the PL for the product (Gusak 2015). High ^{137}Cs concentrations in milk were occasionally observed in 20–30 settlements of Ukraine, mainly in the Rivne region, where wet peat soils are in wide use (Labunska et al., 2018; UIAR, 2018). At present, milk monitoring includes more than 80,000 measurements in Belarus, 10,000 in Russia (only the Bryansk region) and around 60,000 measurements in Ukraine.

Although the quantities of milk with ^{137}Cs concentrations above the PL are at the level of 3–5% of the total currently produced milk in the contaminated territory of Ukraine, high milk contamination remains one of the main reasons preventing the return of the affected regions to normal life conditions (Labunska et al., 2018; Kashparov, 2019).

3.2.4. Sampling of meat

Sampling of meat for radiological inspection was normally performed at the time of the slaughtering of animals at the butcheries. Around 10% of carcasses (half carcasses) of cattle, 5% of carcasses of sheep and pigs, 2% of frozen or chilled blocks of meat (beef, veal, pork, mutton) and offal (liver, lung, kidney etc.) were taken from each homogeneous lot of meat for further sampling.

Individual samples of meat with a weight of 0.2 kg were taken from muscles near the fourth and fifth cervical vertebrae, scapula, pelvic and femoral parts of the animal carcass. Such an approach has allowed for the avoidance of differences in ^{137}Cs concentrations in animal tissues because of variations of the fat content in different parts of the animal. Samples of offal were taken from the packaging units of the lot with mass of samples of 0.1 kg or larger. Samples of poultry meat were taken from whole carcasses. One piece of meat was taken from each frozen block of meat. Individual samples of meat and by-products were being ground, mixed up and the mixture was used to form a mixed sample. A pooled sample of poultry was formed by selecting from each carcass the samples of muscles from breast without skin. Pieces of meat are crushed (with a knife - for 10–15 g or passing meat through a meat grinder) and placed in a sample container. The resulting mass was thoroughly mixed. The quartering method was used to form an average sample weighing not less than 0.5 kg from the pooled sample.

The number of samples taken from the similar level of radioactive contamination (estimated by origin or measurements of the radiation dose rate) lot of animal products was dependent on its size (volume, mass). Thus, from the lot up to 0.5 tonnes only one sample was taken. Two samples were taken from the lot weighing 0.5–3 tonnes; three samples were taken from the lot up to 5 tonnes; five samples were taken from the lot weighing up to 10 tonnes; eight samples were taken from the lot weighing up to 15 tonnes and ten samples were taken from the lot weighing up to 20 tonnes. The requirements for the samplings were regulated by the quality standards adopted in the Soviet Union. These requirements for sampling of animal products were based on the anticipated products volumes, capacities of the monitoring laboratories and requirements to the precisions of radiation control adopted at that time (MG 13-7-2/1056, 1996; SOU, 2006a).

The changes in meat over time with radiocaesium concentrations

exceeding TPLs can be seen in Fig. 3. However, it must be realised that the TPL values have also reduced with time in each of the three countries so the data for individual years are not directly comparable. Differences in the time trends in Fig. 3 amongst the countries largely relate to the scale of the application of the countermeasures, but also to changes in the TPLs (see Table 2). In Russia and Ukraine, the current PLs (suggested for non-accidental exposure situation) of ^{137}Cs in meat is 200 Bq kg^{-1}, whereas in Belarus these levels are 500 Bq kg^{-1} in beef and mutton and 180 Bq kg^{-1} in pork and poultry.

The total quantities of meat with radiocaesium activity concentrations exceeding the TPL were much lower than those observed in milk. These differences reflected the effect of live monitoring of the animals, preventing slaughtering animals with high ^{137}Cs activity concentrations in their muscles. Live monitoring of livestock was applied extensively after the Chernobyl accident from 1988 to 1989 to measure in situ activity concentrations of $^{134/137}$Cs in muscle tissue. These measurements were performed largely before slaughtering to confirm that action levels are not exceeded. If the activity concentration is above the TPLs, management options - such as decontamination by clean feeding – or administration of Cs binders which reduce its absorption in the gut - were used to lower the activity concentration before slaughter. Clean feeding was used extensively after the Chernobyl accident in the former USSR, Norway and Sweden (IAEA, 2012).

