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Current radiological situation in areas of Ukraine contaminated by the Chernobyl accident: Part 1. Human dietary exposure to Caesium-137 and possible mitigation measures

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ABSTRACT

This study reports for the first time temporal trends for the period of 2011–2016 in ^{137}Cs content in cow's milk originating from private farms and households of 14 settlements located in the territories of the Rivne region, Ukraine. These areas are still radioactively contaminated as a result of the accident at the Chernobyl Nuclear Power Plant (ChNPP) in 1986. In 2016, the average ^{137}Cs activity concentration in milk exceeded the Ukrainian Permissible Level (PL) for adults of 100 Bq/l in samples from 6 settlements and the PL for children of 40 Bq/l in 8 settlements, reaching activity concentration of around 500 Bq/l in some samples. Estimated annual effective doses calculated utilizing two different methodologies were in the range of 1.4–2.6 mSv/year and 1.2–1.8 mSv/year, respectively. The estimated effective period of milk semi-purification ($T_{1/2\text{–}eff}$) from ^{137}Cs in these settlements was in the range from 8 to 17 years. The estimated ecological period of milk semi-purification ($T_{1/2\text{–}eco}$) from ^{137}Cs was in the range from 11 to 36 years. The optimization of the remedial actions strategy for investigated settlements exposed to an effective dose above 1 mSv/year (as estimated in 2016) has shown that a diversity of measures can decrease effective dose for a representative person to below 1 mSv/year. Such measures include application of Ferrocyn to cows, mineral fertilization of potato fields, information campaigns on consumption of wild mushrooms and other forestry products, and feeding pigs with uncontaminated fodder. The total costs of such measures are estimated to be about 71,000 Euro per year for the combined population (8336 inhabitants) of the six villages investigated in this study that showed the highest median residual ^{137}Cs activity concentrations in milk, with a subsequent decrease in cost in the future. This would result in an averted collective dose of 11 man-Sv, at an average cost of 6.5 kEuro/man-Sv averted. In the absence of governmental programs for implementation of necessary protective measures to reduce radiological risks to impacted populations, the exceedance of PL for the activity concentration of ^{137}Cs in cow's milk for adults of 100 Bq/l in the Chernobyl-affected areas of Ukraine could persist for many more years – until at least 2040.

1. Introduction

As a result of the accident at the Chernobyl Nuclear Power Plant (ChNPP) in 1986, over 40,000 km^2 of the territory of Ukraine were contaminated with ^{137}Cs at deposition density above 40 kBq/m^2, including some 10,500 km^2 of agricultural land (Fesenko et al., 2007a; IAEA, 2006; Kholosha, 2008; Nadtochiy, 2003). One month after the accident, in June 1986, ^{134}Cs and ^{137}Cs were the main contaminants deposited within residential areas outside of Zones 1 and 2, the Chernobyl Exclusion Zone and the Zone of Unconditional (Obligatory) Resettlement, respectively. Thirty years later, both external and internal human exposure to Chernobyl-derived radionuclides are mainly due to ^{137}Cs.

We have previously shown (Kashparov et al., 2005) that milk contributes on average more than half to the ^{137}Cs dietary intake for the residents of rural Chernobyl-contaminated areas of the Rivne region, a consequence of the abnormally high bioavailability of ^{137}Cs from the peat/boggy soils (Maloshtan et al., 2015), while potatoes account

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Fig. 1. Location of Rivne region on the map of Ukraine (dark blue area). ChNPP – Chernobyl Nuclear Power Plant. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

for < 10%. However, in some forest-based areas, wild berries and mushrooms may contribute up to 50% of the dietary ^{137}Cs intake during specific years (Kashparov et al., 2005).

Although the high ^{137}Cs bioavailability of local soils was known before the Chernobyl accident (Marey et al., 1974), little attention was paid to this problem in the first years after the accident because of the comparatively low density of contamination by ^{137}Cs (< 100 kBq/m^2) measured at these distances (about 200 km) from ChNPP. High activity concentrations of ^{134,137}Cs in milk (exceeding the Ukrainian Temporary Permissible Level (TPL-89) of 370 Bq/l) were nevertheless reported for settlements located within Rivne and Volyn' regions through the work of the research expeditions of the Ukrainian Institute of Agricultural Radiology in 1989–90 (Nadtochiy, 2003).

Due to the natural decay of ^{137}Cs, its activity in soils has decreased twofold from values recorded immediately after the accident, while in foodstuffs, activities have decreased by factors between ten and a hundred, partly as a result of immobilisation by the soil matrix and partly through various remedial actions and countermeasures taken (Fesenko et al., 2007a; IAEA, 2006; UN, 2011). The number of settlements with annual effective dose to the population above the established limit of 1 mSv/year (Supreme Council of Ukraine, 1991a) fell from 826 in the early 1990s to only 25 settlements of the Rivne and Zhytomyr regions by 2011–12, decreasing by a factor of 7 between 2004 and 2012 alone (Bazyka et al., 2016). Nonetheless, in these remaining settlements, internal exposure is still elevated, especially by the consumption of local milk with ^{137}Cs activity concentration above the Permissible Limits (PL) for adults of 100 Bq·l^{-1} (see Fig. S2b in SI) (Lihtarov et al., 2013), and well above the PL for children (40 Bq/l). Children born decades after the Chernobyl accident are therefore still exposed to significant doses of harmful radionuclides in these settlements. Efforts to minimise and prevent further exposure of populations residing in territories of Ukraine affected by the Chernobyl accident through the consumption of radioactively contaminated food are therefore still urgently needed.

Government monitoring of Chernobyl affected settlements in Ukraine, including the measurements of ^{137}Cs in cow's milk, ceased in 2012. This has created a gap in the systematic knowledge and consequent difficulties to assess the current radiological situation in these communities and, therefore, to implement effective response measures. A number of relatively simple remedial actions could otherwise be applied in a targeted fashion to limit exposures (IAEA, 2006; UN, 1997), including the application of hexaferrocyanide compounds such as Prussian Blue or Ferrocyn, to cow fodder. Ferrocyn application is one of the remedial actions specifically recommended for territories which are characterised by high soil-plant transfer of ^{137}Cs and yet where other countermeasures (e.g., top soil removal, drainage of wet peat soil) are ineffective or not possible to conduct (Ulanovsky et al., 2011). It has been used successfully in Norway and Sweden to reduce the activity concentration of radiocaesium in lamb and venison by up to 5 times, and in cows and goats milk by 3–5 times, respectively (IAEA, 2006).

A study based on monitoring of soil and agricultural products for radiological contamination in Ukraine in 2004 (Jacob et al., 2009) evaluated various scenarios for implementation of remedial measures and agricultural practice in radioactively contaminated areas, and identified a high potential to reduce radiological, economic and social consequences in affected areas through some relatively simple interventions. A more recent study (Waddington et al., 2017) also reported high cost-effectiveness of such remedial measures, though based largely on the same data set and hence not reflecting the current situation in Ukraine.

