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Optimising sampling strategies for emergency response: Vegetation sampling

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ABSTRACT

The lessons learned from the plant sampling campaigns implemented in the most Chernobyl affected countries are described. The variability of ^{137}Cs activity concentrations in plants taken from a variety of sampling sites, as well as the uncertainties around the aggregated transfer factors (T_{ag}) from soil to plants were estimated. The sampling sites covered both agricultural and natural lands in different landscapes: floodplain, plains, and watershed meadows. To determine parameters of the lognormal distribution of the ^{137}Cs activity concentration in plants and the values of corresponding aggregated transfer factor (T_{ag}) values, from 25 to 49 plant and soil samples were collected at each sampling site with the grid increment that varied from 1 to 10 m. The gradients of deposition i.e. monotonic changes (trends) of the contamination density conditioned by the global (in respect to study area) gradient of fallout were not observed in any of the study sites. Therefore, the variability of radionuclide contamination density (and activity concentrations in the soil) within the study sites were determined by only random factors such as microheterogeneity of radioactive deposition in a sampling point. The mean standard deviation of the logarithms of ^{137}Cs activity concentrations in plants sampled in all such sites and the corresponding transfer factors were similar for all sites studied and were not dependent on the mean soil contamination density at the site, the type of radioactive fallout and the vegetation type. The values of the average standard deviation of the ^{137}Cs activity concentration logarithms in plants and the corresponding transfer factors for the vegetation sampling area $\geq$1 m^2 and the relative activity measurement uncertainties $\leq$10% were estimated as 0.4 ± 0.1 and 0.5 ± 0.1, respectively. A new simple method for optimization of the number of linked (conjugated) plant and soil samples as well as estimates of the activity concentration and transfer factor uncertainties when measuring composite samples were proposed. Based on the results of these studies, the recommendations were made to the sampling of plants for radionuclides.

1. Introduction

The deposition of radionuclides onto the soil and vegetation is the first step for further radionuclide transfer in the terrestrial environment, including to the food chain. After initial contamination of the environment, the radionuclides are distributed between the soil and plant surface. The fraction of radionuclide associated with plants depends on time lapsed after the emergency, type of depositions (dry or wet), aboveground biomass, leaf area index (LAI), climatic conditions etc. (IAEA, 2009;Neil Cape, 2009; Henner et al., 2013; Hurtevent et al., 2013; Holiaka et al., 2017).

Interception fraction (i.e. the radionuclide fraction initially retained by plants), weathering (the rate of radionuclide loses from plants due to removal by wind, rain, leaching and cuticular abrasion) and translocation are the main processes determining activity concentrations of radionuclides in the vegetation during the first year after the deposition. In the case of the Chernobyl deposition, effective weathering half-lives ($T_{1/2}^{Eff}$) (i.e. half-life that takes radioactive decay into account) ranged from 4 to 7 days for ^{131}I and from 8 to 11 days for caesium isotopes (^{134,137}Cs), respectively (IAEA, 2010).

After a relatively short period of dominating foliar uptake (a few months after the accident), root uptake becomes the major pathway for the plant contamination. The extent of the root uptake is dependent on

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the physico-chemical properties of the radionuclide; the physico-chemical characteristics of the soil environment; type of crop, crop management practices (application of fertilizers, irrigation, ploughing, liming, etc.) and climate conditions (Sanzharova et al., 1994, 2009; IAEA, 2006; Kashparov et al., 2005a,b, 2007a,b, 2018). The presence of plants may result in formation of local heterogeneities of the depositions, because of plant differences in radionuclide interception and weathering.

The coefficients of variation of ^{137}Cs activity concentrations in the samples of plants (i.e. the ratio of the standard deviation to the mean) are often used for assessment of the overall uncertainty of the contamination of agricultural plants, trees and shrubs and natural wild grasses. It was shown that for vegetation samples taken from the relatively small sites (465–3600 m^2) this parameter can vary from 0.12 to 0.20 (McGee et al., 1995; ICRU, 2006). It was also found, that the coefficient of variation of ^{137}Cs activity concentrations in graminaceous vegetation and non-graminaceous species (*Calluna vulgaris*) near sheep farms varied from 0.09 to 0.63 (Beresford et al., 2007) and for ^{90}Sr in natural wild grasses of alpine pasture up to 0.59 (Schimmack et al., 2003).

The permissible levels (or food safety standards) of radionuclides in foodstuffs, including grains, potatoes, root vegetables, vegetables, medicinal plants, fruits and berries were introduced shortly after the Chernobyl accident to reduce radiation exposure to the population (Balonov et al., 2018). In some Chernobyl affected areas, the ^{137}Cs activity concentrations in milk from the cows grazing natural fodder lands are exceeding the permissible levels (Labunska et al., 2018). This shows that an accurate estimate of the statistical characteristics of ^{137}Cs concentrations in natural grass to optimize implementation of remedial actions is needed (Fesenko et al., 2001, 2013; Jacob et al., 2009; Ulanovsky et al., 2011). Thus, a proper justification of the number of vegetation samples required to maintain the accuracy of the assessment, minimizing associated cost and other resources is important. (Alexakhin et al., 1996; Theocharopoulos et al., 2001).

Concentration ratio (CR[1]) and aggregated transfer factors (T_{ag}) are basic parameters used for predicting the plant contamination (IAEA, 2009, 2010). Concentration ratio is defined by the ratio of the activity concentration of radionuclide in the plant (Bq·kg^{-1}, dry mass) to that in the soil (Bq·kg^{-1}, dry mass) and T_{ag} is the radionuclide activity concentration (Bq/kg, dry mass) in the plant per unit area activity density (kBq/m^2) in the soil.

