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# A protocol for the radiological assessment for agricultural use of land in Ukraine abandoned after the Chornobyl accident



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#### ABSTRACT

There is a need in Ukraine for re-evaluation of the status of lands outside the Chornobyl Exclusion Zone which were abandoned in the years after the accident. Since the 1991 criteria for zoning were put in place, there has been no re-classification of abandoned lands even though radionuclide contamination density and, for radio-caesium isotopes, mobility have both declined. This study describes the development of a protocol for assessment of abandoned lands in Ukraine based on a 100 ha experimental plot. A simple method of quantification of <sup>137</sup>Cs contamination density was developed using external dose measurement whilst other relevant radionuclides (<sup>90</sup>Sr; <sup>241</sup>Am and Pu isotopes) were quantified using selected soil samples. Modelling of uptake of radionuclides in eight key crops shows that the study field could be re-used for agriculture according to Ukrainian regulatory limits. Monte Carlo modelling of potential dose to farm workers showed that dose was dominated by external exposure and that doses were significantly below 1 mSv y<sup>-1</sup>. Based on statistical analysis of soil-plant concentrations ratios, criteria were derived for assessment of suitability of agricultural land for production. The criteria are applicable to areas of soddy-podzolic sandy and sandy loam soils (Podzoluvisol) typical in these regions of Northern Ukraine. They are not applicable to high organic matter soils (Histosols) where soil-plant concentration ratios are likely to be much higher for radiocaesium.

# 1. Introduction

There is an identified need in Ukraine to develop a new strategy for the management of the Chornobyl contaminated areas (Presidential Decrees N $^{\circ}$ s 141/2016 and 174/2016). This study presents a field test of a practical method for the radiological assessment of contaminated land to ensure that agricultural crops are below regulatory limits. A further aim is to evaluate whether annual effective radiation dose rates to agricultural workers are below relevant limits if currently abandoned lands were to be brought back into production. As an example of contaminated land evaluation for derestriction, the methods developed here could be applied to other radioactively contaminated sites worldwide.

Over the years since the Chornobyl accident, radiation doses have

declined significantly due to radioactive decay, redistribution in the soil profile and erosion from surface soils. For radiocaesium, reduction in more bioavailable forms has led to long term declines in surface waters and foodstuffs, particularly in areas of mineral soils such as those studied here (Fesenko et al., 2023; Smith et al., 2000). Milk consumption from areas of high organic content soils remains a significant source of internal dose (Labunska et al., 2018) in some parts of Northern Ukraine. Internal radiation doses from agricultural ecosystems tended to be much lower than those from forest and semi-natural ecosystems (Drozdovitch et al., 2022; Fesenko et al., 2000). The large extent of agricultural land which currently remains officially abandoned is therefore not expected to give rise to high ingestion doses from agricultural products (Kashparov et al., 2022). To remove restrictions on such land, however, it is necessary to demonstrate that crops can be grown which meet the

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relatively strict radiological food safety criteria in Ukraine.

#### 1.1. Land zoning and food protection criteria

The radiation exposures relevant to the reuse of abandoned lands in Ukraine outside the Chornobyl Exclusion Zone are within the chronic low dose exposure category as defined in (ICRP, 2007). Given the long time period since the 1986 Chornobyl accident, the contamination is categorised as an existing (rather than an emergency) exposure situation.

The criteria for the zoning of land in Ukraine were set out in 1991 based on the objective of ensuring that effective equivalent radiation exposures to the public were no greater than  $5.0 \text{ mSv y}^{-1}$  and that, over time, maximum doses should reduce to  $1 \text{ mSv y}^{-1}$  or less (Smith and Beresford, 2005). In practice, these zones were defined on the basis of contamination density of radionuclides, as summarised in Table 1. Fig. 1 shows the extent of lands currently classified in zones 1 ("Chornobyl Exclusion Zone", CEZ), 2 ("Zone of Unconditional (Obligatory) Resettlement" (ZoR), and 3 ("Zone of Guaranteed Voluntary Resettlement"). In accordance with the Law of Ukraine, lands in Zones 1 and 2 are radiation hazardous, where agricultural production is prohibited. In Zone 3, land is considered radioactively contaminated, but can be used for agricultural production, with protective countermeasures applied if necessary.

The zoning of settlements and agricultural land in Ukraine has not changed significantly since 1991 (Kashparov et al., 2022). The original zoning was based on extensive measurement of gamma-emitting radionuclides and <sup>90</sup>Sr activity concentration in soil samples (Baryakhtar, 1995). For each bulked sample, a field plot of approximately 50 ha was divided into five 10–15 ha sectors and a sample collected in each sector to 20 cm depth using an auger. The samples from each of the five sectors were then combined to form a single sample for subsequent analysis. The external gamma exposure rate was measured at all the sampling points. The results obtained by (Baryakhtar, 1995) formed the basis for the subsequent zoning of Ukrainian territory (Table 1).

In Ukraine, restrictions on an area of land can only be changed by legislative change of the zoning procedure, whereas in Russia and Belarus restricted areas were reviewed every five years based on changes in the radiological situation and the need for their use. In Russia and Belarus, a large discrepancy between zoning and measured radioactivity levels was found and a revision of the boundaries of contamination zones recommended.

Current Ukrainian limits for <sup>137</sup>Cs and <sup>90</sup>Sr in locally important

agricultural products are shown in Table 2. These limits are broadly consistent with those in force in Russia and Belarus for <sup>137</sup>Cs but, for <sup>90</sup>Sr, permissible levels were generally significantly lower in Belarus than in Ukraine and Russia (Balonov et al., 2018); these authors recommended a scheme (not yet implemented) for harmonisation and simplification of standards between Ukraine, Belarus and Russia. The permissible level for radiocaesium in foods in Japan, set from one year after the Fukushima Daiichi accident, is 100 Bq kg<sup>-1</sup> (Nihei et al., 2016) for all foods except milk and infant foods which have an upper limit of 50 Bq kg<sup>-1</sup>.

The Permissible Levels for agricultural products in force in Ukraine are lower than the European Union permitted levels of radioactive contamination of food following the Chornobyl accident. The Euratom Regulations (European Union Council, 2016) gave maximum activity concentrations of <sup>90</sup>Sr of 125 Bq kg<sup>-1</sup> for dairy products, 75 Bq kg<sup>-1</sup> for infant foods and 750 Bq kg<sup>-1</sup> in other major foodstuffs. For longer lived isotopes other than <sup>90</sup>Sr and alpha emitters (i.e. in this context <sup>137</sup>Cs): 1000 Bq kg<sup>-1</sup> for dairy produce, 400 Bq kg<sup>-1</sup> for infant foods and 1250 Bq kg<sup>-1</sup> for other major foodstuffs.

In the Ukrainian system, limits on crops are based on both  $^{137}$ Cs and  $^{90}$ Sr concentration in the crop. So, for example, if  $^{137}$ Cs was at 70 % of the relevant limit and  $^{90}$ Sr was at 31 % of its relevant limit, then the crop would be considered to be above the limit and unsuitable for market distribution. In practice, the limiting activity concentration in crops is determined by considering the transfer of radioactivity to crops and accounting for the uncertainty in this factor. This will be discussed further in the Methods section.