Therefore, the aims of the current study were: a) to fill gaps in the data concerning the current situation in relation to human exposure to ^{137}Cs via consumption of locally produced foods in areas of Ukraine that were radioactively contaminated as a result of the Chernobyl accident; b) to investigate temporal trends of ^{137}Cs activity concentrations in cow's milk originating from settlements in Ukraine in which the last governmental monitoring (in 2012) reported exceedance of ^{137}Cs permissible levels in milk; c) to forecast ^{137}Cs activity concentrations in milk for these settlements in the future and; d) to evaluate the scope of the countermeasures that are still necessary more than 30 years after the Chernobyl accident, in order to reduce or prevent further exposure of Ukrainian populations to radioactive elements.

2. Materials and methods

2.1. Monitoring network

The study area included fourteen settlements in three districts (Rokytne, Sarny and Dubrovytsya districts) of the Rivne region (area: 20,047 km^2, population: about 1.2 million) (see Fig. 1 and Fig. 2) that

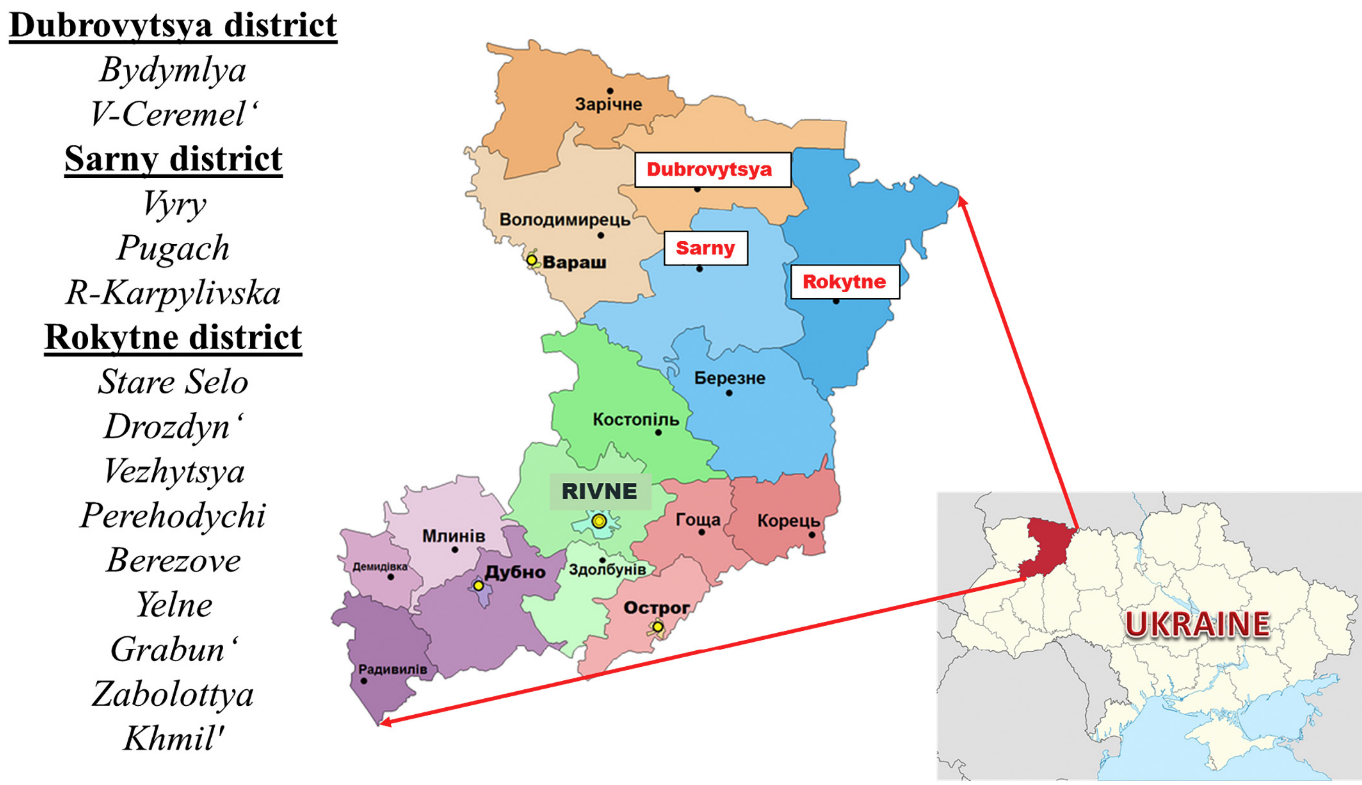

Fig. 2. Map of the Rivne region and a list of monitored villages in Dubrovytsya, Sarny and Rokytne districts.

are located about 200 km to the west of ChNPP and have reported ground contamination by ^{137}Cs in the range of 17–83 kBq/m^2 (Lihtarov et al., 2012).

These settlements, according to the last government monitoring data obtained in 2011–2012, had the exceedance of both annual effective dose and average ^{137}Cs activity concentration in milk above their respective limits (1 mSv/year and 100 Bq/l, respectively) (Lihtarov et al., 2012, 2013).

Table 1 describes these settlements, including their location, population, number of milking cows in each settlement in 2016, and the data for ground contamination calculated for 2016 based on previous research (Lihtarov et al., 2012), taking into account ongoing ^{137}Cs radioactive decay. The density of the contamination by ^{137}Cs within these settlements' boundaries was in the range from 17 kBq/m^2 to 83 kBq/m^2 (see Table 1). There were several hotspots detected outside of the settlements where ^{137}Cs contamination of about 200 kBq/m^2 was recorded. This, however, was not considered to influence substantially the human exposure to radioactive substances in the investigated area as these hotspots were not located within inhabited areas or in fields used for cultivation or pasture.

2.2. Sample collection

Between 2011 and 2016, three categories of local food products (milk, potatoes and wild mushrooms) were collected from 14 villages in the Rivne region (Fig. 2) that are located within the zone of guaranteed voluntary resettlement and which were identified by the most recent official data as having estimated effective internal does above 1 mSv/y (Lihtarov et al., 2012). One exception was the village of Bydymlya in Dubrovytsya district, which is located in the zone of unconditional (obligatory) resettlement, and yet which is still occupied and grazed. Foods were sampled opportunistically, based on availability of milk and

Table 1
Description of investigated settlements located in Rivne, Sarny, and Dubrovytsya districts of the Rivne region, Ukraine.