The concept of transfer factors includes additional uncertainties due to the assumption that all roots and activity are presented in standardised rooting zones, therefore aggregated transfer factors used in this study relate to radionuclide activity inventory within the top 20 cm of soil.

The paper provides outputs of the sampling programs performed from 1990 to 2018 by the Ukrainian Institute of Agricultural Radiology (UIAR) and some other sister institutions in areas of Ukraine contaminated by the fallout from the Chernobyl accident, including the Chernobyl Exclusion Zone (ChEZ). The aim of this study was to systematically investigate the spatial variability of ^{137}Cs activity concentrations in vegetation growing in cultivated fields and in natural environments. Various factors, such as the species of plants, properties of agricultural lands, number of samples, size of samples, and spatial arrangement of the samples were examined to improve the efficiency of the vegetation sampling and optimize the sampling planning (Khomutinin et al., 2001). From the spatial variability observed, the minimum number of plant samples was estimated which is necessary to obtain the mean activity concentration of ^{137}Cs in vegetation with a given accuracy.

[1] CR (concentration ratio) used as synonym of the transfer factors B_v.

2. Material and methods

The sampling design used for vegetation sampling was similar to that implemented for soil sampling (Khomutinin et al., 2020). Thus, the area of the sampling units ranged 6 × 6–60 × 60 m and the variation of the contamination density because of the gradient of the deposition (systematic change along gradient of deposition) at each site was lower than variations of radionuclide concentrations in the soil due to the effects of random factors.

The contamination density measured for any sampling point can be presented as a sum of three major components (Khomutinin et al., 2020):

- Systematic component, i.e. fraction of the deposition that follows a monotonic change (trend) of the contamination conditioned by the global (in respect to study area) gradient of fallout;
- Spot component that accounts for localities where values of density of contamination are statistically significantly different from the trend;
- Random component that accounts for the effects of:
  - o microheterogeneity of radioactive deposition in a sampling point,
  - o the sample collection method and handling procedures and,
  - o the analytical procedure applied for measurement of radionuclide activity concentration.

For the purpose of the study the boundaries of sites were chosen to exclude effects of systematic and spot components; such sites are termed here as sampling units (or sampling sites).

In case of environmental contamination by γ-emitting radionuclides (such as radiocaesium), the subdivision of the surveyed territory to the sampling unit can be done based on the measurements of the ambient dose rate. This method was widely used in the survey of the territory of settlements, forests and agricultural lands affected by the Chernobyl accident. The 30-50%-deviations of the measured dose rates from the mean value was taken as a threshold level to identify such sampling units for further sampling of soil and vegetation.

Individual (single) vegetation samples taken within the sampling site are often merged in one composite sample for further measurement of radionuclides activity. The activity of radionuclides in such a pooled sample is normally considered as a mean radionuclide concentration in vegetation at the sampling site and the measurement error is the only parameter used to characterize the overall uncertainty of the radionuclides in vegetation at the sampling site. Our study was aimed to estimate total variability of radionuclides accumulation in plants growing in the same site and the radionuclide activity concentrations were measured in each collected sample. These data were further used to identify a probability distribution of radionuclide concentrations in the vegetation samples and to estimate mean values, standard deviation, and other required statistical parameters. Such an approach can provide estimating the total uncertainty of radionuclide activity concentration in vegetation growing in the surveyed field and improve the efficiency of the vegetation sampling.

Field sampling of vegetation was conducted at the sites (Table 1), which were previously used to investigate the spatial variability of soil contamination density (Khomutinin et al., 2020). The study areas covered both agricultural and natural lands of different landscape types: floodplain, plains and watershed meadows.

2.1. Sites description

The sampling units were selected in the fields with the crops which were typical for the contaminated part of the Ukrainian Polessie and on the abandoned former agricultural fields with typical wild (natural) vegetation (Table 1). Plant species included basic crops of the Chernobyl affected areas: rye (*Secale cereal* L.), oat (*Avena sativa* L.) and winter

Table 1
Major characteristics of sampling sites with grassy vegetation, geometric mean (GM) and geometric standard deviation (GSD) of ^{137}Cs activity concentration, T_{ag} and the Pearson pair correlation coefficient between ^{137}Cs activity concentrations in conjugated samples of plants and soils contamination density at sampling units.

Site #	Year	Latitude N°	Longitude E°	Type of plant	Increment[a], m	Unit size, m	Quantity of samples	^{137}Cs activity concentration (DM), Bq·kg^{-1}		^{137}Cs T_{ag} (DM), (Bq·kg^{-1})·(kBq·m^{-2})$^{-1}$		Correlation coefficient
								GM	GSD	GM	GSD	
Cultivated plants on arable land												
S9	1999	51.340	30.126	Rye2	X1 = 10	60 × 60	49	275	1.57	0.19	1.69	+0.04
					X2 = 2	10 × 8	30	268	1.47	0.29	1.74	
S13	1999	51.153	30.353	Oat[b]	X1 = 10	60 × 60	49	92	1.38	0.81	1.64	−0.04
					X2 = 2	10 × 8	30	57	1.31	0.58	1.33	
S17	1999	51.182	29.492	Winter wheat[b]	X1 = 10	60 × 60	49	497	1.49	0.66	1.55	−0.13
S3	1999	51.377	30.036	Couch grass	X1 = 2	12 × 12	49	87054	1.86	2.17	2.9	−0.11
Natural vegetation on abandoned agricultural fields												
S7	1999	51.341	30.103	Bushgrass	X1 = 1	6 × 6	49	519	1.25	0.23	1.69	−0.16
S11	1999	51.264	30.096	Bushgrass	X1 = 2	8 × 8	25	245	1.46	0.60	1.77	−0.27
S15[c]	1999	51.303	29.642	Suncups	X1 = 10	60 × 60	49	2510	1.43	0.27	1.62	+0.13
					X2 = 2.5	10 × 10	25	3210	1.44	0.31	1.68	
S16[d]	1999	51.289	29.853	Bushgrass	X1 = 10	40 × 40	25	67	1.72	1.63	1.93	−0.06
S18	2003	50.632	32.619	Bushgrass	X1 = 10	50 × 40	30	7.9	1.46	2.88	1.48	+0.14
					X2 = 2.5	10 × 10	25	7.9	1.31			