# 1.2. Deposition patterns of relevant radionuclides

The radionuclides of primary concern outside the Chornobyl Exclusion Zone are <sup>137</sup>Cs and <sup>90</sup>Sr, though this study also considers isotopes of Pu. The activity ratio <sup>137</sup>Cs: <sup>90</sup>Sr for the fuel component of Chornobyl fallout is approximately 1 (Kashparov et al., 2020). This ratio (as well as the ratios of <sup>137</sup>Cs to other radionuclides of the fuel component, including <sup>241</sup>Am and Pu isotopes) increases with distance from the Chornobyl nuclear power plant (Mück et al., 2002). This is due to the increase in the fraction of condensed radiocesium, which leaked out during the high-temperature annealing of nuclear fuel during the accident. Therefore, there is not a strong correlation between the <sup>137</sup>Cs activity and the activity of those radionuclides (including <sup>90</sup>Sr, Pu isotopes and hence also <sup>241</sup>Am) which were released during the accident mainly in the form of fuel particles.

# Table 1

Radiological zoning criteria in Ukraine<sup>a</sup>.

Zone 1: The Chornobyl Exclusion Zone (CEZ)

Territories adjacent to the Chornobyl NPP which were evacuated in 1986. The criterion for evacuation was that dose to the population should be less than 100 mSv y<sup>-1</sup> for the first year after the accident.

Zone 2: Zone of an unconditional (obligatory or compulsory) resettlement (lands where people cannot live and cannot carry out agricultural production, or any other economic activity).

Effective annual dose is greater than 5 mSv  $y^{-1}$  (the average effective dose for the population of the village). The effective dose is calculated using the Ministry of Health approved method (Guidance, 1998), with external dose calculated from the average density of contamination and internal dose from the average consumption rate of food in Ukraine, which is estimated using measured average activity concentrations in milk and potatoes from the village;

OR

Based on contamination density, if

 $^{137}$ Cs > 555 kBq m<sup>-2</sup> or

 $^{90}\text{Sr} > 111 \ \text{kBq} \ \text{m}^{-2}$  or Pu isotopes (  $^{238}\text{Pu};^{239}\text{Pu};^{240}\text{Pu})^{**} > 3.7 \ \text{kBq} \ \text{m}^{-2}$ 

Zone 3: The zone of a guaranteed voluntary resettlement (where people can live and agriculture is permitted, but aid is available for people to relocate if they wish to): Effective annual dose is greater than 1 mSv  $y^{-1}$  but less than 5 mSv  $y^{-1}$  (the average effective dose for the population of the village calculated as described for Zone 2).

$$\begin{split} 185 <^{137} Cs < 555 \ kBq \ m^{-2}, \ or \\ 5.5 <^{90} Sr < 111 \ kBq \ m^{-2}, \ or \\ 0.37 <^{238\cdot239,240} Pu < 3.7 \ kBq \ m^{-2} \end{split}$$

<sup>a</sup> The contamination density criteria presented here are for radionuclides of Chornobyl origin only. Thus, radioactivity from nuclear weapons testing is not included when comparisons are made against the contamination density criteria. \*\* Note that the law enforcing these zoning criteria mistakenly states all Pu isotopes – it should have specified only these alpha-emitting isotopes and not<sup>241</sup>Pu. This has led to confusion: although the dose from<sup>241</sup>Pu (which emits a low-energy beta particle) is insignificant, its contamination density is much higher than the alpha-emitting Pu isotopes.



Fig. 1. Zoning of contaminated territory of Ukraine according to current regulations (Ministry of Emergencies of Ukraine, 2008) and field study plot. The "Compulsory relocation zone" is Zone 2 in Table 1 and "Zone of guaranteed voluntary relocation" is Zone 3.

Current Ukrainian Permissible Levels for  $^{137}$ Cs and  $^{90}$ Sr in foodstuffs per unit dry mass (dm) or fresh mass (fm) as they go to market (MHPU, 2006).

Food or drink	Permissible Level for <sup>137</sup> Cs Bq kg <sup>-1</sup> or Bq l <sup>-1</sup>	Permissible Level for <sup>90</sup> Sr Bq kg <sup>-1</sup> or Bq $l^{-1}$
Grains, including wheat, rye, oats, barley, millet, buckwheat, rice, maize, sorghum and other cereals (dm; air dried)	50	20
Dried legumes including peas, beans, soybean, lentils (dm)	50	30
Sunflower seeds	70	10
Milk (fm)	100	20
Potatoes (fm)	60	20
Fresh vegetables including leafy vegetables, stone fruits, root vegetables, legumes, cultivated mushrooms (fm)	40	20
Meat of domestic animals including poultry (fm)	200	20

The western trace of radioactive fallout (where almost all "Zone 2" abandoned lands are located) was formed mainly by condensed radiocesium and washout with rainfall was an important deposition process, leading to a patchy pattern of deposition. Due to the distance from the ChNPP, there was only a very small fraction of radionuclides of the fuel component, particularly <sup>90</sup>Sr, <sup>241</sup>Am and Pu isotopes. In comparison to the highly heterogeneous <sup>137</sup>Cs contamination, the contamination radionuclides associated with the fuel component is more consistent. To the West, the <sup>90</sup>Sr condensed component is only found in significant quantities at closer distances (up to 80 km) from the ChNPP.

# 1.3. Scope of this study

This study provides a standardized approach for estimating <sup>137</sup>Cs, <sup>90</sup>Sr and <sup>239-240</sup>Pu contamination densities of agricultural lands, which were withdrawn from economic use after the Chornobyl accident (i.e. that land currently classified as Zone 2), to assess the current suitability for using the land for agricultural production.

The developed protocol is suitable for evaluating potential re-use of land for all main agricultural crops including:

Cereals (wheat, rye, oat, barley)
Maize.
Sunflower (seeds)
Potatoes.
Root crops (beet, carrot)
Leafy vegetables.
Leguminous crops (soybean)
Grass and pasture.
Animal products.
Where "grass" is defined here as cu

Where "grass" is defined here as cultivated grasses for cattle fodder and "pasture" is defined as uncultivated meadow with a mix of grass and other plants edible by domestic animals. As discussed below, grass and pasture limits are based on calculation of uptake to milk and meat to ensure that the limits for milk and meat are not breached.

This study applies to the soddy-podzolic sandy and sandy loam (Podzoluvisol) soils typical of areas currently in the ZoR (Zone 2). It does not apply to highly organic soils where soil-plant concentration ratios are likely to be significantly higher than those used in this assessment. The study further evaluates doses to agricultural workers to ensure that occupational dose limits are not exceeded in lands de-restricted for agricultural activity. It does not consider doses to civilians living in settlements close to the agricultural land.

## 2. Methods

## 2.1. Site description and survey

A detailed site survey was carried out at a test plot at Mezhiliska (Fig. 2), approximately 60 km to the south-west of the Chornobyl reactor and located close to Osyka village (Narodychi district, Zhytomyr region  $51^{\circ}04'26''$  N  $29^{\circ}21'31''E$ ). The district is currently in Zone 2 (Zone of Unconditional (Obligatory) Resettlement) and most of the population was relocated in the early 1990's. The area of the field is about 100 ha. After the Chornobyl accident, this field was used for agriculture for several years and deep ploughing to 25 cm is likely to have been part of the land management regime. At the time of the survey (July 24, 2018), the field was predominantly natural herbaceous vegetation.