District	#	Settlement name	Coordinates		Distance to ChNPP, km	Estimated soil surface density of ^{137}Cs, kBq/m^2 (for 2016)	Number of	
			Latitude N°	Longitude E°			Villagers	Milking cows
Rokytne	1	Stare Selo	51.608	27.127	208	28	3507	905
	2	Drozdyn'	51.646	27.235	201	31	2186	573
	3	Vezhytsya	51.566	27.098	210	55	1097	382
	4	Perehodychi	51.619	27.005	217	77	509	148
	5	Berezove	51.580	27.335	194	35	3236	771
	6	Yelne	51.452	27.137	206	55	751	187
	7	Grabun'	51.553	27.245	200	40	333	86
	8	Zabolottya	51.600	27.294	197	36	510	120
	9	Khmil'	51.479	27.358	192	17	1175	237
Sarny	10	Vyry	51.243	26.951	220	39	2307	111
	11	Pugach	51.341	26.900	223	39	668	6
	12	R-Karpylivska	51.393	26.780	232	59	495	61
Dubrovytsya	13	Bydymlya	51.667	26.972	220	59	872	122
	14	V-Ceremel'	51.537	26.988	218	83	165	37

other produce from individual households, such that no selective bias was applied. In total, 977 samples of milk, 28 samples of potatoes, and 57 samples of mushrooms were collected for analysis of radioisotope content.

The samples of whole milk were collected in 250 ml pre-cleaned polyethylene bottles and placed immediately into a portable refrigerator for transportation. On arrival at the laboratory, milk samples were homogenized by thorough shaking and the exact volume and weight were recorded. Samples of dried mushrooms and fresh potato were collected in paper bags. Potatoes were peeled and thoroughly washed with tap water.

Before measurements of radioactivity were carried out, potato and mushroom samples were crushed and thoroughly homogenized using a blender (Braun, Germany). About 250 ml, 500 g and 100 g of milk, potato, and mushrooms, respectively, were used for ^{137}Cs activity concentration measurements.

2.3. ^{137}Cs activity concentration measurement

Activity concentration of ^{137}Cs in food samples was measured using a low-level gamma-spectrometer with a high-purity germanium detector (GEM-30185, EG&G Ortec, USA) equipped with a multichannel analyzer (*ASPEC-927*) and a passive protection device, and was operated using the *GammaVision* software (ORTEC, 2015). Polyethylene cylindrical containers (130 cm^3) and Marinelli beakers (500 cm^3) were utilized for measurements. Calibration of the spectrometer was conducted according to a previously published method (ASTM, 2014) using certified standards (water equivalent multinuclides standard, V. G. Khlopin Radium Institute, Russia). QA/QC procedures were frequently performed and included regular monitoring of the system performance, X-charts for activity/efficiency, background and full width at half maximum (FWHM) for the ^{137}Cs peak (662 keV). To validate accuracy and precision of the method employed for ^{137}Cs activity concentration measurement, quality control samples (i.e., different matrix samples including water, soil, and sawdust spiked with known certified activities of radionuclides) and Certified Reference Materials (CRM) have been analysed alongside of the samples. The analysis of CRM IAEA-375, "Radionuclides and trace elements in soil", (in triplicate) showed satisfactory results for ^{137}Cs mean activity concentration of 5320 ± 300 Bq/kg in comparison with the certified value (5280 Bq/kg) and was within the 95% confidence interval (5200–5360 Bq/kg) recommended for this material (IAEA, 2000). The limit of detection for ^{137}Cs in all samples was 1 Bq (per sample).

2.4. Estimation of effective doses

For comparison, we utilized two models for effective doses estimation:

1) the established model developed and applied by the Ukrainian government since the early 1990s, referred to officially as Method-96 (Lihtarov et al., 1996); and.

2) a more recent, specialised model, Remediation Strategies after the Chernobyl Accident (ReSCA) (Ulanovsky et al., 2011), based on data obtained during some two decades of agricultural countermeasures performed in all rural settlements (in Belarus, Russia and Ukraine) in which the dose to the "Representative Person" from the Chernobyl accident in 2004–2010 exceeded 1 mSv/year (IAEA, 2012; Nisbet et al., 2004; Fesenko et al., 2007b; Jacob et al., 2009). The "Representative Person" is an individual, who will almost always be a hypothetical construct, who receives a dose that is representative of the more highly exposed individuals in the population as defined in the reports by ICRP (ICRP, 2006, 2007) and IAEA (IAEA, 2011).

According to Method-96, we calculated the average annual effective dose due to external exposure for a given settlement based on the average density of contamination of its territory by ^{137}Cs (A_s^{Cs-137}, kBq/m^{-2}), taking into account factors of outdoor and indoor occupancy (i.e., the fraction of time spent indoors and outdoors) (Eckerman and Ryman, 1993; IAEA, 2011). Temporal variations of irradiation due to vertical re-distribution of radionuclides in soils have also been accounted for using this methodology. The average annual effective dose due to external exposure ($\overline{D_{ext}}$, mSv/year) was calculated for each village according to Eq. (1), taking into account the value of dose coefficient for ^{137}Cs exposure from contaminated soil $B_{ext}{}^{Cs-137}$ of 2.9·10^{-3} (mSv·y^{-1})·(kBq·m^{-2})$^{-1}$ reported for rural populations in Ukraine (Lihtarov et al., 1996):

$$\overline{D_{ext}} = B_{ext}^{Cs-137} \cdot A_s^{Cs-137} \quad (1)$$

The contribution of other Chernobyl-derived radionuclides to external dose outside of the ChNPP Exclusion Zone is small in comparison with ^{137}Cs→^{137m}Ba (e.g., < 0.7% for ^{90}Sr and < 0.07% for ^{241}Am) and, therefore, those have not been taken into account.

According to the Method-96, we estimated the internal annual effective dose utilizing a combination of dietary consumption rates obtained in the middle of the 1990s for the population residing in rural areas of Ukraine (Lihtarov et al., 1996) and annual data on ^{137}Cs activity concentrations in milk and potatoes from each of the settlements obtained in the current study in 2016.

The average annual effective dose due to internal exposure ($\overline{D_{\text{int}}}$, mSv/year) was calculated by Eq. (2):

$$\overline{D_{\text{int}}} = B_{ing}^{Cs-137} \cdot \sum_{i=1}^{n} (A_i^{Cs-137} \cdot M_i) \quad (2)$$

where $B_{ing}{}^{Cs-137}$ = 1.3·10^{-5} mSv/Bq is committed effective dose per unit ingestion intake of ^{137}Cs (IAEA, 2011);

$A_i{}^{Cs-137}$ is activity concentration of ^{137}Cs in *i* foodstuff (Bq/kg, fresh weight);

M_i is an average annual consumption rate of *i* foodstuff (kg/year).

For the ReSCA model, the external exposure of the Representative Person to radiation emitted from radionuclides deposited on the ground was estimated based on most recent ^{137}Cs contamination data for each settlement reported for 2012 (Lihtarov et al., 2013) and recalculated for 2016 taking into account further radioactive decay of ^{137}Cs, with a half-life of 30 years (Table 1). Estimates of the internal exposure due to radionuclide uptake via ingestion of foodstuffs were calculated by Eq. (2) and based on:

1) Estimates for consumption rates of the staple foods determined for Stare Selo village in Rivne region (see Table S1 in SI), based on our 2012 survey, recognising that, to the authors' knowledge, no more recent or more extensive published data are available.