[a] Distance between the grid nodes/sampling points.
[b] Stems and shoots.
[c] Former arable land.
[d] Meadow.

wheat (*Triticum* L.) and widely distributed wild vegetation:predominantly couch grass (*Elymus repens* L.), bush grass (*Calamagrostis epigejos* L.) and suncups (*Oenothera* L.).

The sampling sites were selected based on the external dose rate measurements, as described in Khomutinin et al. (2020). Thus, the gradients of deposition i.e. monotonic changes (trends) of the contamination density conditioned by the global (in respect to study area) gradient of fallout were not observed in any of the study sites. Therefore, the variability contamination density of radionuclide (activity concentrations in the soil) within the study sites were mainly determined by the random factors such as microheterogeneity of radioactive deposition in a sampling point.

One site (S18) was selected where the ^{137}Cs in the soil (at the deposition density of 2–3 kBq m^{-2}) was of global fallout origin. Most of the selected sites (S3, S 7, S11, S13, S15 and S16) were in the ChEZ and only sampling sites S17 and S18 were located outside of the zone (Table 1).

The sampling sites S7, S11 and S18 were located on fallow, S15 on old arable land, S16 on dry meadows, covered by natural grass with plants of 20–40 cm of height (Table 1). The agricultural vegetation sampling sites were also chosen on arable lands which were part of crop rotation and had been ploughed several times within experimental studies both inside of the ChEZ (S9 with rye and S13 with oat) and beyond it. Site S17, which followed traditional farming practices, was used for planting winter wheat (Table 1).

2.2. Sampling of vegetation

Sampling of vegetation and soil was carried out at a rectangular or square grid with different distance between the grid nodes (Table 1). Samples were collected with the various sampling grid resolutions. Thus, spatial increments (distances between the grid nodes) varied from 1 to 10 m. Quantity of conjugate (interconnected) plant-soil samples (i.e. soil and plant samples taken at the same point at the same time) varied from 25 to 49, depending on the sampling grid resolution applied for the sampling site. For agricultural plants only the green mass (stems and shoots) was sampled.

Plant samples were collected by cutting the biomass of chosen species of vegetation (Table 1) at each 1 × 1 m (1 m^2) sampling site at the height of 3–5 cm above ground to avoid contamination of samples by soil particles. Samples of soils at 20 cm depth were also taken at each sampling point to determine density of contamination (Khomutinin et al., 2020) and transfer factors evaluations. Conjugated samples of the soil were taken at the same point and time as samples of the vegetation (sampling area 1 m^2). The plant material was weighed, dried at 105 °C and milled.

2.3. Measurements

Dried vegetation samples were analysed for ^{137}Cs by a high-resolution gamma spectrometer in a range of counting geometries from 15 cm^3 containers to 1000 cm^3 Marinelli beakers. Analyses were carried out using high-purity germanium detectors (GEM-30185), equipped with a multichannel analyser (Model 919) and with a standard 10 cm lead shielding (EG&G Ortec, USA). Calibration of the spectrometer was conducted using a commercial certified standard (sawdust matrix multi-nuclides standard provided by V.G. Khlopin Radium Institute, Russia) and subsequently checked through participation in national and international inter-comparison exercises.

QC/QA 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 (661.6 keV), as well as analysis of internal control samples and external reference materials (CRM) using the QA option of the software. The relative total uncertainties of measurements (for 95% confidence levels) did not exceed 10% for the sites contaminated with Chernobyl deposition, and 25% for the sites where radionuclide contamination was mostly of a global fallout origin. Higher uncertainty in measuring ^{137}Cs in the vegetation sampled at the sites contaminated by the global fallout can be explained by low ^{137}Cs concentrations in the plants of these sites and a lack of possibility for long-term measurement of these samples.

All activity concentration of radionuclides in the soil and vegetation are given in Bq per weight of dry mass (DM).

2.4. Data analysis

Sampling planning and evaluation of the data on radionuclide activity concentrations in the soil-plant samples are often based on as-

sumptions concerning the probability distributions of the corresponding parameters. It is well known that in the Earth's crust, the concentration of elements (and their radioisotopes) often follows a lognormal distribution (Khomutinin et al., 2001; Malanca et al., 1996). The results of this study have shown that the variability of the radionuclide concentrations in the soil and the vegetation samples, terrestrial density of contamination, *CR* and TF_{ag} were asymmetrical, and could be fitted by a lognormal distribution (Stuart and Ord, 1987; Fesenko et al., 1993, 1997; Khomutinin et al., 2001, 2020). The probability density of the log-normal distribution can be presented as:

$$f(Q_v) = \frac{1}{Q_v \cdot s \cdot \sqrt{2\pi}} \exp\left(-\frac{1}{2}\left[\frac{\ln(Q_v) - \mu}{s}\right]^2 \right) \quad (1)$$

where Q_v is the radionuclide activity concentration in the vegetation; μ is the mean of natural logarithm of the radionuclide activity concentration and s is the standard deviation of the logarithms of the radionuclide activity concentrations. The median of radionuclide activity concentration, that is equal for log-normal distribution to the geometric mean (GM), was defined as:

$$M_C = \exp(\mu) \quad (2)$$

The log-normal distribution was defined as a distribution where the logarithms of the values and not the values themselves are normally distributed. Another approach that could be used to describe the central tendency of the values of interest can be based on the application of the geometric mean (GM) and geometric standard deviation (GSD) where GSD is the exponent of standard deviation of the natural logarithms of the individual values of the radionuclide activity concentration in vegetation.