The ambient gamma dose rate survey was carried out with STORA TU dosimeters (Ecotest, Ukraine) along transects with readings taken at approximately 30 m intervals along the transect at a height of approximately 1m above the ground. Transects were spaced in an approximate grid with less than 200 m between transects. The STORA-TU transferred measurements in real time to a smartphone running Android OS. Using the GS Ecotest application, this information was processed, displayed and stored in a database. Spatial interpolation of the gamma dose rate was conducted using QGIS software (QGIS, 2018).

## 2.2. Soil sampling and analysis

In total, 19 composite soil samples were collected within the field. Each composite sample consisted of 5 soil cores collected randomly to a depth of 25 cm within a sampling plot (approx. area of  $100 \text{ m}^2$ ) using a cylindrical corer of 3.5 cm diameter. Sampling points were approximately uniformly distributed within the field (Fig. 2).

The soil samples were dried, homogenised and weighed into Marinelli beakers (1000 cm<sup>3</sup>) prior to analysis. Activity concentrations of <sup>137</sup>Cs and naturally occurring gamma-emitting radionuclides in all soil samples were measured using a low-level gamma-spectrometer with a high-purity germanium detector (GEM-30185, EG&G Ortec, USA) equipped with a multichannel analyser (Model *919*, EG&G Ortec, USA)

and a passive protection device, and using the *OMNIGAM* software (EG&G Ortec, USA). Calibration of the spectrometer was conducted using certified standards (soil matrix multi-nuclides standard).

Activity concentrations of  ${}^{90}$ Sr in soil were measured using radiochemical separation of the radioisotope (ISO18589-5) followed by beta counting of its daughter,  ${}^{90}$ Y, using a beta-spectrometer (SEB-70, AKP (Ukraine)). Activity concentrations of plutonium isotopes ( ${}^{238}$ Pu,  ${}^{239,240}$ Pu) were determined in the same soil samples as those used for  ${}^{90}$ Sr analyses. A standard method (ISO18589-4), which is based on the selective chromatographic extraction of Pu using specific resins (TRU), was applied. Electrodeposited sources of Pu were measured using a Soloist  $\alpha$ -spectrometer with a Soloist-U0300 detector (EG&G ORTEC).

Samples were decay corrected to the date of sampling – July 27, 2018. All measured values or radionuclide activities were reported with combined uncertainties (p = 0.95).

## 2.3. Estimation of activity concentration in crops

Activity concentrations in crops were predicted using the measured soil activity concentrations and published transfer parameters based on the following equation:

$$C_p = \alpha \cdot C_r \cdot C_s, \tag{1}$$

Where  $C_p$  is the radionuclide activity concentration in a plant product (fresh mass, fm), Bq kg<sup>-1</sup>;  $\alpha$  the dry matter content;  $C_s$  the radionuclide activity concentration in soil (dry mass, dm) in Bq kg<sup>-1</sup> and  $C_r$  the plant-soil concentration ratio for the plant (dry mass basis). Mean values of concentration ratio for simplified soil categories and plants can be found in TRS 472 (IAEA, 2010). However, individual values of  $C_r$  can vary by over orders of magnitude even for the same plant and soil group. Thus, available regional data on soil-to-plant factors of <sup>137</sup>Cs and <sup>90</sup>Sr, which have been collected during the last 10 years, (see Supplementary Data) were compared to those from TRS 472. The radiological assessments presented here were based on both regional and TRS 472  $C_r$  values (sandy soil category), with regional data collated here compares well with that in TRS472. In general (5 out of 6 Cr values for <sup>137</sup>Cs and 3



Fig. 2. Test field at Mezhiliska and location of soil sampling points. Image from Google Earth.

Geometric means (i.e. EXP(mean of LN( $C_r$ ) values)) and geometric standard deviations (i.e. EXP(std dev of LN( $C_r$ ) values)) of soil to plant transfer factors ( $C_r$ ) of  $^{137}$ Cs and  $^{90}$ Sr.  $C_r$  is calculated on a dry mass basis (Bq/kg dry crop  $\div$  Bq/kg dry soil).  $\alpha$  is the dry mass/fresh mass ratio for the crop.

Crop	α	<sup>137</sup> Cs			<sup>90</sup> Sr		
		N	GM	GSD	N	GM	GSD
Potatoes	0.21	35 <sup>a</sup>	0.063	1.7	39	0.22	2.6
Leafy vegetables (salad)	0.08	96	0.12	4.1	72	1.7	4.1
Root crops (red beet)	0.16	37	0.062	2.5	26	1.1	3.7
Leguminous crops (soya seeds)	0.87	15 <sup>a</sup>	0.057	2.0	15 <sup>a</sup>	0.98	2.0
Maize (grain)	0.85	18 <sup>a</sup>	0.018	2.0	18 <sup>a</sup>	0.03	1.8
Cereals (grain)	0.87	67 <sup>a</sup>	0.020	2.3	108 <sup>a</sup>	0.47	2.3
Sunflower (seeds)	0.87	5 <sup>a</sup>	0.037	1.5	2 <sup>a</sup>	0.30	1.32
Grass (hay)	0.85	18 <sup>a</sup>	0.037	1.8	10 <sup>a</sup>	1.4	1.7
Pasture	0.85	25 <sup>a</sup>	0.31	2.2	14 <sup>a</sup>	1.8	1.9

N - size of sample; GSD - geometric standard deviation.

<sup>a</sup> Regional data of UIAR (see Supplementary Data); other  $C_r$  values are from TRS-472 (IAEA, 2010) for sand soil group.



**Fig. 3.** Comparison of mean region-specific Cr values collated for this study with those from (IAEA, 2010). Each data point represents a different crop type (see Table 3 for Cr values used and their source).

out of 5 for <sup>90</sup>Sr), the regional specific values of Cr were lower than those from IAEA TRS-472. This gives some confidence that for those crops where TRS-472 data were used (i.e. no local data available), the model is likely to be conservative.

It can be seen in Table 3 that  $C_r$  values for the category "Pasture" are

significantly (8x) higher than those for the category "Grass". This is due to the fact that pasture (defined here as semi-natural uncultivated land) is generally composed of more organic, often floodplain soils which less strongly bind radiocaesium than more mineral and cultivated agricultural land. Lack of fertilisation and liming will also increase radiocaesium uptake. In addition, pasture is composed of a wide range of plant species which may have higher transfer ratios than grasses.

Plant-soil  $C_r$  values have most often been described by a lognormal probability distribution (see also (Khomutinin et al., 2020)) and this has been assumed here (this was tested for those crops for which ranges of  $C_r$  data were available). Assuming a lognormal probability distribution, the average value (median) of the concentration of activity of radionuclides in each crop and also the probability of exceeding the permissible levels can be estimated using the equation:

$$q = 1 - \int_{0}^{C_{\rho}^{0}} \frac{1}{\sqrt{2\pi} \cdot C_{\rho} \cdot \mathbf{s}_{p}} e^{-\frac{1}{2} \left( \frac{\ln(C_{\rho}) - \ln(\overline{C}_{\rho}))}{\mathbf{s}_{p}} \right)^{2}} \cdot dC_{\rho}$$
(2)

Where *q* is the probability of exceeding the permissible level  $(C_p^o)$  and  $s_p$  is the geometric standard deviation (i.e. the standard deviation of  $\ln C_p$  values). If a field is contaminated homogeneously, equations (1) and (2) can be used directly. The geometric mean radionuclide activity concentration in a plant product is calculated from the geometric mean activity concentration of that radionuclide in soil using Eq. (1).