2) Most recent data (2015–2016) on activity concentration of ^{137}Cs in three products (milk, potato and wild mushrooms) measured in the current study. These three products accounted for over 90% of the human ^{137}Cs intake in investigated areas (Kashparov et al., 2005) with milk as a main contributor (over 50%) in villages where ^{137}Cs activity concentrations in milk were > 70 Bq/l. In cases with ^{137}Cs activity concentrations in milk < 70 Bq/l, wild berries and mushrooms contributed greatly (up to 50%) to the human ^{137}Cs intake.

3) For settlements in which experimental data were not possible to obtain for some foodstuffs in the period 2015–2016, the activity concentrations of ^{137}Cs in these products were estimated using soil-specific ^{137}Cs aggregated transfer factors (Tfag) for wet peat/boggy Histosol soils (Stolbovoi, 2000) that predominate in the investigated areas. For potato, beef, pork and mushrooms, Tfag values of 1, 10, 4 and 20 Bq/kg fresh weight (fw) per kBq/m^2 respectively (Ulanovsky et al., 2011) have been used. Histosols in the investigated areas have not been changed or modified and are still characterised by high biological availability of radiocesium, as was recently confirmed (Maloshtan et al., 2015) for two villages in Rivne region (Vezhytsya and Yelne) that are also considered in our study. Hence, the use of the above-mentioned Tfag was considered as justified. For reference, the current Ukrainian PL for ^{137}Cs activity concentration for milk, meat, fresh wild mushrooms and berries, and dried wild mushrooms and berries are 100 Bq/l, 200 Bq/kg,

500 Bq/kg, and 2500 Bq/kg, respectively (MHPU, 2006). For both models considered in the current study, the total dose to the Representative Person (D_{total}), defined as the 95%-ile of the individual dose distribution in a settlement and expressed as settlement-average doses, $\overline{D_{ext}}$ and $\overline{D_{int}}$, have been calculated as in Eq. (3):

$$D_{total} = f_{ext} \cdot \overline{D_{ext}} + f_{int} \cdot \overline{D_{int}} \quad (3)$$

where $f_{ext} = 1.8$ and $f_{int} = 3.0$ are contemporary values of the factors defined for rural settlements in Ukraine (IAEA, 2007).

2.5. Estimation of the effectiveness of remedial actions

We evaluated seven remedial actions that have been specified previously as optimal for Ukraine (Ulanovsky et al., 2011) including:

1. **RI - R**adical **I**mprovement of grassland, which includes removing vegetation, ploughing, liming, fertilization, and reseeding;
2. **D - D**rainage (used only in connection with RI on wet peat soil);
3. **FA - F**errocyn **A**pplication to cows;
4. **FP** - **F**eeding **P**igs with uncontaminated fodder before slaughter;
5. **MF** - **M**ineral **F**ertilizer application on potato fields;
6. **IM** - **I**nformation campaign on **M**ushrooms and other forest produce consumption;
7. **RS** - **R**emoval of contaminated **S**oil from populated areas in a settlement.

The principal criterion for the success of remedial actions was the reduction of the annual effective dose (< 1 mSv y^{-1}) for the "Representative Person" of the population as defined in the reports by ICRP (ICRP, 2006, 2007) and IAEA (IAEA, 2011).

The quantitative characteristics we used with the ReSCA model to estimate the effectiveness of remedial actions in the study area were obtained from Ulanovsky et al. (2011). The cost of applying each remedial action in Ukraine and the projected reduction factors (i.e., the ratio between contamination of soil or a food product before and after remedial action implementation) (Ulanovsky et al., 2011) are presented in Table S2 and Table S3 in SI. For each village, the cost of an action was estimated using quantitative characteristics for each action multiplied by the number of inhabitants (for actions FP, MF, IM, and RS) or the number of cows (in case of actions RI and FA). The total cost of the remedial actions for an individual village was then derived as the sum of the costs of the actions proposed for that village.

Due to the impossibility of locating a parcel of land within the study area where cultivated pastures and hay-meadows could be newly created, the action D (drainage), which is used only in combination with the action RI (radical improvement), was not considered further in this study.

3. Results and discussion

3.1. ^{137}Cs activity concentration in milk in 2011–2016

Our data for ^{137}Cs in milk obtained for 2011 were in good agreement (Spearman's rank correlation coefficient r_s was 0.77 and the two-tailed value of P was 0.0014) with the governmental official data for the same year (Lihtarov et al., 2012), which confirms the representativeness of our experimental data (see Table 2). Data for ^{137}Cs activity concentration in milk during the period 2011–2016 (see Table 2) suggest little systematic change in the patterns of contamination over that period for most of the settlements of the Rivne region investigated in the current study. The milk from some of the settlements had activity concentration of ^{137}Cs below the Ukrainian PL in some of the years for which samples were taken. Some of the year to year variability apparent in the data may be due to fluctuations in weather conditions, and seasonal variation in the feed given to cows and its degree of contamination (e.g., fresh range forage and/or hay), as well as due to other anthropogenic factors (e.g., changes in land use structure). For example, in the village of Yelne, ^{137}Cs average activity concentration in milk decreased from 134 ± 111 Bq/l (measured in 2014) to 18 ± 5 and 10 ± 3 Bq/l (measured in 2015 and 2016, respectively). The most probable reason for such a decrease was the complete destruction of pasture near the village as a result of extensive operations of amber extraction in these areas, meaning that the cattle stock of Yelne village has decreased and remaining cows have been kept within the boundaries of each household on small patches of land or grazed on another pasture. Soils adjacent to houses in this village (i.e., Podzoluvisol) tend to be different from those in the pastures, and in particular are not characterised by a high soil-plant ^{137}Cs transfer coefficients (IAEA, 2006) due to the fact that villages in the area are historically built on dry, sandy soils while pastures are mostly characterised by wet peaty soils (Maloshtan et al., 2015). This could help explain why the average content of this radionuclide in cow's milk sampled in 2015 and 2016 was about 10 fold lower in comparison to 2014. The other villages that did not have exceedance of the Ukrainian PL for ^{137}Cs in milk (for average values) during the last 3 years of the study period included Grabun', Zabolottya, Khmil', Vyry, Pugach and R-Karpylivska, although the activity concentration of ^{137}Cs in individual milk samples from these villages was sometimes above the PL (up to a maximum of 160 Bq/l). In 2016, ^{137}Cs median activity concentration in milk was still found to exceed by far the Ukrainian PL for adults (100 Bq/l) in 6 villages of Rivne region, 4 of which are located in the Rokytne district (Stare Selo, Drozdyn', Vezhytsya, Perehodychi), and 2 in the Dubrovytsya district (Bydymlya and V-Ceremel'). A box and whisker plot summarising ^{137}Cs activity concentration values in milk samples collected in July 2016 from these villages is presented in Fig. 3.