3. Results and discussion

3.1. Distribution of radionuclide activity concentrations in vegetation

Typical spatial distributions of ^{137}Cs activity concentrations in the vegetation taken at the sampling sites are shown in Fig. 1. Variability of the radionuclide activity concentrations in the vegetation samples originated from the spatial inhomogeneity of radionuclide concentrations in the top soil, physico-chemical form of radionuclides in the fallout and soil properties - i.e. parameters that are out of the control. This uncertainty was merged with the measurement errors.

Overall, the data of Fig. 1 allow consideration of the activity concentrations of ^{137}Cs in vegetation samples taken at the sampling sites (Q_v) as a random positive value with a significant scattering and a right-sided asymmetry (Fig. 1).

The distribution of this random value corresponding to concentrations of ^{137}Cs in vegetation samples, obeys the log-normal law of probability distribution (see Eq. (1)) (Albers et al., 1998, 2000; Khomutinin et al., 2001; Schimmack et al., 2003). The dispersion of logarithms of the radionuclide activity concentrations in the vegetation did not depend on time (on the decay of radionuclides). Therefore, the standard deviation of the logarithms of the radionuclide activity concentrations in plants calculated for the uniformly contaminated sampling sites (the parameter ***s*** in Eq. (1)) was selected as a basic parameter for further estimations. One of the objectives of our research was to obtain estimates of this parameter for ^{137}Cs activity concentrations in the vegetation at the sites located on different landscapes, with different vegetation and properties of the Chernobyl depositions and to identify the most significant factors affecting variations of this parameter.

Table 2 provides the values of standard deviation (***s***) of the logarithms of the activity concentration of vegetation with ^{137}Cs at various sampling units for different sampling pitches and quantity of samples (Table 1). Since the values of standard deviation (***s***) were estimated based on a limited quantity of samples, they follow the χ^2-distribution (Stuart and Ord, 1987), which determines the non-symmetry of the errors (Δ^- and Δ^+) presented in Table 2.

The estimates of the parameters characterizing the scattering of ^{137}Cs concentrations in the vegetation at the experimental sites are illustrated in Fig. 2, together with the confidence intervals for a significance level of 0.05 (Table 2).

The average values of standard deviation (***s***) of the logarithms of the

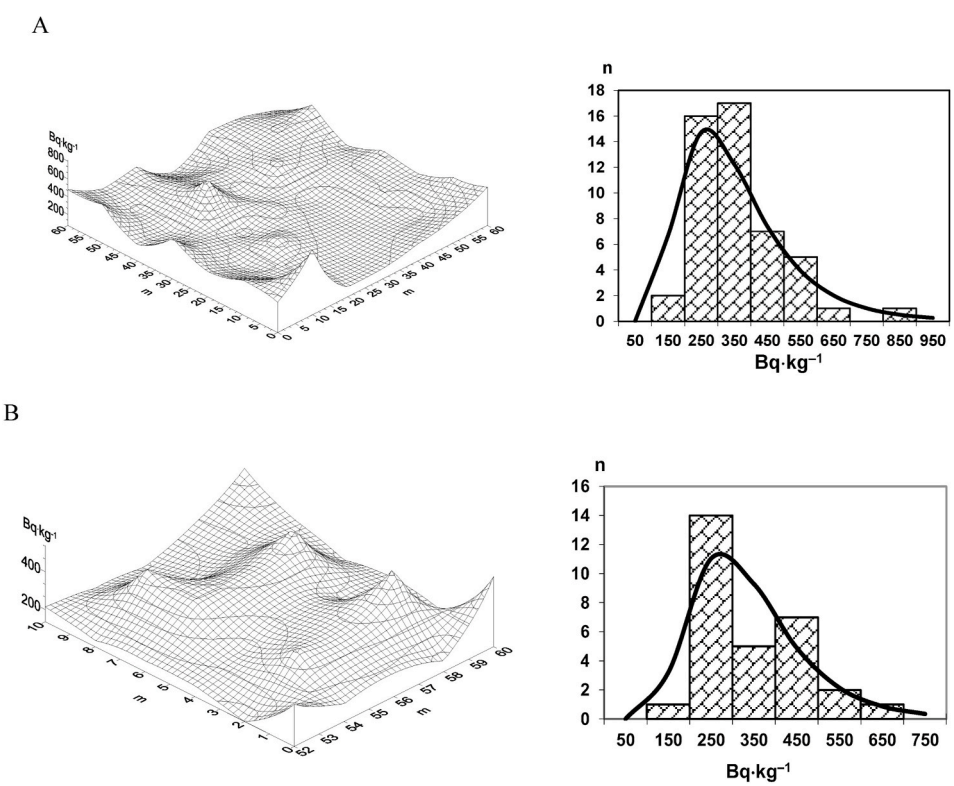

Fig. 1. Spatial variability of ^{137}Cs activity concentrations in stems and shoots of rye samples (left figures) taken in sampling unit No S9 (see Table 1). A represents the data taken within an area of 60 × 60 m (increment of 10 m) (left figure) and the corresponding distribution function (GM = 275; GSD = 1.57) (right figure) and B represents the data taken from samples within an area of 10 × 8 m (increment of 2 m) (left figure) and the corresponding distribution function (GM = 268; GSD = 1.47) (right figure). Bars present the numbers of samples within defined ranges ^{137}Cs activity concentrations in the vegetation (discrete distribution); the solid line is the approximation of the data by the log-normal distribution.