Equations for the case where the field is divided into different sections (for example where there is a gradient of contamination) are presented in Supplementary Information.

The risk of exceeding permissible levels of <sup>137</sup>Cs and <sup>90</sup>Sr in foodstuffs which could be produced on the field was estimated using this approach and geometric mean values of concentration ratios for the main crops as well as their variabilities (GSD) (Table 3). Table 3 only presents data for <sup>137</sup>Cs and <sup>90</sup>Sr since it will be shown that in the ZoR (Zone 2), these are the limiting radionuclides for crop production.

The area under assessment is considered suitable for the production of a crop (Labunska et al., 2018) if:

$$B + 0.6 \Delta B \le 1.0 \tag{3}$$

where  $B = \frac{C_p^{Cs}}{PL_{cs}} + \frac{C_p^{Sr}}{PL_{Sr}}$  is the factor determining compliance or noncompliance with the regulatory limit (*PL*) and

$$\Delta B = 1.1 \cdot \sqrt{\left(\frac{\Delta C_p^{Cs}}{PL_{Cs}}\right)^2 + \left(\frac{\Delta C_p^{Sr}}{PL_{Sr}}\right)^2} \tag{4}$$

where  $\Delta C_p^{RN}$  is the 95 % uncertainty of the predicted  $C_p$  value for the particular radionuclide. The 95 % uncertainty level was chosen to give a high probability that limits will not be exceeded. It is noted that the protocol includes a validation step (checking of crop activity concentration after first harvest and comparison with predicted value) to further ensure that crops do not exceed the limit for human consumption. Radionuclides other than <sup>137</sup>Cs and <sup>90</sup>Sr were not considered because their contribution to crop contamination is negligible in the regions outside the CEZ studied here (Ukrainian Institute of Agricultural Radiology, unpublished results) and their content in products is not regulated in Ukraine.

Using data on transfer factors, it is possible to estimate upper values of radionuclide contamination densities (Bq m<sup>-2</sup>) at which activity concentrations of <sup>137</sup>Cs and <sup>90</sup>Sr in crops will meet the above requirement (Eq. (3)) without taking into account the potential use of countermeasures. The upper levels of radioactive contamination of land correspond to the upper contaminations of land for the 3rd zone but limited for some crops where the exceedance level for crop contamination was above that of the 3rd zone limit as discussed below.

## 2.4. Exposure of farm workers

This radiological risk assessment is based on estimation of dose to a farmer/farm worker carrying out all soil preparation, crop spraying, seeding/planting and harvesting operations across a 100 ha farm, a reasonable farm size estimate for one farmer/farm worker. They are therefore relevant to commercial mechanised farm operations and not to self-production.

A Monte Carlo approach (1000 model runs) was taken to evaluate external dose rate variability using estimated parameter ranges and distributions. It was assumed that field operations (soil preparation, seeding, spraying and harvesting) took 4.6 h ha<sup>-1</sup>, a conservative estimate based on UK data (Williams et al., 2006), but reflecting potential use of smaller farm machinery than is typical in the UK. Potential variation was assumed to be a factor of 1.5 above or below this value, with a uniform distribution between upper and lower limits. External exposure is estimated for a soil activity-depth profile corresponding to a mixed (ploughed) depth of 0.25 m, a soil density of 1400 kg m<sup>-3</sup>, the maximum allowable <sup>137</sup>Cs contamination density of 555 kBq m<sup>-2</sup>) and isotope ratio data from the Narodychi District. Variability in the external dose coefficient was assumed based on variation in measured values. Inhalation dose coefficients were taken from (IAEA, 2004). Parameters used and assumed probability distributions are summarised in Table 4.

#### 3. Results

## 3.1. Site gamma survey

The results of the gamma dose rate survey (Fig. 4) indicated the presence of a north west to south east gamma dose gradient within the field. The values of dose rate varied from 0.11  $\mu$ Sv h<sup>-1</sup> in the northern part of the area to 0.35  $\mu$ Sv h<sup>-1</sup> in the southern part.

#### Table 4

cab)

Parameter values and distributions for Monte Carlo estimation of external and inhalation effective dose rates. Ratios of Sr, Pu, Am to<sup>137</sup>Cs are representative of the Narodychi region and would be higher at higher contamination densities and closer to the Chornobyl NPP.

Parameter	Central estimate	Assumed variability	Notes
<sup>90</sup> Sr/ <sup>137</sup> Cs ratio	9.1 E-3	Uniform (6.9–11.3E-3)	This study
<sup>238</sup> Pu/ <sup>137</sup> Cs ratio	1.3 E-4	Uniform (9.6–16.4 E–5)	This study
<sup>239,240</sup> Pu/ <sup>137</sup> Cs ratio	6.3E-4	Uniform (3.9–8.7 E–4)	This study
<sup>241</sup> Pu/ <sup>239,240</sup> Pu ratio	1.67E+1	N/A	Ratio in fallout ( UNSCEAR, 2000a).
<sup>241</sup> Am/ <sup>137</sup> Cs ratio	2.9E-4	Uniform (2.0–3.8 E–4)	This study
Soil dry bulk density	1400  kg m <sup>-3</sup>	Normal (S.D. 200)	Range for clay soils $1100-1600 \text{ kg m}^{-3}$ and sandy soils $1300-1700 \text{ kg m}^{-3}$
Plough mixed depth	0.25 m	Uniform (0.2–0.3)	Typical range
$\begin{array}{l} \mbox{Ploughed field dose} \\ \mbox{for}^{137}\mbox{Cs} = 555 \ \mbox{kBq} \\ \mbox{m}^{-2} \end{array}$	$\begin{array}{l} 0.38 \ \mu Sv \\ h^{-1} \end{array}$	Uniform (0.24–0.6)	This study
Occupancy farm worker	4.6 h ha <sup>-1</sup> year <sup>-1</sup>	Uniform (3.1–6.9)	Williams et al. (2006)
Adult breathing rate (activity level "light")	0.86 m <sup>3</sup> h <sup>-1</sup>	Normal (S.D. 0.15)	Moya et al. (2011)
Inhalable dust soil tillage (open tractor	$\begin{array}{l} 2\times10^{-5} \\ \text{kg m}^{-3} \end{array}$	Uniform (5 $\times$ 10 <sup>-6</sup> -4 $\times$ 10 <sup>-5</sup> )	Arslan and Aybek (2012)

#### 3.2. Soil sampling results and interpolation of contamination density

The  $^{137}$ Cs contamination measured from the soil cores taken on 28 07 2018 within the test field varied from 100 to 490 kBq m $^{-2}$  with an average of 210 kBq m $^{-2}$  and with a standard deviation of 106 kBq m $^{-2}$ . Analysis of data showed that there was a linear relationship between gamma dose rates and contamination density by  $^{137}$ Cs (Fig. 5). The correlation was good enough to be used in estimation of areal contamination by  $^{137}$ Cs by co-kriging (Isaaks and Srivastava, 1989).

Co-kriging was used for interpolation (mapping and contouring) of  $^{137}$ Cs contamination within the field (Fig. 6) from the combination of soil cores and dose rate measurements. The regression method took advantage of the covariance between two regionalized variables that are related, and are appropriate when the main attribute of interest ( $^{137}$ Cs activity concentration in soil samples) is sparse, but related secondary information (gamma dose rates) is abundant. The geostatistical method yields more reliable models because it capitalizes on the strengths of both data types. This approach can significantly reduce the uncertainties associated with the variability of radioactive contamination within the field.