Within the period from 2011 and 2016, the highest average values recorded were for milk from Vezhytsya village (in 2012), at 560 ± 183 Bq/l, which is about 6 and 14 times above the Ukrainian PL for adults and children, respectively. In terms of individual milk samples, the highest recorded value for ^{137}Cs activity concentration was 1000 Bq/l, found in an individual milk sample from Vezhytsya village collected in 2012, with individual samples collected from 4 other villages (Drozdyn', Perehodychi, Stare Selo and Yelne) above 500 Bq/l in 2011–2016 (Table 2).

Among livestock products, milk is the most sensitive to fluctuations in the level of radioactive contamination in the cattle's diet (IAEA, 2009; IAEA, 2010). This was clearly demonstrated by our previous analysis of seasonal dynamics of milk contamination by ^{137}Cs revealing that the content of this radionuclide in milk decreases during the autumn-winter season when cattle are kept inside (see Fig. S3 in SI) (Khomutinin et al., 2001). Apparently, during the autumn the cattle's diet is enriched with the newly harvested agricultural crops that have been grown on cultivated lands and which are not as contaminated with ^{137}Cs in comparison with natural grasses. Hence, this leads to a decrease of the ^{137}Cs content in the animal's daily diet and, respectively, to a decrease of milk contamination by this radionuclide. In addition, depending on the climatic conditions during the pasture period, the difference between maximum and minimum levels of milk pollution by ^{137}Cs at private households and farms in the Rivne region can reach an order of magnitude and more (See Fig. S3 in SI) (Khomutinin et al., 2001). Indeed, while the average value for ^{137}Cs in 20 samples of milk collected from Stare Selo village in May 2015 was 313 ± 28 Bq/l ($n = 20$), in September of the same year the average ^{137}Cs activity concentration in milk decreased to 213 ± 20 Bq/l (n = 20). A similar situation was observed in Vezhitsya village with a seasonal decrease in the average ^{137}Cs activity concentration in milk from 420 ± 36 (n = 20) to 270 ± 17 Bq/l ($n = 15$) (see Table S4 in SI).

3.2. Milk contamination forecast

The overall decreasing trend observed during the last few years in

Table 2
Most recent available official data (Lihtarov et al., 2013) and current study data on ^{137}Cs activity concentration in milk (mean value ± STD (range)) in settlements of Rivne region, Ukraine, in 2011–2016. n – number of samples.

Settlement	Official data			Current study											
	n	2011	2012	2011		2012		2013		2014		2015		2016	
		Bq/l	Bq/l	n	Bq/l	n	Bq/l	n	Bq/l	n	Bq/l	n	Bq/l	n	Bq/l
Stare Selo	5	380	381	16	303 ± 193 (130 - 670)	26	303 ± 107 (135 - 445)	20	225 ± 98 (74 - 430)	20	267 ± 115 (150 - 470)	40	263 ± 120 (68 - 590)	13	288 ± 124 (18 - 349)
Drozdyn′	5	429	186	16	434 ± 215 (110 - 690)	28	453 ± 169 (180 - 795)	20	186 ± 41 (90 - 260)	20	273 ± 120 (110 - 510)	35	344 ± 132 (150 - 640)	20	332 ± 96 (137 - 515)
Vezhytsya	5	308	288	20	482 ± 128 (240 - 660)	25	560 ± 183 (250 - 1000)	20	258 ± 94 (139 - 388)	20	436 ± 113 (280 - 620)	35	353 ± 145 (140 - 660)	11	333 ± 146 (177 - 525)
Perehodychi	5	234	179	20	346 ± 197 (80 - 750)	6	285 ± 157 (160 - 590)	20	137 ± 22 (100 - 160)	20	240 ± 101 (90 - 470)	20	467 ± 229 (160 - 720)	7	246 ± 73 (188 - 396)
Berezove	5	93	538	7	124 ± 43 (50 - 170)	–	- -	20	104 ± 75 (24 - 265)	20	80 ± 41 (3 - 130)	19	81 ± 67 (11 - 210)	20	16 ± 6 (8 - 28)
Yelne	5	212	364	16	193 ± 147 (45 - 430)	13	185 ± 99 (30 - 320)	11	350 ± 197 (149 - 710)	14	134 ± 111 (37 - 300)	14	18 ± 5 (12 - 28)	15	10 ± 3 (7 - 16)
Grabun′	5	330	319	13	185 ± 54 (150 - 330)	–	- -	–	- -	10	12 ± 3 (8 - 16)	10	73 ± 9 (60 - 88)	10	40 ± 35 (7–87)
Zabolottya	5	227	229	7	221 ± 39 (180 - 300)	5	35 ± 8 (12 - 44)	–	- -	10	11 ± 2 (7 - 14)	10	58 ± 59 (11 - 160)	8	19 ± 4 (9 - 22)
Khmil′	5	102	249	16	125 ± 52 (80 - 220)	–	- -	–	- -	10	96 ± 28 (70 - 140)	10	17 ± 4 (14 - 25)	10	72 ± 27 (11 - 123)
Vyry	5	119	129	25	87 ± 32 (50 - 160)	–	- -	–	- -	10	50 ± 34 (24 - 130)	10	18 ± 8 (10 - 35)	–	- -
Pugach	5	153	179	5	47 ± 16 (20 - 60)	–	- -	–	- -	5	32 ± 20 (14 - 53)	5	17 ± 6 (12 - 24)	–	- -
R-Karpylivska	5	123	129	10	89 ± 61 (30 - 205)	5	64 ± 35 (20 - 100)	–	- -	10	62 ± 36 (13 - 120)	10	52 ± 15 (24 - 80)	–	- -
Bydymlya	5	154	134	15	183 ± 96 (70 - 360)	–	- -	–	- -	10	176 ± 54 (100 - 250)	10	53 ± 19 (23 - 86)	7	165 ± 110 (18 - 310)
V-Ceremel′	5	149	300	14	216 ± 58 (120 - 300)	–	- -	–	- -	10	359 ± 151 (24 - 530)	10	299 ± 100 (180 - 448)	10	272 ± 73 (90 - 360)

the radiological contamination of milk originating from settlements where radioprotection measures have not been applied (Table 2) is consistent with, and therefore likely mostly attributable to, physical ^{137}Cs decay. The results of our long-term monitoring of ^{137}Cs contamination in foodstuffs produced in selected areas of the Rivne region therefore suggest that the natural remediation/mitigation processes (e.g., immobilisation of radiocaesium by soil) are virtually exhausted and that without application of additional appropriate countermeasures, more rapid improvement in the radiological situation in these areas cannot be expected in the immediate future.