Table 2
Standard deviation of the log of the ^{137}Cs activity concentrations in plants at the sampling units.

Site #	Sampling increment, m	S	Error of s	
			Δ^-	Δ^+
S9	X1 = 10	0.45	0.06	0.09
	X2 = 2	0.38	0.07	0.11
S13	X1 = 10	0.32	0.05	0.07
	X2 = 2	0.27	0.05	0.07
S17	X1 = 10	0.40	0.08	0.13
S3	X1 = 2	0.62	0.09	0.13
S7	X1 = 1	0.22	0.03	0.05
S11	X1 = 2	0.38	0.07	0.12
S15	X1 = 10	0.36	0.06	0.10
	X2 = 2.5	0.36	0.07	0.11
S16	X1 = 10	0.54	0.10	0.17
S18	X1 = 10	0.38	0.07	0.11
	X2 = 2.5	0.27	0.05	0.09
Mean ± STD		0.38 ± 0.11		

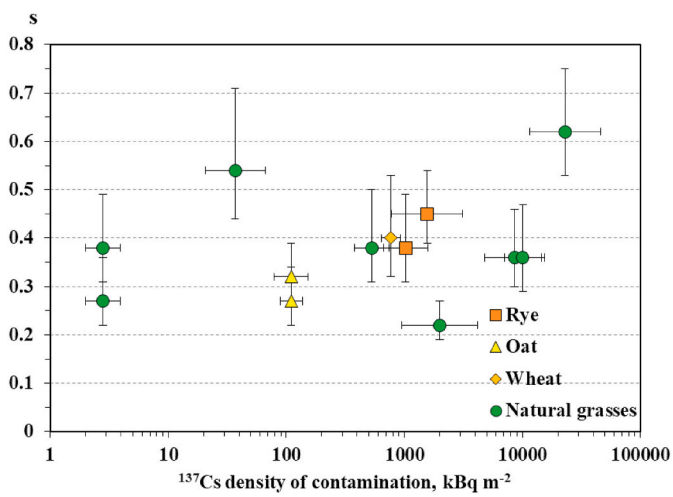

Fig. 2. Dependence of the average standard deviation of the logarithms of the ^{137}Cs activity concentrations in plants (s±Δ, Table 2) on ^{137}Cs contamination density (GM·GSD$^{\pm 1}$) at uniformly contaminated sites (Khomutinin et al., 2020).

^{137}Cs activity concentrations in the vegetation of 0.38 ± 0.11 (Table 2) were not statistically different for sampling units with a size of 60 × 60 m and of 10 × 8 m (pair correlation coefficient was of 0.22), sampling increment of 10 m and <2.5 m (Table 2, Fig. 3). The correlation analysis of these results did not reveal a statistically significant dependence of the "*s*" parameter estimates on the contamination density (Fig. 2). The Pearson pair correlation coefficient at the significance level α = 0.05 was statistically insignificant, as its average value was 0.29, and the corresponding confidence intervals for that value were −0.32 and 0.72, respectively.

It can be seen from Fig. 3 that the "*s*" parameter is also independent of the type of radioactive fallout (condensed aerosol - s = 0.36 ± 0.09 (N = 8) or fuel particles - s = 0.41 ± 0.14 (N = 5) of the Chernobyl origin) (Khomutinin et al., 2020) and vegetation type (Table 1). Thus, the observed differences were of a random nature (Figs. 2 and 3).

Thus, a geometric mean of radionuclide activity concentrations in vegetation samples collected from a sampling unit is a good estimator of plant contamination within the unit. Standard deviations of the distribution were estimated for the specific condition in this work (Table 2).

The main purpose of the further evaluation was estimating the minimum necessary number of vegetation samples that could guarantee the required precision of the GM estimate for a given dispersion of contamination density values. This can be done based on the theory of

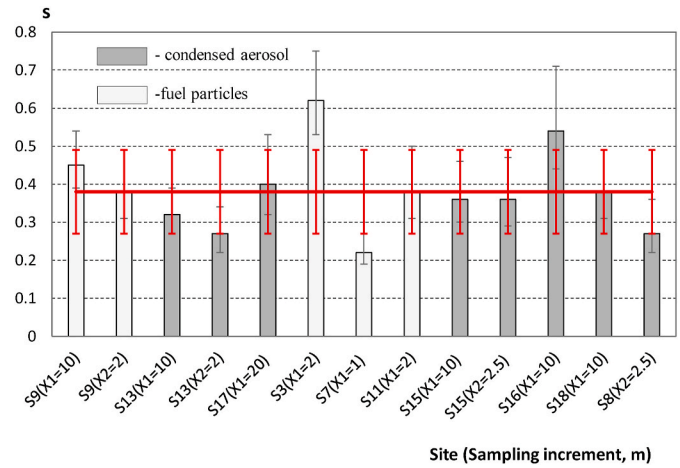

Fig. 3. Values of the standard deviations of the ^{137}Cs activity concentration logarithms in plant samples (s±Δ, Table 2) collected from various experimental sites with the different increments (with the different sampling grid resolutions). The solid line is the average $s = 0.38 \pm 0.11$ for all plants and sampling sites (Khomutinin et al., 2001). Dark grey and light grey bars represent sites contaminated by condensed aerosol and fuel radioactive fallouts, respectively.

interval estimates of unknown parameters presented in Kendall and Stuart (1973). Application of the theory with respect to the soil sampling is described in detail elsewhere (Khomutinin et al., 2020).