## 3.3. Radionuclides other than Cs-137

Activity concentrations of <sup>137</sup>Cs, <sup>90</sup>Sr and plutonium isotopes determined in soil samples are provided in Table 5. Radioactive contamination of the land comprises fallout both from global nuclear weapons testing (NWT) and Chornobyl. The most intensive series of the nuclear weapon tests in the atmosphere took place during 1954-1958 and 1961–1962 (Hardy et al., 1973). The relevant long-lived radionuclides from these nuclear tests were <sup>239+240</sup>Pu, <sup>241</sup>Pu, <sup>241</sup>Am, <sup>137</sup>Cs and <sup>90</sup>Sr. Surface depositions of <sup>238</sup>Pu originating from the weapon tests in the Northern Hemisphere were low in comparison with other plutonium isotopes: the average weapons <sup>238</sup>Pu to <sup>239+240</sup>Pu ratio is 0.017 (Perkins and Thomas, 1980). An accident with a US satellite in 1964 resulted in a 50 % increase in the global fallout of <sup>238</sup>Pu in the middle latitudes of the northern hemisphere (Hardy et al., 1973). Thus, the average global <sup>238</sup>Pu:<sup>239+240</sup>Pu ratio can be estimated as about 0.025. The Pu fallout from the Chornobyl accident had a<sup>238</sup>Pu:<sup>239+240</sup>Pu ratio of about 0.5 (Kashparov et al., 2022) so this allows estimation of the relative contributions of global and Chornobyl fallout in soil contamination. For example, if the activities of  $^{238}\mbox{Pu}$  and  $^{239+240}\mbox{Pu}$  in a soil sample are A<sub>Pu-238</sub> and A<sub>Pu-239+240</sub>, respectively, then the activity of <sup>239+240</sup>Pu originating from Chornobyl can be estimated as (2•A<sub>Pu-238</sub>), and the activity of  $^{239+240}$ Pu originating from the global fallout as (A<sub>Pu-239+240</sub> -2•A<sub>Pu-238</sub>).

The values of the radionuclide activity ratios in soil samples for the surveyed land and in the Chornobyl nuclear fuel are shown in Table 6. When surveying territories in the same region, the expected ratios between radionuclide activities in the soil may differ from those obtained in this work, but their order of magnitude is expected to be similar (Kashparov et al., 2003).

Surface contamination of the field by plutonium isotopes ( $^{238,239,240}$ Pu) varied from 88 to 265 Bq m<sup>-2</sup>, with an arithmetic mean of 156 Bq m<sup>-2</sup> and with a standard deviation of 57 Bq m<sup>-2</sup> (Table 5). Based on the ratio between plutonium isotopes for Chornobyl fallout, it can be concluded that approximately half of the plutonium contamination within the field originates from the global weapons test fallout. No significant correlations between activity concentration of Pu isotopes and other radionuclides measured in the soil were observed.

Surface contamination of the field by  ${}^{90}$ Sr varied from 1 to 3 kBq m<sup>-2</sup>, with an arithmetic mean of 1.8 kBq m<sup>-2</sup> and with a standard deviation of 0.5 kBq m<sup>-2</sup> (Table 5). The contamination densities of  ${}^{137}$ Cs and  ${}^{90}$ Sr were strongly correlated (Fig. 7). In addition to the soil samples collected within the studied field, the data used in Fig. 7 include measurements of samples collected in north-western Ukraine (during 1999–2000) (Romanchuk, 2015). The north-western region was



Fig. 4. Spatial distribution of gamma dose rate (D; interpolated data). Data was collected on 27 07 2018.



**Fig. 5.** Relationship between values of land surface contamination by  $^{137}$ Cs and gamma dose rate (the field had previously been ploughed to 25 cm). Error bars show 95 % measurement uncertainty.

contaminated by the same type of radioactive fallout from the Chornobyl accident as the study field (Romanchuk, 2015). Activities of <sup>137</sup>Cs and <sup>90</sup>Sr were decay corrected to 2018 for plotting on Fig. 7. The relationship between <sup>90</sup>Sr and <sup>137</sup>Cs (Fig. 7) can be used for rough estimation of surface contamination in abandoned land by <sup>90</sup>Sr across north-west Ukraine. The relationship is described by a linear function with the intercept of 0.9 kBq m<sup>-2</sup>, the intercept is in agreement with likely levels

of global weapons testing fallout of  $^{90}$ Sr in this area (UNSCEAR, 1982) of about 1 kBq m<sup>-2</sup>. Levels of  $^{90}$ Sr surface contamination from Chornobyl fallout are relatively low and beyond 80 km from the ChNPP they are comparable with global fallout. The  $^{90}$ Sr/ $^{137}$ Cs activity ratio for the studied field was approximately 0.01 (Table 6).

## 3.4. Predicted radionuclide activity concentrations in crops

Estimates show that the probability of exceeding the permissible concentration of  $^{90}$ Sr activity in crops grown in the study field is extremely low as is the predicted mean value of the  $^{90}$ Sr activity concentration in selected crops (Table 7). Thus, all crops grown in this field are expected to meet permissible levels for  $^{90}$ Sr contamination (including for milk and meat). As shown in Supplementary Information, when a pasture meets requirements on radionuclide contamination to produce milk below PLs it will meet requirements to produce meat.

For <sup>137</sup>Cs, the probabilities of exceeding the established standards in all crops are higher than those for <sup>90</sup>Sr due to higher level of the soil contamination with <sup>137</sup>Cs. Leguminous crops are the most critical crops, since their seeds are predicted to have the highest probability of exceeding permissible limits. The concentration ratio *Cr* is the most important parameter in determining exceedance probabilities with leguminous crops having the highest *Cr* (after accounting for fresh mass/ dry mass ratios) as seen in Table 3.

Soil activity concentrations were calculated which would, with 95 % probability, produce crops which were at the limit for a combination of  $^{90}$ Sr and  $^{137}$ Cs contamination (Eqs. (3) and (4)). By converting these soil



Fig. 6. Field surface contamination by  $^{137}$ Cs interpolated from dose rate measurements (Fig. 4) and based on the relationship between measured contamination per square meter and dose rate (Fig. 5) as of July 2018. The isolines show surface contamination in kBq m<sup>-2</sup>.