In Belarus, the quantities of meat with activity concentrations with ^{137}Cs concentration exceeding the permissible level decreased from 360 tonnes in 1999 to 0.1 tonnes in 2010. This was achieved through rigorous animal monitoring at the butcheries and return for clean feeding animals with high activity concentrations of ^{137}Cs. The incidents with excess of the permissible levels in meat have not been observed since 2011. Incidents of returning animals from slaughterhouses to farms for clean feeding have not been recorded in Belarus in recent years either (Nikolaenko, 2019).

In Russia and Ukraine, the incidents of exceeding the ^{137}Cs PL in meat are observed very rarely (Gusak 2015). After 2014, samples of beef and pork with activity concentrations of ^{137}Cs exceeding the PL were not recorded, whereas in Russia only in 3% of measured meat samples ^{137}Cs concentrations exceeded the permissible level.

Identification of the requirements to the meat monitoring is a more challenging task than that of milk. Milk samples can be taken relatively easy, although the concentrations of ^{137}Cs in milk are quite variable and can be changed relatively fast. Samples of meat can be taken only after slaughtering and is a much more labour-intensive procedure compared to milk. At the same time, it is known that concentrations of radiocaesium in muscles of ruminants is 2-4-fold higher than those in milk for the same animals and similar chronic intake (Tables 1 and 3). There are also rather stable ratios between ^{137}Cs concentrations in milk and meat averaged across some large areas such as group of the settlements (district) or region. This information, together with the data on milk contamination, can be used for identification of areas where rigorous monitoring of meat is of special importance.

3.2.5. Sampling of eggs

Sampling of eggs for radionuclides was performed at the poultry farms from standard packages (boxes) containing 360 eggs (12 trays of 30 eggs). The number of the sampled eggs was dependent on the size the investigated lot and varied from twenty eggs (if there were less than 10 packages in the lot) to 150 eggs, if in the lot there were more than 500 packages with the eggs (Table 4). The eggs and packages for further examinations were taken from the lot random.

3.3. Measurements of radionuclides activity

3.3.1. In vivo measurements

In vivo (or live monitoring) monitoring for radiocaesium in animals was widely used in different countries after the Chernobyl accident. The live monitoring of animals was a rapid, accurate and simple method that

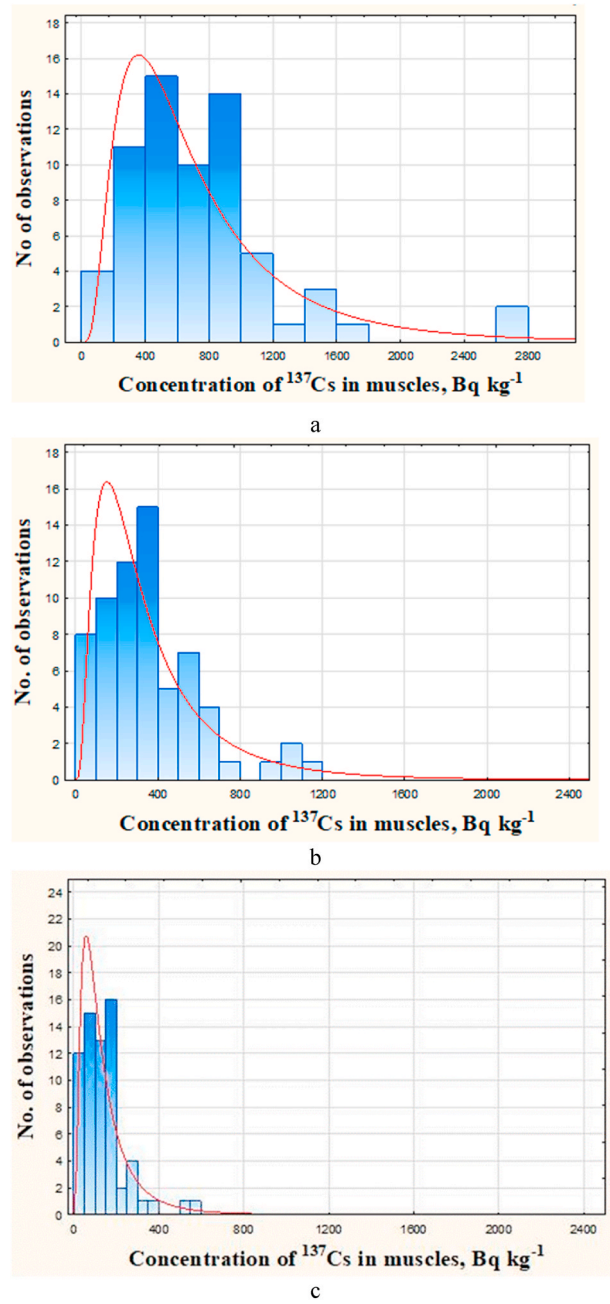

Fig. 4. Assessment of the effects of remediation used in the Veprin settlement of the Russian Federation. a - the frequency distribution of ^{137}Cs activity concentration in muscles of cattle before the remediation; b - after radical improvement of the fodder lands and c - during application of the hexacyanoferrate compounds (Cs binders).