Data on variability of the radionuclide activity concentrations in the vegetation obtained in that study can be used to estimate the number of samples required to fit to the acceptable uncertainty level. The approach, which had been previously suggested for the soil sampling (Khomutinin et al., 2020), can also be applied for calculation of the minimum number of vegetation samples required to determine the GM of the radionuclide concentration with a given relative error for the confidence interval *p*:

$$n \geq s^2 \left\{ \frac{U_p}{\ln(1+\delta)} \right\} \tag{3}$$

where U_p is the quintile of the normal distribution for the level *p*; $\delta = \exp\left(\frac{U_p \cdot s}{\sqrt{n}}\right) - 1$ is the upper level of relative error of the median of the radionuclide concentration in a vegetation sample.

Based on Eq. (3) for $s = 0.38 \pm 0.11$ a two-dimensional nomogram was designed (Fig. 4) for calculation of the number (*n*) of individual vegetation samples required for estimating the geometric mean of the radionuclide concentrations in plants with a given relative error. Variability in the standard deviation of the logarithm of the ^{137}Cs activity concentrations results in the error of *n*. Vertical bars represent possible errors of *n* for a confidence interval of 95%. For instance, only 9 point-vegetation samples should be taken to comply with the desirable relative error of the GM of 25% and measurement errors ($\delta_{\gamma=0.95}$) of 25% (Fig. 4). If only one sample was taken, then the relative error of the plant contamination GM ($\delta_{\gamma=0.95}$) is higher than 50% for any precision of the measurements for radionuclides in that sample (Fig. 4).

It is also worth noting, that the estimated relative error of the average specific activity in the vegetation is 22 ± 8% (Fig. 4) when taking 10 samples of plants from the same areas and the radionuclide activity was measured in one mixed sample.

The analysis of the correlations of the radionuclide activity concentrations in plant samples taken from a homogeneous sampling area of ≥1 m^2 each, showed that the radionuclide concentrations in the samples taken at the points located at the distance between the neighbouring sampling points of 10 m or more are statistically independent (Khomutinin et al., 2001).

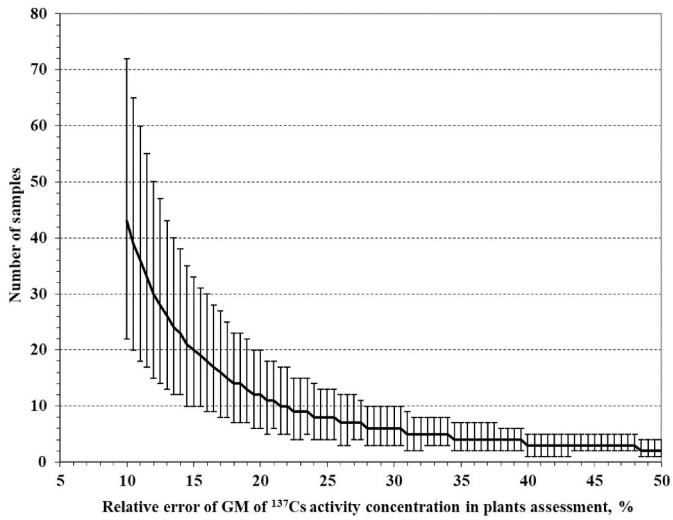

Fig. 4. Nomogram of the minimum necessary number of plant samples to evaluate the geometric mean of ^{137}Cs activity concentrations in vegetation in a uniformly contaminated site (s = 0.38 ± 0.11) to a given relative error at a confidence probability of 95%. Uncertainties of the measurement of the radionuclide activity in vegetation samples ≤10% (p = 0.95).

These findings also allow for a recommendation of a determination of the radionuclide concentrations in a combined vegetation composite sample requiring 5-point samples taken from area ≥1 m^2 each, where the distance between point vegetation samples at the sampling unit is above 10 m. In that case, the relative error of the GM of the activity concentration in vegetation on uniformly contaminated sites does not exceed 30% with a relative measurement error that not exceeding 20% at the ±2σ level of confidence (Fig. 4, Table 3).

The numbers of vegetation samples required to estimate the geometric mean of the ^{137}Cs activity concentration in vegetation with predefined uncertainty are presented in Table 3.

3.2. Statistical evaluation of ^{137}Cs soil to plant transfer factors

Aggregated transfer factors from soil to plant can be presented as the ratio of the activity concentration of radionuclide in the vegetation (Q_v, Bq kg^{-1}) to the radionuclide terrestrial density of contamination (A_s, kBq m^{-2}):

$$T_{ag} = \frac{Q_v}{A_s} \tag{4}$$

Since, Q_v and A_s (Khomutinin et al., 2020) are random values in the small scale of sampling conducted, the aggregated transfer factor (T_{ag}) (i.e. the ratio of Q_v (random value) to A_s (another random value)), is also a random value. A probability distribution of the transfer factors being a combination of two lognormal distributions should also follow the log-normal law (Smirnov and Dunin-Barkovskiy, 1969; Kendall and Stuart, 1973; Stuart and Ord, 1987). The assumption is valid for the specific sites investigated, which are uniformly contaminated.

Table 3
The minimum numbers of the single vegetation samples (≥1 m^2) required to estimate the geometric mean of the ^{137}Cs activity concentration of vegetation in composite sample with predefined uncertainty (SOU 01.1-37-426:2006).

Relative standard error of GM of ^{137}Cs activity concentration in vegetation	Relative error of measuring ^{137}Cs activity, $\delta_{meas.}$, %				
	10	20	30	40	50
10	39	41	45	50	56
20	11	11	12	14	15
30	5	5	6	7	7
40	3	3	4	4	5

Whilst conducting the field studies described here, conjugated (taken at the same sampling point) samples of plants and soils were selected on experimental sites (Khomutinin et al., 2020). Under the term "conjugated soil-plant sample", we mean a bound pair of samples that includes plants and a soil sample taken directly at the same time and at the same sampling point.