Contamination of the study field by	radionuclides of the fuel of	component of the (	Chornobyl radioactive i	fallout in 2018 (sampling	g depth 25 cm).
5 5		1	2	· · · ·	<i>J</i> 1 <i>/</i>

Sample No Specific activity, Bq kg <sup>-1</sup>			Density of contamination					
					Total	Proportion originating from Chornobyl	Total	Total
	<sup>239,240</sup> Pu	<sup>238</sup> Pu	<sup>137</sup> Cs	<sup>90</sup> Sr	$^{238\cdot 239,240}$ Pu, Bq m <sup>-2</sup>	$^{238\cdot 239,240}$ Pu, Bq m <sup>-2</sup>	$^{137}$ Cs, kBq m $^{-2}$	<sup>90</sup> Sr, kBq m <sup>-2</sup>
B5	$0.37\pm0.07^a$	$\textbf{0.08} \pm \textbf{0.03}$	$1150\pm70$	$\textbf{5.4} \pm \textbf{0.7}$	$141\pm24$	$73\pm27$	$370\pm20$	$1.7\pm0.2$
B6	$0.26\pm0.05$	$0.05\pm0.02$	$600\pm30$	$5.6\pm0.7$	$88 \pm 14$	$41 \pm 17$	$173\pm9$	$1.6\pm0.2$
В9	$0.39\pm0.11$	$\textbf{0.06} \pm \textbf{0.04}$	$430\pm30$	$\textbf{5.0} \pm \textbf{0.7}$	$158\pm40$	$65\pm39$	$126\pm8$	$1.7\pm0.2$
B11	$0.55\pm0.17$	$\textbf{0.20} \pm \textbf{0.08}$	$1050\pm80$	$\textbf{6.2} \pm \textbf{0.7}$	$265\pm 66$	$210\pm82$	$370\pm30$	$\textbf{2.2} \pm \textbf{0.2}$
B13	$0.26\pm0.05$	$\textbf{0.06} \pm \textbf{0.02}$	$610\pm40$		$90\pm16$	$54\pm20$	$170 \pm 10$	
B14	$\textbf{0.39} \pm \textbf{0.08}$	$0.08\pm0.03$	$460\pm30$	$\textbf{4.8} \pm \textbf{0.6}$	$148\pm27$	$80\pm30$	$142\pm9$	$1.5\pm0.2$
B16	$\textbf{0.42} \pm \textbf{0.09}$	$0.06\pm0.03$	$340\pm20$	$\textbf{3.9}\pm\textbf{0.6}$	$158\pm30$	$56\pm30$	$113\pm 6$	$1.3\pm0.2$
B17			$300\pm20$	$3.6\pm0.5$			$123\pm7$	$1.5\pm0.2$
B18			$520\pm30$	$\textbf{4.6} \pm \textbf{0.6}$			$205\pm12$	$1.8\pm0.2$
B19	$0.37 \pm 0.05$	$\textbf{0.09} \pm \textbf{0.02}$	$1150\pm70$	$\textbf{6.6} \pm \textbf{0.8}$	$199\pm2$	$122\pm31$	$490\pm30$	$\textbf{2.8} \pm \textbf{0.3}$
$\text{MEAN} \pm \text{SD}$					$156\pm57$	$88\pm55$	$228 \pm 133$	$1.8\pm0.5$

<sup>a</sup> Combined 95 % uncertainty range of measurement.

#### Table 6

Ratios of radionuclide activities in the soil of the surveyed area (Narodychi district) and their ratio in Chornobyl nuclear fuel at the time of the accident.

Activity ratio	Ν	Soil $\pm$ S.E.	Nuclear fuel <sup>a</sup>
<sup>90</sup> Sr: <sup>137</sup> Cs	9	$0.0091 \pm 0.0011$	0.81
<sup>238</sup> Pu: <sup>137</sup> Cs	8	$1.3~(\pm 0.17)  imes 10^{-4}$	$92.1  imes 10^{-4}$
<sup>239+240</sup> Pu: <sup>137</sup> Cs	8	$6.3~(\pm 1.2)  imes 10^{-4}$	$190  imes 10^{-4}$
<sup>241</sup> Am: <sup>137</sup> Cs	3	$2.9~(\pm 0.44)  imes 10^{-4}$	$375\times 10^{-4}$

<sup>a</sup> corrected to 2018.



**Fig. 7.** The correlation relationship between <sup>137</sup>Cs and <sup>90</sup>Sr surface contamination densities (this study; (Romanchuk, 2015); in western regions of Ukraine, which were contaminated by the condensed component of the Chornobyl radioactive fallout decay corrected to 2018. Error bars show 95 % measurement uncertainty.

activity concentrations to contamination density per unit area, threshold contamination densities were estimated for different crops (Table 8). The conversion from soil activity concentrations (Bq kg<sup>-1</sup>) to density per unit area (Bq m<sup>-2</sup>) was performed assuming a soil density, depth of contamination and <sup>90</sup>Sr:<sup>137</sup>Cs ratio from Table 4. A field can typically be used in agriculture for crops and forage production if below the appropriate radionuclide contamination density shown in Table 8. Some crops could be produced at below permissible levels for <sup>137</sup>Cs if the soil contamination is greater than 555 kBq m<sup>-2</sup>, but the classification scheme (Table 1) stipulates that land above 555 kBq m<sup>-2</sup> of <sup>137</sup>Cs must be classified as Zone 2 (where no agricultural activity is allowed). Although some crop types could potentially safely be grown in areas of >555 kBq m<sup>-2</sup> of <sup>137</sup>Cs (Table 8), the Zone 2 classification would disallow this. Crop selection for re-use of assessed fields can be made

#### Table 7

Predictions of <sup>137</sup>Cs and <sup>90</sup>Sr geometric mean activity concentrations in regionally important crops ( $\overline{C_p}$ ) if grown in the study field (as they go to market – in fresh or air dry form) and probabilities (q, to 3 d.p.) of exceeding the permissible levels ( $C_p^0$ ). For example, a q value of 0.02 for exceeding the permissible level for<sup>137</sup>Cs in maize represents a 2 % probability of exceedance of the PL. The grass and pasture limits are based on permissible levels in milk of 100 and 20 Bq kg<sup>-1</sup> for<sup>137</sup>Cs and <sup>90</sup>Sr respectively (see Supplementary Information).

Crop	<sup>137</sup> Cs			<sup>90</sup> Sr		
	$C_p^0$ , Bq kg <sup>-1</sup>	C <sub>p</sub> , Bq kg <sup>-1</sup>	q	$C_p^0$ , Bq kg <sup>-1</sup>	C <sub>p</sub> , Bq kg <sup>-1</sup>	q
Potatoes	60	9.0	0.002	20	0.3	< 0.001
Leafy vegetables (salad)	40	6.5	0.098	20	0.8	0.005
Root crops (red beet)	40	6.7	0.019	20	0.7	0.005
Leguminous crops (soya seeds)	50	24.8	0.166	30	3.7	0.001
Maize (grain)	50	10.4	0.020	20	0.2	0.000
Cereals (grain)	50	11.8	0.051	20	2.5	0.006
Sunflower (seed)	70	22.6	0.000	10	1.6	0.000
Grass (hay)	2400	22.6	0.000	3400	7.7	0.000
Pasture	500	189	0.119	700	6.0	0.000

using the probabilities of exceedance of permissible limits (PL) as shown in Table 7 using site specific contamination densities and  $^{90}$ Sr:<sup>137</sup>Cs ratios.

The radionuclide contamination densities in Table 8 were obtained using available data on radionuclide transfer factors (Table 3) for different crops growing on Podzoluvisols, which are a good representation of the key soil types in this area. The soil contamination densities in Table 8 equate to those at which crops or forage (or subsequent animal derived food products) will, with high probability, be below current Ukrainian permissible levels.