enabled slaughtering animals only with radiocaesium concentrations in the muscles that could be acceptable for human consumption. A handheld gamma-spectrometer with collimated sodium iodide scintillation detector was often used for the measurements. The gamma-spectrometer was calibrated for measurements using a relationship between the count rate in the energy intervals for ^{134}Cs and ^{137}Cs after the in vivo measurements and the radionuclide activity of the muscle tissue of the same animal after slaughtering. Minimum detected activity concentration in the muscles of animals varied from 40 to 200 Bq kg^{-1} depending on background radiation and the detector efficiency. The method was well documented and used in the former Soviet Union countries (Fesenko et al., 2006; IAEA, 2006; SOU, 2006b) as well as in Norway (Brynildsen

and Strand, 1994).

Live monitoring data were also used for assessment of the remediation effectiveness applied after the Chernobyl accident. One example of use of the live monitoring of ^{137}Cs in animals for assessment of effect of remediation (application of Cs-binders for animals and radical improvement) in one of the settlements of the Bryansk region (Veprin, the Russian Federation) is given in Fig. 4. Hexacyanoferate compounds (Cs binders) with the administration rate of 6 g were administered in Veprin to the private cows, and additionally private households were provided with the cultivated pastures for grazing the cows. These two remedial options, most popular in the affected areas of Russia, were implemented in that settlement in 2005–2006 (Fig. 4).

The geometric mean of ^{137}Cs activity concentrations in the muscles of animals (590 Bq kg^{-1}) measured before implementation of the remedial actions were much higher than the permissible level in meat (160 Bq kg^{-1}), and 94% of animals were not suitable for slaughtering (Fig. 4a). Around two-fold reduction was observed upon remediation of the fodder lands and provision of the cultivated lands for grazing animals (Fig. 4b). Despite this decline the number of animals suitable for slaughtering has decreased up to 29%. Only additional application of hexacyanoferrate compounds allowed achievement of the reduction of the geometric mean of ^{137}Cs activity concentration in the muscles of animals up to 110 Bq kg^{-1}, and the fraction of the cattle that were not suitable for slaughtering was decreased up to 12% (Fig. 4c).

3.3.2. Measurements animal sample for radioactivity

The experience of the large radiation accidents, such as the Chernobyl and the Fukushima Daiichi accidents, demonstrates that even countries with well-developed nuclear power are not technologically prepared to mitigate the consequences of serious nuclear accidents, especially at an early stage of the emergency (IAEA, 2006; IAEA, 2014). Therefore, at the early stages after the accident, all possible options should be used for the evaluation of food contamination, even those where some of the instruments are not directly intended to measure radioactivity in food and the agricultural environment. A simple measurement method based on the use of simple widely available gamma radiometer/dosimeters with 25 × 40 mm NaI (Tl) scintillation detector (e.g. SRP-68-01, Electron, Ukraine) was suggested following the Chernobyl accident for the rapid sorting and banning of milk in large tanks or containers. The dose rate measurements were made on the surface of the container (canister, tank) or by placing the sealed detector into the container with liquid: milk or water. These dosimeters were calibrated with similar containers with known activity concentrations of radiocaesium measured by the spectrometric method (Prister et al., 2016).

Based on such measurements, the relationship between the dose rate values and radiocaesium concentrations in milk was identified together with the threshold value of the dose rate that ensures compliance with the milk TPL (see Table 2). Such dose rate levels were documented as reference levels used for the milk inspection for radionuclides with the aid of the dosimeters-radiometers. It was found that in large tanks with a volume above 100 L the increment of the external dose rate for 0.5 μSv h^{-1} above background comply with the milk activity concentration of ^{131}I about 350 Bq L^{-1}, ^{134}Cs–100 Bq L^{-1} and ^{137}Cs–250 Bq L^{-1} (Eckerman and Ryman, 1993).