The long-term dynamics of radioactive contamination decline in milk in the late phase of radiological accidents, due to a combination of

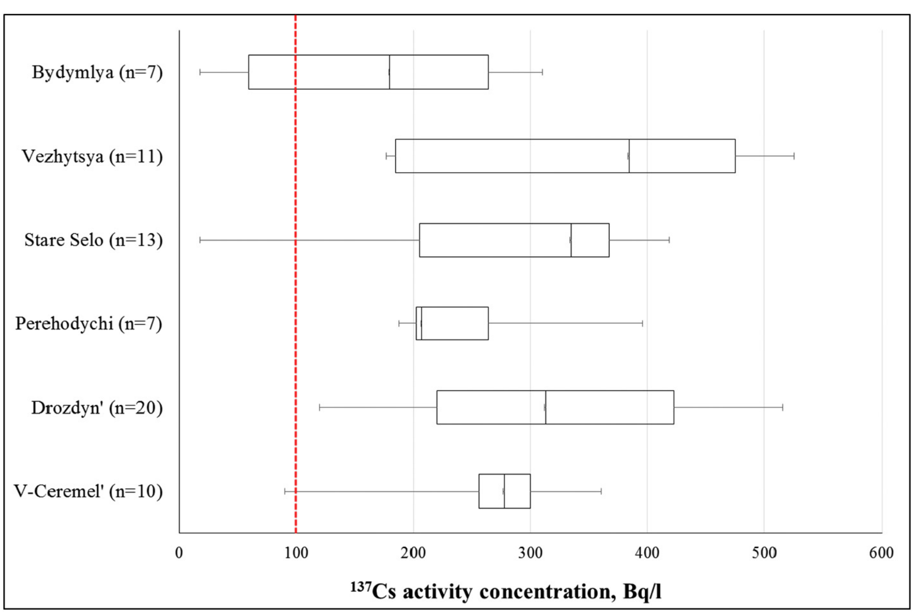

Fig. 3. Box and whisker plot of ^{137}Cs activity concentration (Bq/l) in milk samples collected from six villages of Rivne region, Ukraine, in 2016. The left and right limits of the boxes represent the 25th and 75th percentiles of the values, respectively. The solid vertical line within each box represents the median value. The left and right whisker are equal to the minimum and maximum values, respectively. Red vertical line – Ukrainian PL (Bq/l) for milk; n – number of samples. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Table 3
The parameters of dynamic ^{137}Cs activity concentration decline in cow's milk in settlements of Rivne region (see Eq. (4)) and the predicted year when the content of caesium in milk will be < 100 Bq/l. GSD – geometric standard deviation, STD – standard deviation.

Settlement	A(0), Bq/l (geometric mean·GSD)	(k + λ), year^{-1} (mean ± STD)	R^2	$T_{1/2\_eff}$, years (mean ± STD)	k/year (mean ± STD)	$T_{1/2\_eco}$, years (mean ± STD)	Predicted year for ^{137}Cs in milk < 100 Bq/l (mean ± STD)
Stare Selo	955·1.3$^{\pm 1}$	0.042 ± 0.004	0.8	17 ± 2	0.019 ± 0.004	36 ± 8	2040 ± 6
Drozdyn′	1350·1.5$^{\pm 1}$	0.051 ± 0.007	0.7	14 ± 2	0.027 ± 0.007	26 ± 7	2037 ± 8
Vezhytsya	2156·1.4$^{\pm 1}$	0.065 ± 0.006	0.8	11 ± 1	0.043 ± 0.006	17 ± 2	2033 ± 5
Perehodychi	1254·1.9$^{\pm 1}$	0.063 ± 0.011	0.6	11 ± 2	0.04 ± 0.01	17 ± 4	2026 ± 9
Bydymlya	1445·1.7$^{\pm 1}$	0.085 ± 0.010	0.8	8 ± 1	0.062 ± 0.01	11 ± 2	2019 ± 2
V-Ceremel′	1649·1.5$^{\pm 1}$	0.069 ± 0.008	0.8	10 ± 1	0.046 ± 0.008	15 ± 3	2027 ± 6

Table 4
Estimated annual effective doses for 2016 and last governmental official data for 2012 (Lihtarov et al., 2013). n/a – not available. Village names and associated internal doses are marked in bold to indicate estimates arising from application of the ReSCA model in the current study that were higher than those previously estimated using governmental Method-96.

Settlement	External, mSv/year			Internal, mSv/year			Total, mSv/year			
	2012 (official)	2016[a] Method-96	2016[b] ReSCA	2012 (official)	2016[a] Method-96	2016[b] ReSCA	2012 (official) Estimated	2012 (official) Measured Internal dose[c]	2016[a] Method-96	2016[b] ReSCA
Stare Selo	0.06	0.05	0.06	2.8	2.10	1.23	2.82	0.45	2.16	1.29
Drozdyn′	0.06	0.05	0.07	1.4	2.35	1.37	1.51	0.56	2.41	1.44
Vezhytsya	0.11	0.10	0.12	2.1	2.43	1.61	2.18	0.5	2.53	1.73
Perehodychi	0.16	0.15	0.17	1.3	1.80	1.49	1.48	0.44	1.94	1.66
Berezove	0.07	0.06	0.08	3.8	**0.12**	**0.35**	3.88	0.24	0.18	0.42
Yelne	0.11	0.10	0.12	2.6	**0.07**	**0.49**	2.74	n/a	0.17	0.61
Grabun′	0.08	0.07	0.09	2.3	**0.29**	**0.47**	2.35	0.43	0.36	0.56
Zabolottya	0.08	0.07	0.08	1.6	**0.14**	**0.37**	1.70	0.41	0.21	0.44
Khmil′	0.04	0.04	0.04	1.8	0.53	0.39	1.85	0.22	0.56	0.43
Vyry	0.08	0.07	0.08	0.9	**0.13**	**0.39**	1.02	0.09	0.20	0.47
Pugach	0.08	0.07	0.08	1.3	**0.12**	**0.38**	1.37	0.14	0.20	0.47
R-Karpylivska	0.11	0.10	0.13	0.9	**0.38**	**0.67**	1.04	0.32	0.48	0.80
Bydymlya	0.11	0.10	0.13	1.0	1.20	1.06	1.11	0.23	1.30	1.19
V-Ceremel′	0.17	0.15	0.18	2.2	1.99	1.63	2.35	0.43	2.14	1.81

[a] Estimated in the current study utilizing official governmental Method-96 (Lihtarov et al., 1996).
[b] Estimated in the current study utilizing ReSCA model (Ulanovsky et al., 2011).
[c] Human body measurements conducted using WBC (Lihtarov et al., 2013).

strong absorption of ^{137}Cs by soil particles and radioactive decay, are usually described by an exponential dependence as per Eq. (4) (IAEA, 2010):

$$A(t)_{milk} = A(0)_{milk} \bullet e^{-(k+\lambda)\bullet t} \quad (4)$$

where $A(t)_{milk}$ and $A(0)_{milk}$ - the activity concentration of ^{137}Cs in cow's milk (Bq/l) at the time-point t (years) after 1986 and at time zero immediately after initial radiological contamination of the soil, respectively;

λ - radioactivity decay constant that defines as $\lambda = Ln(2)/T_{1/2}$. For ^{137}Cs, $\lambda = 0.023$/year and $T_{1/2} = 30.17$ years;

k – coefficient that defines as $k = Ln(2)/T_{1/2\_eco}$, where $T_{1/2\_eco}$ - the ecological half-life of ^{137}Cs activity concentration in milk (years). The effective half-life $T_{1/2\_eff} = Ln(2)/(k+\lambda)$ (years).