### 3.5. Risk assessment for farm workers

Effective equivalent doses from external, inhalation and inadvertent ingestion of soil were calculated using the Monte Carlo (MC) model to estimate uncertainty (Fig. 8). For comparison, doses were also calculated using the RESRAD (Yu et al., 2007) and NORMALYSA (IAEA, 2023) software. Mean external dose at 555 kBq m<sup>-2</sup> of <sup>137</sup>Cs was 74 µSv y<sup>-1</sup> in both of these models and inhalation dose was 0.045 and 0.13 µSv y<sup>-1</sup> respectively. These are broadly consistent with doses estimated in the MC model (median: 200 µSv y<sup>-1</sup> external, 0.90 µSv y<sup>-1</sup> inhalation), though lower due to a lower ploughed field soil-external dose rate coefficient assumed in these models compared to the MC model.

Whilst external and inhalation pathways likely represent the most

Illustrative soil contamination densities of <sup>137</sup>Cs and <sup>90</sup>Sr for different crops and forage products based on a 95 % probability that permissible levels of these radionuclides will not be exceeded. The central 50th percentile estimate (i.e. based on mean values of parameters) is also shown. The values are only illustrative as they depend on site specific<sup>90</sup>Sr:<sup>137</sup>Cs ratios<sup>a</sup>. In practice, <sup>137</sup>Cs is the limiting radionuclide as (depending on crop type) it makes a between 2 × and  $26 \times$  greater contribution to exceedance of PL than<sup>90</sup>Sr. Note also that in practice, the maximum<sup>137</sup>Cs contamination level is limited by the regulatory criterion for Zone 2: 555 kBq m<sup>-2</sup>.

Crop	<sup>137</sup> Cs, kBq m <sup>-2</sup>		<sup>90</sup> Sr, kBq m <sup>-2</sup>	
	50 %	95 %	50 %	95 %
Potatoes	1350	570	12	5.1
Leafy Vegetables	890	122	8.1	1.1
Root crops (Red Beet)	850	210	7.7	1.9
Legumes (Soya Bean)	250	89	2.3	0.81
Maize (grain)	1120	360	10	3.3
Cereals (grain)	560	160	5.1	1.5
Sunflower (seeds)	480	260	4.4	2.4
Grass (hay) <sup>b</sup>	20000	8700	180	80
Grass (pasture)	330	100	3.0	0.9

 $^{\rm a}\,$  The values given here assume a  $^{90}{\rm Sr}{:}^{137}{\rm Cs}$  ratio between 6.9  $\times$  10  $^{-3}$  and 11.3  $\times$  10  $^{-3}.$ 

<sup>b</sup> Limit is very high because only a small fraction of cattle feed is typically hay. The limiting activity concentration would be lower if a greater proportion of feed was hay.

significant doses to an agricultural worker, other dose pathways have also been considered (Supplementary Information). Exposures to agricultural or process workers were also evaluated from.

- skin contact with contaminated soil;
- inadvertent ingestion of soil;
- dust inhalation when handling crops.

These dose pathways resulted in an effective equivalent dose of order 1  $\mu$ Sv y<sup>-1</sup> from inhalation and ingestion and 0.1 mSv y<sup>-1</sup> skin dose from contact with soil. These do not add significantly to external exposure (see Supplementary Information) and are much less than the (ICRP, 2007) recommended dose limit for exposures to the skin of 50 mSv y<sup>-1</sup> to members of the public.

## 4. Discussion

The study has presented a method for evaluating radionuclide transfers to crops and the suitability of land for return to agricultural use. As shown in Table 9, the mean contamination density of the Mezheliska field plot studied is within criteria for Zone 3. Based on the <sup>137</sup>Cs contamination density of the whole field, the land could be used for growing all crops except leguminous vegetables (see Table 8) without the application of countermeasures. Alternatively, due to the zonal nature of the contamination, it was found that the northern part of the field could alternatively be reclassified as being out of the radioactively contaminated (and hence restricted use) zones and could be completely returned to economic use. The southern part of the field (contamination up to 555 kBq m<sup>-2</sup>) could be reclassified as Zone 3 and would require restrictions on the type of crop which could be grown. Based on the zoning criteria in Ukrainian legislation, the surveyed field does not qualify as a radioactive zone based on <sup>90</sup>Sr and plutonium contamination densities (Table 9).

It should be noted that this study assumes farming practices typical of Ukraine. In general, it is expected that other farming practices (e.g. greater use of mineral fertilisers, in particular potassium) would decrease <sup>137</sup>Cs concentration ratios. Potential increase in concentration ratios due to different farming practices cannot be completely excluded, but we think this unlikely as fertilisation levels are currently generally low in this region.

Given the complex nature of the radioactive contamination of the region, it can be concluded that <sup>137</sup>Cs cannot be used as an accurate tracer for the transuranic elements (TUE) contamination. Nevertheless, as shown in Tables 5 and 6, the level of contamination of this area by TUE outside the CEZ in the west direction is rather low and approximately the same as the contribution of global weapons test fallout and Chornobyl components to the total contamination by these radionuclides. For this reason, there is no significant issue with hot particle contamination at this distance from the Chernobyl site – areas significantly contaminated with hot particles have much higher TUE:<sup>137</sup>Cs ratios than those observed here.

The correlation between <sup>137</sup>Cs and <sup>90</sup>Sr in soil that has been observed (see Fig. 7) can only be used to provide a rough estimate of <sup>90</sup>Sr contamination since, even outside the CEZ, <sup>90</sup>Sr to <sup>137</sup>Cs ratios can vary (e.g. (Labunska et al., 2018)). Estimation of the level of <sup>90</sup>Sr contamination of land being returned to use should be based on monitoring, so it is recommended that sampling and measurement of <sup>90</sup>Sr is carried out to determine this ratio in each field plot.

## 4.1. Radiation protection principles and ethical issues

The approach used here to evaluate the study field is based on internationally accepted principles of radiation protection developed by the International Commission for Radiological Protection (ICRP) (ICRP, 2007, 2009). Robust estimates of activity concentrations and potential doses are essential to stakeholder dialogue on contaminated land issues. However, radiological assessment is only one part of the decision-making process for change of use of contaminated land (ICRP, 2009). A decision to change an existing exposure situation must be based on dialogue between all stakeholders that have an interest in, or are potentially affected by the proposed change (Lochard, 2013; Rollinger et al., 2016). Ethical aspects of countermeasures and change in use of abandoned lands have been discussed by, for example, (IAEA, 2022; Oughton, 2016; Oughton et al., 2004). Such issues include (in addition to dose optimisation and justification), minimisation of detriments, respect for the dignity and integrity of affected stakeholders, and a fair distribution of benefits from the change.

It is also noted that there are limited radiological benefits of further reductions in doses of order 1 mSv  $y^{-1}$  or less. Thus, countermeasure implementation (either restrictions or physical countermeasures such as intensive fertilisation) should be based primarily on social impact considerations. For example, countermeasures may be implemented to provide reassurance even when doses are very low. On the other hand, interventions which are not strictly necessary from a radiological perspective can also give the impression of significant risk where no such risk is present. This societal context is particularly complex in less developed countries where expenditure on radiological protection, or controls on "contaminated" land may (where doses are low) divert resources from more important health, safety and economic issues.

#### 4.2. Dose to farm workers

The recommended occupational exposure limit for non-classified workers is up to 6 mSv per year effective dose in the UK and up to 1 mSv per year in Ukraine. Doses to farm workers using the MC model presented here (and two other models) were dominated by external exposure. Estimated doses (assuming 555 kBq m<sup>-2 137</sup>Cs) were broadly consistent between the different models and were all significantly lower than 1 mSv y<sup>-1</sup>. Hence, it is pertinent here to consider issues relevant to dose rate reduction at < 1 mSv annual Chornobyl-derived dose. At this additional dose rate, the majority of a person's total annual dose is from natural background radiation so social and economic factors are of greater consequence in consideration of remediation measures.