Standard methods of gamma spectrometry for ^{60}Co, ^{106}Ru, ^{110m}Ag, ^{131}I, ^{134}Cs, ^{137}Cs and ^{241}Am, beta radiometry for ^{89,90}Sr, and alpha spectrometry for $^{238\text{-}240}$Pu and ^{241}Am were also widely used for determination of radionuclide concentrations in the animal samples (Prister et al., 2016).

Gamma spectrometry is the simplest and fastest method of analysis. This method is non-destructive and allows simultaneous determination in the sample of many gamma-emitting radionuclides. Gamma spectrometers with coaxial semiconductor detectors of high pure germanium (HPGe) or scintillation detectors from various materials (NaI (Tl), CsI, $LaBr_3$, etc.) were normally used to measure radionuclide activity concentrations in animal samples. HPGe detectors have better energy resolution compared to a scintillation type of detector and can be applied to detect a complex mixture of radionuclides. At the same time, together with a similar registration efficiency of gamma radiation, HPGe is still more expensive than spectrometers with scintillation detectors.

Despite that, both types of detector can be used to measure samples of animal products. However, at the early phase of a nuclear emergency when a mixture of short-lived radionuclides in the deposition should be measured, use of the gamma-ray spectrometer with HPGe detectors to control radioactive contamination of forage is preferable.

Application of HPGe detectors for separation of ^{110m}Ag and ^{137}Cs concentrations in the liver of the cattle evacuated from the Chernobyl 30-km zone is a good example of a need to use the detectors with high-energy resolution for the monitoring of animals. The lines of gamma-spectrum 658 keV and 662 keV are used for detecting ^{110m}Ag and ^{137}Cs in environmental samples. These gamma lines are close to one to another. Therefore, a separation of these gamma-radiation lines, required to detect both ^{110m}Ag and ^{137}Cs separately, is not possible using scintillation detectors and only the application of the high energy resolution HPGe detectors allowed a measurement of specific activity of these radionuclides in the animal tissues. Thus, these measured data show that ^{110m}Ag concentrations in the liver of cattle was higher than those of radiocaesium although ^{110m}Ag concentrations in the environment were much lower compared to those of ^{137}Cs (V. Kashparov, personal communication, July 22, 2018). It is worth mentioning that this effect was specific only for the liver, since for the other animal tissues the ^{137}Cs concentrations were higher than those for ^{110m}Ag. In turn, this allowed for getting a proper dose estimate and explanation of the related biological effects in cows in the 30 km zone.

Radionuclides that do not emit gamma radiation or have very weak gamma lines (e.g., ^{89,90}Sr, $^{238\text{-}240}$Pu) cannot be identified directly by gamma spectroscopy. In most cases, they must be analysed after extraction from a sample matrix by means of radiochemistry (Vajda and Kim, 2012; Brun et al., 2002; Maxwell et al., 2011). Such methods are labour consuming and require at least one day to analyse a set of samples. However, if a ratio can be established between a gamma emitting nuclide and a non-gamma emitter, the radioactivity of the latter can be estimated indirectly by gamma spectrometry. In the late phases of the emergency, a non-destructive, rapid and simple beta-spectrometric method (Courti et al., 2002) was also applied to determine ^{137}Cs and ^{90}Sr activity concentration in animal products. The method has been widely used since the 1990s to measure ^{90}Sr in agricultural products and foodstuffs in the former Soviet Union countries affected by the Chernobyl accident.

3.4. Optimizing sampling program

Information on the source of radioactive contamination gives an indication of the radionuclides that are expected to be in the food samples. Such information is useful for planning the emergency sampling, because a request for a complete analysis for all radionuclides in each sample can lead to long and expensive sample processing and measurement.

Food monitoring following a nuclear emergency should provide information on radioactive contamination of food with some predefined degree of the precision. The uncertainty of data on radionuclide activity concentrations in individual samples, can be used for estimating the number of samples to be taken for provision of the required degree of the assessment's precision. To illustrate such an approach, the data on ^{137}Cs concentrations in the cow milk produced in the private households of the Ukrainian settlements were used to estimate the number of samples needed to determine the average radiocaesium concentration in the milk with the required precision (UIAR, 2018).