The probability distribution of the T_{ag} fits the log-normal law quite well (Fig. 5). This confirms the correctness of the previously chosen hypothesis about the log-normal law of both distributions of the soil contamination density (Khomutinin et al., 2020) and ^{137}Cs concentrations in the vegetation contamination (see section 3.1) on sampling sites.

Clear evidence of such a statement was also reported by Prister et al. (1991), who has shown that ^{137}Cs T_{ag} values reported for wide sets of agricultural plants can be successfully approximated by the log-normal distribution.

Based on the definition of aggregated transfer factor, the conclusion can be made that the standard deviation of the T_{ag} logarithms (s_{Tag}) depends on two factors, namely, standard deviation of radionuclide concentrations in the plant samples (s_{Qv}) and standard deviation of radionuclide concentrations in the soil of the sampling site (s_{As}). As shown earlier, these parameters estimated for our sampling sites were similar for all sites studied, including sites that were different in vegetation species composition, contamination density and landscape. Therefore, this assumption can also be accepted for a standard deviation of the T_{ag} logarithms (Khomutinin et al., 2020) and can be calculated as follows (Smirnov and Dunin-Barkovskiy, 1969; Kendall and Stuart, 1973):

$$s^2_{T_{ag}} = s^2_{Q_v} + s^2_{A_s} - 2 \cdot r_{As,Qv} \cdot s_{Q_v} \cdot s_{As} \tag{5}$$

where $r_{As,Qv}$ is the correlation coefficient between the logarithm of the contamination density (A_s) and the logarithm of radionuclide concentration in plants (Q_v).

Statistically significant correlation between ^{137}Cs concentrations in conjugate soil–plant pairs samples was not found for any of the sampling sites that ranged in size 6 × 6–60 × 60 m (Fig. 6). The Pearson pair correlation coefficients between the density of soil contamination with ^{137}Cs and concentration of radionuclides in plants of all studied sampling sites varied from −0.27 to +0.14 (Table 1). At the same time, there is a linear correlation between GM values of the ^{137}Cs concentrations in the plants estimated for every study site and corresponding GM values of the ^{137}Cs concentrations in the soil demonstrating acceptability of the T_{ag} concept.

In case of the composite soil samples taken with the sampler with sampling area >0.005 m^2 and the measurements error lower 10% (s_{As} amounts to 0.3 ± 0.1 (Khomutinin et al., 2020) and s_{Qv} amounts to 0.4 ± 0.1, see section 3.1), the standard deviation of T_{ag} ($s_{T_{ag}}$), calculated based on Eq. (5), can be presented as follows:

$$s_{T_{ag}} = \sqrt{s^2_{Q_v} + s^2_{A_s}} = \sqrt{0.3^2 + 0.4^2} = 0.5 \tag{6}$$

On the other hand, this value can be obtained directly based on the data of the field sampling of the conjugated samples of soil and vegetation presented in Table 1 and in Fig. 7 (Khomutinin et al., 2020).

The mean value of the standard deviation of logarithms of the ^{137}Cs T_{ag} to plants calculated based on all available conjugate data on ^{137}Cs in soil and plants ($s_{T_{ag}}$) amounted to 0.52 ± 0.08 (Fig. 7). This value nearly coincided with the above estimate for this value of 0.5 ± 0.1 that follows from Eq. (7).

Evaluation of the field data on radionuclide transfer to grain and straw of 29 agricultural plants planted at the soddy-podzolic sand soil (Bondar' et al., 1993), also showed that the standard deviations of the T_{ag} logarithms were very similar for grain and straw. Those values were similar for all plants considered and amounted to 0.5 ± 0.1 (Khomutinin et al., 2001).

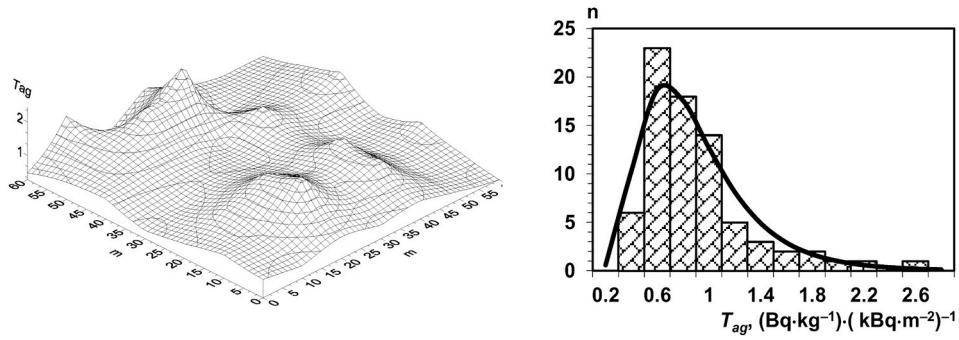

Fig. 5. Spatial variability of the ^{137}Cs soil-to-plants aggregated transfer factors (left figure) and their distribution function (GM = 0.81; GSD = 1.64) (right figure) measured in uniformly contaminated sites No. S13. Bars present the numbers of samples within defined T_{ag} ranges (discrete distribution); the solid line is the approximation of the data of the log-normal distribution.