**Fig. 8.** A: External dose for farm worker cultivating 100 ha of land estimated at  $^{137}$ Cs density of 555 kBq m<sup>-2</sup> compared with range in annual external dose rates worldwide from naturally occurring terrestrial gamma emitters (UNSCEAR, 2000a) assuming a conversion from absorbed dose in air to external effective dose of 0.7 Sv Gy<sup>-1</sup> (UNSCEAR, 2000b) and cosmic radiation (Bouville and Lowder, 1988). Natural terrestrial external dose rates in Northern Ukraine are at the lower end of this range REF. **B:** Inhalation dose from all Chornobyl radionuclides for farm worker at  $^{137}$ Cs density 555 kBq m<sup>-2</sup>. The illustrative range of effective equivalent dose rates worldwide from inhalation of natural alpha-emitting radionuclides is also shown (based on data in (Appleton, 2007; Dubois, 2005)).

Comparison of radionuclide contamination density (with Standard Deviation S. D. and Standard Error S.E.) at Mezheliska field plot with Zone 2 and Zone 3 Criteria.

Radionuclide	Mean (S.D.; S.E.) kBq m <sup>-2</sup>	Zone 2 Criterion kBq m <sup>-2</sup>	Zone 3 Criterion kBq m <sup>-2</sup>
<sup>137</sup> Cs	210 (116; 25)	>555	185–555
<sup>90</sup> Sr	1.8 (0.46; 0.15)	>111	5.55–111
Pu (Total	0.087 (0.055;	>3.7	0.37-3.7
Chornobyl)	0.02)		

# 4.3. Validation

Following the field radionuclide characterisation, sunflower seeds were harvested from the Mezheliska study plot. Their mean activity concentration in 2019 was  $35 \pm 3$  Bq kg<sup>-1</sup> d.w. of <sup>137</sup>Cs compared to the model predicted activity concentration of 23 Bq kg<sup>-1</sup>. For <sup>90</sup>Sr the mean activity concentration was  $1.5 \pm 0.3$  Bq kg<sup>-1</sup> d.w. compared to the model predicted value of 1.6 Bq kg<sup>-1</sup>. Measured values were significantly below the permissible level of 70 Bq kg<sup>-1</sup> for <sup>137</sup>Cs and 10 Bq kg<sup>-1</sup> d.w. for <sup>90</sup>Sr

#### 5. Conclusion

This evaluation of soil-crop transfer of contaminated land has shown that large areas of land in Zone 2 ("The Zone of Obligatory Resettlement") could potentially be de-restricted and used for agriculture. The key determinant of land use is the <sup>137</sup>Cs contamination density. Strontium-90 has higher soil-crop transfer factors but is at much lower deposition densities than <sup>137</sup>Cs so is unlikely to be a limiting factor in reuse of land in this area. It is noted, however, that <sup>137</sup>Cs:<sup>90</sup>Sr ratios are higher nearer to the Chernobyl site. Transuranium radionuclides are at very low levels in Zone 2 and therefore are not significant in dose formation compared to <sup>137</sup>Cs. The key factors in determining re-use of land and potential crop type are therefore the <sup>137</sup>Cs deposition density and the soil-plant concentration ratio.

The pilot field site could be used to produce crops which are below permissible levels for consumption, with some restrictions (at least for part of the field) on which crops could be grown. Doses to agricultural workers are very small compared with variation in natural radiation doses worldwide and are significantly below 1 mSv y<sup>-1</sup>. Large areas of land in Northern Ukraine currently in Zone 2, the Zone of Unconditional (Obligatory) Resettlement (see "Compulsory Relocation Zone" in Fig. 1) are at similar or lower contamination densities to our study plot and so, based on this evaluation, could potentially officially be brought back

#### into production.

Based on this study, we recommend the following protocol for evaluation of radioactively contaminated land in Northern Ukraine (where agricultural production is currently not allowed) for potential reuse. The protocol consists of five key steps.

- 1. External gamma dose rate survey and mapping of plot;
- 2. Soil sampling to determine local ratios of <sup>137</sup>Cs to <sup>90</sup>Sr and use of generic ratios (Table 6) for transuranium radionuclide estimation;
- 3. Determination of crops which can be grown based on high confidence that permissible limits will not be exceeded (see Table 7 for example)
- 4. Comparison of surface contamination with zone criteria presented in Table 9
- 5. Validation of crop activity concentrations after the first harvest. This could alternatively be done on a small experimental sub-plot prior to planting the whole field.

Based on the experience of this study, fieldwork required for characterisation of an approximately 50 ha plot requires about 12 personhours. The field size over which activity concentrations should be averaged should be no greater than 50 ha, where a "field" is defined, for this purpose, as an area of land used to grow a single crop type. The maximum field size of 50 ha is based on the observed scale of spatial variation of radioactivity in distant zones from Chornobyl. If, subsequent to evaluation using this protocol, a field is sub-divided into different crop types, then the individual sub-plots should be reevaluated. This re-evaluation may either use the initial contamination density mapping or re-measurement to ensure that the average contamination density in each sub-plot is below the limits set out in this protocol for the allowable crop types.

Given the relatively wide variation in plant-soil concentration ratios for a given crop species, the approach presented here is likely to overestimate plant activity concentrations, since decisions on restrictions for crops are based on the 95 percentile predicted value (ie including uncertainty in  $C_r$ ). It may be possible to reduce this conservatism using the observation that plant-soil concentration ratios are correlated between species for a given soil type (Willey, 2014). In a field where crop type is restricted, measurement (rather than model estimation) of  $C_r$  for a crop which is possible to grow may allow more accurate estimation of  $C_r$  for other crops and thus may allow removal of restrictions for that particular field. This should be considered in any further development of the protocol presented here.

As an example of contaminated land evaluation for derestriction, the methods developed here could be applied to other radioactively contaminated sites worldwide. The method could potentially be adapted for other soil types using soil-plant transfer databases, by evaluation of relative soil radionuclide absorption ( $K_d$ ), or by  $C_r$  extrapolation methods discussed above.

#### CRediT authorship contribution statement

J.T. Smith: Writing – review & editing, Writing – original draft, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Conceptualization. S.E. Levchuk: Writing – review & editing, Writing – original draft, Visualization, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. D.A. Bugai: Writing – review & editing, Investigation. N.A. Beresford: Writing – review & editing, Investigation. N.A. Beresford: Writing – review & editing. M.D. Wood: Writing – review & editing. Khomutinin Yu: Methodology. G.V. Laptev: Writing – review & editing. V.A. Kashparov: Writing – review & editing, Writing – original draft, Methodology, Formal analysis, Conceptualization.

## Declaration of competing interest

relationships which may be considered as potential competing interests: J T Smith reports a relationship with UK Department of Health Committee on Medical Aspects of Radiation in the Environment that includes: travel reimbursement. J.T. Smith and G.V. Laptev are Directors of the not-for-profit Chernobyl Spirit Community Interest Company which produces and sells spirits made from crops grown in radioactively contaminated lands in Ukraine. Profits support communities in the affected areas. If there are other authors, they 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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# Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jenvrad.2025.107698.

## Data availability

We have uploaded data with the MS files

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