It was found that the differences in radionuclide activity concentrations in the cow's milk samples from one herd taken from one sampling area for one day could differ by more than 10 fold (Fig. 5) (Khomutinin et al., 2001; UIAR, 2018). A statistical analysis has shown

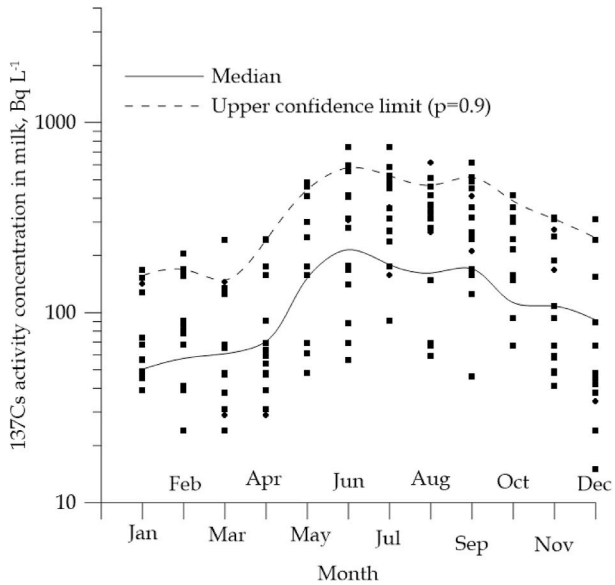

Fig. 5. Annual dynamic of ^{137}Cs concentrations in milk in Myliach settlement, Rivne region, Ukraine, 1997.

$$Me_{A_{RN}} = \exp(\mu_{RN}) \tag{5}$$

The parameter s_{RN} is the approximation for the coefficient of variation $W_{A_{RN}}$ of the radionuclide activity concentrations in the product:

$$s_{RN} = \sqrt{Ln\left(\left(\frac{SD_{A_{RN}}}{\bar{A}_{RN}}\right)^2 + 1\right)} \cong W_{A_{RN}} \quad GSD = \exp(s_{RN}) \tag{6}$$

The analysis of ^{137}Cs concentrations variability in milk samples, which were collected in 18 villages within contaminated areas of Ukraine (the number of milk samples was 1700), has not revealed the characteristic peculiarities connected with the sampling site disposition, and deviations between sites had a random character (Khomutinin et al., 2001). The high variability of ^{137}Cs activity concentrations in the milk of cows was due to both the individual animals' characteristics (daily milk yield, fat content in the milk, diet, etc.) and the variations in ^{137}Cs concentrations in the feeds. Therefore, we consider the s_{RN} as a random variable, which follows a normal probability distribution. It was also found that the dispersion of the logarithms of the activity ^{137}Cs concentrations in milk taken at the various sites with different properties (contamination density, soil properties ets.) were not statistically different ($p > 0.05$). Therefore, we suppose that the dispersion of the logarithms of the activity ^{137}Cs concentrations in milk does not does