Eq. (4) has been used to forecast milk contamination with ^{137}Cs. The parameters of the exponential dependence described by Eq. (4) are provided in Table 3.

Extrapolation from the measurements of ^{137}Cs activity concentration in cow's milk in 6 settlements of Rivne region (Stare Selo, Drozdyn', Vezhytsya, Perehodychi, Bydymlya, and V-Ceremel') reported by the Ukrainian Government in 1991–2012 (Bazyka et al., 2016; Lihtarov et al., 2012, 2013), together with the data reported in this study for the period 2011–2016, indicates that the effective half-life for ^{137}Cs in milk ($T_{1/2\_eff}$) in these settlements may vary from 8 to 17 years. Hence, the activity concentration of ^{137}Cs in cow's milk may persist above 100 Bq/l for another 23 years or more (until 2040 or later).

However, it has previously been shown (Fesenko et al., 2013, 2007a; Jacob et al., 2009; Ulanovsky et al., 2011) that the application of protective countermeasures, such as Ferrocyn administration, may decrease the activity concentration of ^{137}Cs in milk threefold even over only a few days. For example, a decrease in milk contamination by ^{137}Cs has been reported during the period of 1994–1996 as a result of extensive application of various remedial actions, including Ferrocyn application (see Fig. S1 in SI). Hence, if Ferrocyn had been applied to cows in settlements in the Rivne region, the majority of these settlements (with the possible exception of Drozdyn' and Vezhytsya) might then have access to milk with radiocaesium activity concentrations below the Ukrainian PL.

The practical application of Ferrocyn ceased in Ukraine in 2009 (UN, 2011), despite the fact that neither the legal requirements in Ukraine nor the international nuclear safety standards proclaimed by the International Atomic Energy Agency in relation to food (IAEA, 2016, 2011) had been met.

In order to evaluate the likely effectiveness of the different radiological remedial/mitigation actions that could be usefully applied within the Rivne region, taking into account the recent (2015–2016) radiological situation in this area, and therefore to help optimise the combination of such measures, we have applied the ReSCA model (Ulanovsky et al., 2011).

3.3. Optimization of a remedial strategy

Optimizing a radiological remediation/mitigation strategy requires consideration of a wide range of technical (e.g. effectiveness, cost,

Table 5
Recommended strategy of remedial actions (RA).

Settlement	RA	Dose (mSv/year)		Cost of RA[a]	Averted dose	Cost/averted dose (kEuro/man-Sv)
		before RA	after RA	(kEuro)	(man-Sv)	
Bydymlya	FA	1.19	0.84	4.5	0.415	10.8
Drozdyn′	FA	1.44	0.75	17.2	3.096	5.6
Stare Selo	FA	1.29	0.67	27.2	4.374	6.2
Perehodychi	FA	1.66	1.13	4.4	0.611	7.3
	FA + MF	1.13	1.04	0.4	0.016	25.5
	FA + MF + IM	1.04	0.98	1.5	0.019	81.2
V-Ceremel′	FA	1.81	1.23	2.0	0.297	6.6
	FA + MF	1.23	1.13	0.1	0.005	23.6
	FA + MF + IM	1.13	1.07	0.5	0.006	75.3
	FA + MF + IM + FP	1.07	0.86	0.9	0.011	86.9
Vezhytsya	FA	1.73	1.02	11.5	2.135	5.4
	FA + MF	1.02	0.95	0.9	0.025	35.7
Total				71.1	11.010	6.5

RI - Radical Improvement of grassland, which includes removing vegetation, ploughing, liming, fertilization, and reseeding; D - Drainage (used only in connection with RI on wet peat soil); FA - Ferrocyn Application to cows; FP - Feeding Pigs with uncontaminated fodder before slaughter; MF - Mineral Fertilizer application on potato fields; IM - Information campaign on Mushrooms and other forest produce consumption; RS - Removal of contaminated Soil from populated areas in a settlement.
a - Costs of Remedial Actions estimated according to Ulanovsky et al. (2011).

feasibility), environmental, and social (acceptability, opportunities for self-help actions) factors. To the authors' knowledge, no such strategy has been in place in Ukraine since 2012.

The last available governmental dosimetry certification data revealed important discrepancies between estimated average annual effective dose and the dose measured in humans using the Whole Body Counter (WBC) (see Table 4). Indeed, the official estimated effective doses to population were 3–10 times higher than those obtained during population monitoring with WBC, and the doses obtained during monitoring never exceeded 1 mSv/year. This highlights the conservatism in data analysis utilizing the old methodology established in 1996 (Lihtarov et al., 1996). Also, the consumption of locally produced milk in the Rivne region, and in Ukraine as a whole, has significantly declined in comparison to that in 1990–1996, due in part to a decrease in dairy cow numbers in Ukraine. The cow population in 2012 decreased by a factor of 3.3 and 2.9 times in comparison to 1990 and 1996, respectively. This has caused a decrease in milk production by a factor 2.2 and 1.4 times, respectively (SSC of Ukraine, 2017). In addition, due to a challenging economic situation in Ukraine, rural populations have tended to sell more milk to milk processing plants rather than consume it themselves, in order to generate income.

Nevertheless, our estimation of the total effective dose for 14 villages, using either the official Method-96 (Lihtarov et al., 1996) or ReSCA model (Ulanovsky et al., 2011), has shown (see Table 4) that the populations of 6 settlements (Stare Selo, Drozdyn', Vezhytsya, Perehodychi, Bydymlya, and V-Ceremel') are still receiving an effective dose above 1 mSv/year in 2016. According to the Law of Ukraine those settlements can be considered as contaminated and they require implementation of remedial actions to decrease the effective dose to the population.

For these 6 settlements, average annual effective dose, which was calculated using Method-96, ranged from 1.30 to 2.53 mSv/year, and was, on average, 1.4 times higher than that obtained using ReSCA model for the Representative Person (range: 1.19–1.81 mSv/year). The higher estimates obtained using Method-96 are explained in large part by the high milk consumption rates estimated for this region in 1996, at 355 l/year (Lihtarov et al., 1996). Average estimated milk rates from our own survey in 2012 were substantially lower, at 68 ± 8 l/year, and these lower intakes were used for the internal dose estimation utilizing the ReSCA model. In contrast, for the 7 villages, which had estimated effective doses below 1 mSv/year (marked in bold in Table 4), the ReSCA model yielded internal dose estimates that were higher than those obtained using Method-96. Such a discrepancy could result from a number of factors, including:

- the difference in the number of food products considered for the internal dose estimation in the two methods applied. While Method-96 takes into account only contamination of milk and potatoes by ^{137}Cs, the ReSCA model accounts for milk, beef, pork, potatoes, and mushrooms;
- the relatively low (< 40 Bq/l) content of ^{137}Cs in milk in these seven villages. This has led to low estimated values of internal doses in case of Method-96 application, while internal dose calculated using ReSCA model was higher due to additional contributions from consumption of pork, beef, and mushrooms that have not been accounted for in Method-96.