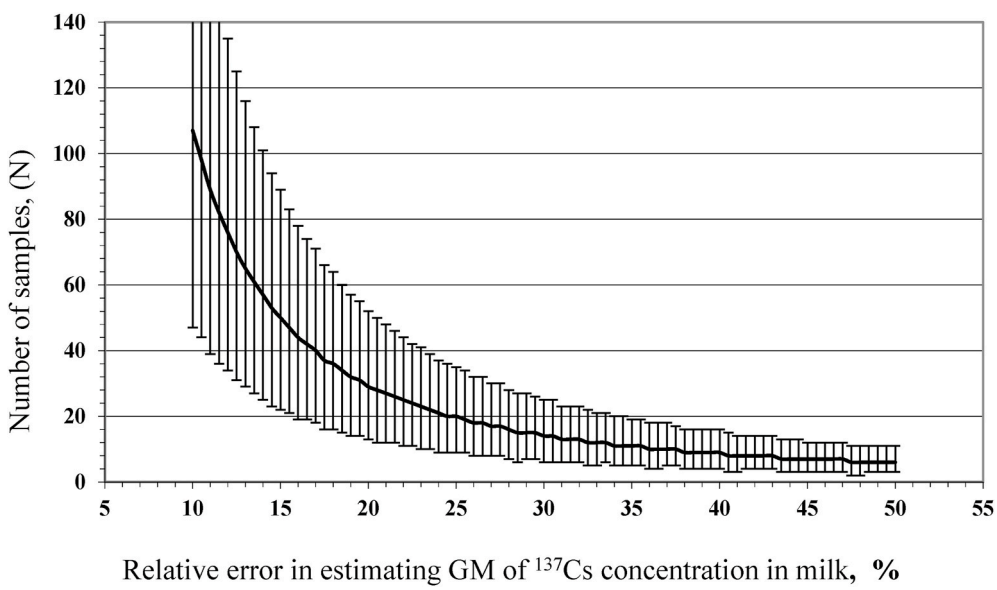

Fig. 6. The nomogram for estimating minimum number of samples (n) required to evaluate the geometric mean (GM) of ^{137}Cs activity concentration in cow milk for a given relative error ($p = 0.95$) in the definite moment of grazing period when measurement error is less than 15% ($p = 0.95$).

that the distribution of ^{137}Cs activity concentration in these samples may be satisfactorily fitted by the log-normal distributions (Khomutinin et al., 2001; ICRU, 2006.)

$$f\left(A_{RNso}\right) = \frac{1}{\sqrt{2\pi}\cdot A_{RN}\cdot s_{RN}} \exp\left(-\frac{1}{2}\left(\frac{\ln(A_{RN}) - \mu_{RN})}{s_{RN}}\right)^2\right. \tag{4}$$

where: A_{RN} is the radionuclide activity concentration in milk; μ_{RN} is the average logarithm[5] of the radionuclide activity concentrations in the product; s_{RN} is the standard deviation of the logarithms of the radionuclide activity concentration in the product. Therefore, the geometric mean of radionuclide activity concentration in the milk is equal to:

depend on the local settlement specific factors and the variability of the data for each site can be described only by the value of standard deviation of the logarithms of ^{137}Cs activity concentrations in milk (s_{Cs}) of 0.6 ± 0.2, if a relative measurement error of ^{137}Cs activity in samples was less than 15% ($p = 0.95$).

Data on variability of the activity ^{137}Cs concentrations in milk obtained in Ukraine were used to assess the number of samples to be taken required to fit to the acceptable uncertainty level (Khomutinin et al., 2001). Using the interval estimate (see Khomutinin et al., 2020), a two-dimensional diagram was designed (Fig. 6). The nomogram shown in Fig. 6 allows for estimating the minimum number of samples (N) taken from the herd of cows, which is required to estimate the geometric mean of the radionuclide activity concentration in milk with a relative error ($p = 0.95$).

For example, let us suppose that required uncertainty of estimating a geometric mean of milk is 30% and the relative error of ^{137}Cs in samples

[5] The natural logarithm of x is generally written as log x, $\log_e$ x, ln x.

is less than 15%. The conclusion can be made based on data from Fig. 6 that fourteen independent milk samples (N = 14) should be taken from the herd of cows to comply with the above conditions (on average). Conversely, the error of the resulting average milk contamination in the herd would be more than 50%, if five samples (N = 5) of milk were taken from five cows of one herd and from five samples one composite sample was prepared to measure ^{137}Cs activity (Fig. 6). The results obtained show that the relative error in determination of the annual internal doses to the population of the settlement (Likhtarov et al. 1996, 2012), may exceed 50% if only ten samples of milk were taken per year - five samples when the cows were housed indoors and five samples during outdoor grazing on pasture (Khomutinin et al., 2001).

4. Conclusion

The paper gives an overview of 35 years of experience of monitoring of animal products after the Chernobyl accident. At the early stages after the accident, application of simplified in situ techniques was a highly effective tool for assessment of radionuclide concentrations in animal products, even though these instruments were not originally developed to measure radioactivity in food. Overall, in-situ measurements tools calibrated based on data on radionuclide activity concentrations in animal products were one of the key elements for monitoring of animal products at all time periods after the accident. Information on properties of radionuclides of concern are important to identify areas where radionuclide activity concentrations in animal products can result in a need in use of monitoring programmes. Mobility of radionuclides in the environment, parameters of radionuclide transfer to animals and biological half-lives in the animal tissues can be listed among such data. The analysis of the data on milk sampling in 18 settlements affected after the Chernobyl accident shown that radionuclide concentration in the animal products can be fitted by the log-normal distributions and the dispersion of the logarithms of the activity ^{137}Cs concentrations in milk is not dependent on the local settlement specific factors. Based on these findings the approach for determination of number of samples to be taken to provide the required precision of the GM estimate for given variability of the data is suggested.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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