In cases in which ^{137}Cs content of milk was > 70 Bq/l, milk was the primary contributor to estimated internal dose when applying both Method-96 and ReSCA model accounting for over 50% of the estimated internal dose. Hence, the addition of contributions from pork, beef, and mushrooms in the ReSCA model, which are not considered in Method-96, played a comparatively less significant role in estimation of the internal dose in such cases. However, when ^{137}Cs activity concentration in milk was < 70 Bq/l, mushrooms contributed greatly (up to 50%) to the estimated internal dose, which resulted in higher values obtained with the ReSCA model than using Method-96.

In comparison to other countries that were immediately affected by the accident at ChNPP (Russia and Belarus), Ukraine had smaller number of the settlements (e.g., 25 settlements officially reported for 2012) (Lihtarov et al., 2013) with the average annual effective dose above 1 mSv/year. For Belarus and Russia, more recent official data are available; for example, in Belarus in 2015, the exceedance of average annual effective dose above 1 mSv/year was recorded in 91 settlements (NSPCRMHE, 2015), while in Russia in 2017, this was still the case for 135 settlements (Bruk et al., 2017), with 2 settlements above 5 mSv/year. One of the main reasons for the relatively high number of settlements in Russia with the annual effective dose exceedance is that remedial actions have been less extensively applied than those performed in Ukraine and Belarus (Jacob et al., 2009).

Our proposed optimized Remedial Actions (RA), including minimization of the averted dose, may substantially reduce the exposure of the population of the 6 settlements mentioned above to radioactive elements (see Table 5). For example, the application of Ferrocyn (FA) to cows would be expected to result in decrease of the effective dose in three settlements (Stare Selo, Drozdyn', and Bydymlya) below 1 mSv/year and would cost about 50,000 Euro.

To achieve a similar result in another three settlements (V-Ceremel', Perehodychi, and Vezhytsya), it would be necessary to conduct additional actions in combination, including:

- application of Mineral Fertilizers (MF, cost 3380 Euro) for potato fields in all three settlements;
- conduct of an Information campaign on Mushrooms consumption (IM, cost 1989 Euro) in V-Ceremel' and Perehodychi settlements;
- Feeding Pigs (FP, cost 924 Euro) with uncontaminated fodder before slaughter in V-Ceremel settlement'.

Hence, with estimated costs of about 71,000 Euro for a total of 8336 inhabitants, with the FA action being the most expensive (over 66,000 Euro), our calculations indicate that the effective dose to the population in 6 settlements considered in this study could be decreased to below 1 mSv/year (see Table 5).

The averted collective dose would then be 11 man-Sv with costs of about 6500 Euro/man-Sv, which is consistent with world-wide practice (Los et al., 1999) and amounts to less than the internationally accepted standard of 10,000 Euro/man-Sv commonly used for economic justification of remedial measures (Fesenko et al., 2007a).

In summary, improved radiation protection of those communities in Ukraine that are among the most affected by the Chernobyl accident could be achieved by implementation of a combination of relatively simple and inexpensive remedial actions, leading to: a) a decrease in radiocaesium activity concentration in agricultural products below PLs and b) a reduction in effective dose to population below 1 mSv/year – as specified by the Law of Ukraine (Supreme Council of Ukraine, 1991a). The costs for such RA implementation in Ukraine amount to < 100,000 Euro, which is only a fraction (0.03%) of the total budget allocated, for example, in 2015 (373 million Euro) for measures to remediate/mitigate impacts of the Chernobyl accident and improve protection of the affected population in Ukraine. This overall budget covers a range of the funds including the Chernobyl Foundation "Shelter" and several international projects (including those sponsored by the EBRD) designed to provide technical assistance with these initiatives. In addition, in the recent IAEA Safety Standards (IAEA, 2016) it is stated in Requirement 9 ("System for protective actions to reduce existing or unregulated radiation risks") that "The government shall establish an effective system for protective actions to reduce undue radiation risks associated with … contamination from past activities or events, consistent with the principles of justification and optimization". However, to the best of our knowledge, no such actions have been taken in Ukraine since 2009 in fulfilment either of the provisions of Ukrainian Law or of IAEA safety standards with respect to the implementation of remedial actions on the radioactively contaminated territories of Ukraine.

Similarly, the absence of government monitoring of the radioactively contaminated territories of Ukraine for the last few years also prevents any re-evaluation of the existing boundaries of the contaminated zones that were established in 1991. Of 826 villages in these zones that showed exceedance of the effective dose of 1 mSv/year in 1991 (Lihtarov, 2012), such exceedance remained in 25 villages by 2012 (Lihtarov et al., 2013). However, under Ukrainian Law (Supreme Council of Ukraine, 1991b), formal re-evaluation of the zone boundaries could be done *only* on the basis of an official expert analysis of the radio-ecological situation, which effectively ended in 2012.

4. Conclusion

The current study reports for the first time the temporal trends of ^{137}Cs in cow's milk originating from private households and farms located in radioactively contaminated areas of Ukraine for the period of 2011–2016. Our results show that during this period 14 settlements in Ukraine, with about 18,000 inhabitants in total, continued to consume ^{137}Cs contaminated cow's milk that exceeded Ukrainian permissible levels by up to 6 and 14 times for adults and children, respectively.

By 2016, there were still 6 settlements in the Rivne region for which populations continue to receive estimated effective internal doses above 1 mSv/year through consumption of radioactively contaminated locally produced foods. Unfortunately, the situation in these settlements will remain largely unchanged in the immediate future as natural remedial/mitigation capacities in the area are mostly exhausted. Our results show that the ecological half-life ($T_{1/2\_eco}$) of ^{137}Cs in milk from these settlements is in the range of 11–36 years, which is comparable with the period of physical ^{137}Cs decay (30 year). Without the implementation of the RA discussed above, the activity concentration of ^{137}Cs in cow's milk from the Rivne region could remain above PL (i.e., 100 Bq/l) for the following 23 years – until at least 2040. The annual effective doses estimated for 2016 for populations of these 6 settlements, using both Ukrainian official methodology (Method-96) and the alternative ReSCA model, exceed the PL of 1 mSv/year.

The strategy of the remedial actions implementation proposed in the current study suggests that the effective dose to population in 6 considered settlements in Ukraine could be decreased below 1 mSv/year with a funding of about 70,000 Euro.

Our findings are in good agreement with previously reported data (Fesenko et al., 2007a; Jacob et al., 2009; Waddington et al., 2017) and highlight that remedial measures implementation in investigated areas in Ukraine are still of a great importance and can be achieved with a high effectiveness in terms of both radiological and economic factors.

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Declarations of interest

none.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envint.2018.04.053.

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