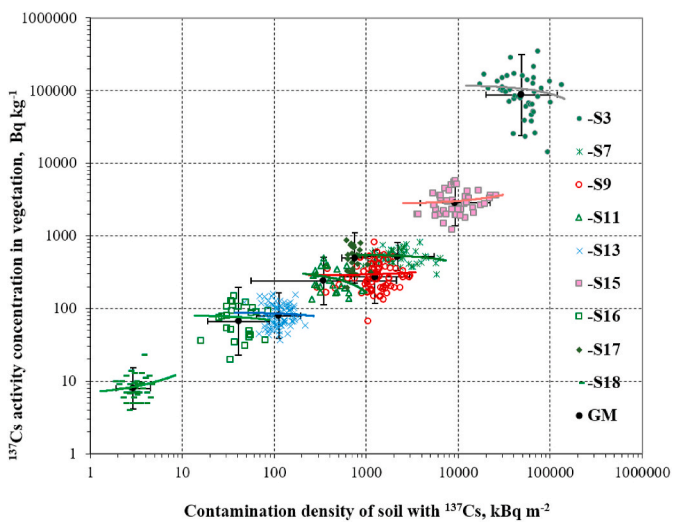

Fig. 6. Variations in the ^{137}Cs activity concentrations in the vegetation with the ^{137}Cs contamination density calculated based on conjugated soil samples. Samples of the vegetation and the soil were collected at the same point and the same time. The sampling area of the vegetation was 1 m^2. Geometric mean (GM) values for various sampling sites are given together with 95% confidence intervals.

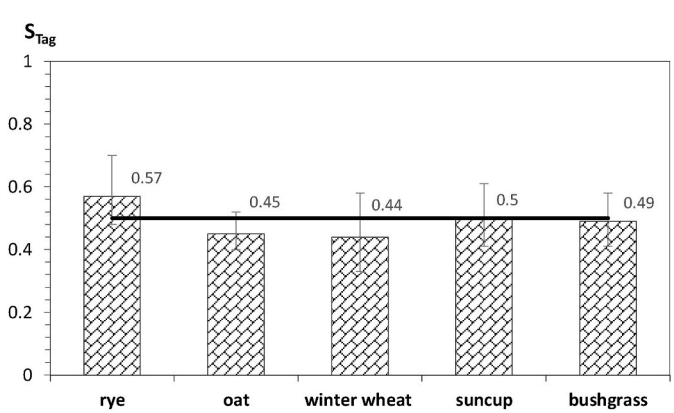

Fig. 7. Values of the average standard deviations (*s*) of the T_{ag} logarithms of ^{137}Cs for plants (s_{Tag}) with 95% confidence intervals for various stems and shoots of plant species: rye (*Secale cereal* L.), oat (*Avena sativa* L.), winter wheat (*Triticum* L.), suncup (*Oenothera* L.) and bushgrass (*Calamagrostis Epigejos* L.). The solid line is the average $s_{Tag} = 0.5$ for all plants and sampling sites (Khomutinin et al., 2001).

Thus, it can be assumed that the logarithm standard deviation of the aggregated transfer factors for ^{137}Cs based on the sampling of the vegetation from area ≥1 m^2 and sampling of the soil from area ≥0.005 m^2 is 0.5 ± 0.1 for studied plant species, soils and deposition types. Data on variability of T_{ag} values obtained in this study can be used to assess the number of samples required to comply with the required uncertainty level. Using the interval estimate (Khomutinin et al., 2020), a two-dimensional diagram was designed (Fig. 8).

The diagram allows estimating the number (n) of the conjugate soil-plant samples required to obtain a GM value of the soil-plant T_{ag} for ^{137}Cs for a uniformly contaminated site with a given relative error. For example, for the relative error of the T_{ag} geometric mean of 20% and relative error of measuring ^{137}Cs activity of sample ≤10%, it is necessary to take on average 20 conjugated composite vegetation and soil samples. The vegetation samples should be taken from the area ≥1 m^2, whilst soil samples from area ≥0.005 m^2.

On the other hand, the estimate of standard deviation of the mean T_{ag} value is (30 ± 8) % for sampling 10 conjugate soil-plant samples and radionuclide measuring activity concentration in one mixed composite sample (Fig. 8).

The reduction factor (*R*) defined as the ratio of the radionuclide transfer factors to plants before (T_{ag}) and after application of remedial options (T'_{ag}) is a basic parameter used for the remediation planning:

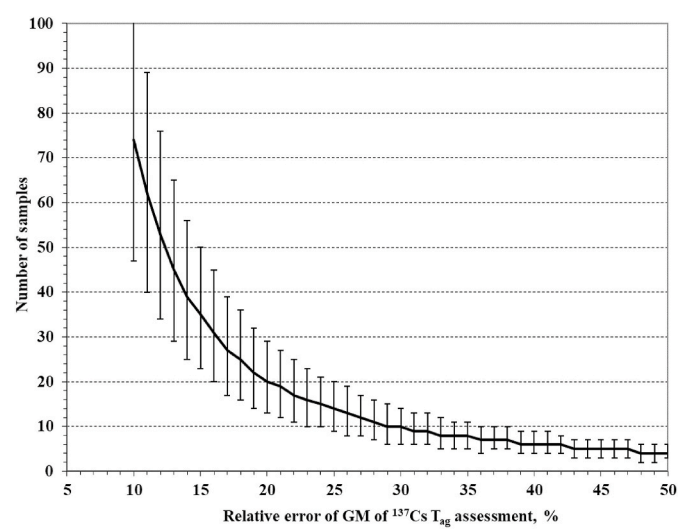

Fig. 8. Nomogram of the number of conjugate soil–plant samples required ($s_{Tag} = 0.5 \pm 0.1$) to obtain the geometric mean of ^{137}Cs T_{ag} with a prescribed relative error at a confidence probability 0.95% in a uniformly contaminated site. The uncertainties of measurement of the radionuclide activity in samples ≤10% (p = 0.95).

$$R = \frac{T_{ag}}{T'_{ag}} \quad (7)$$

As it was stated earlier (Khomutinin et al., 2001), a number of conjugated composite plant and soil samples which were required to obtain statistically significant differences between the geometric mean values of the two transfer factors (T_{ag} and T'_{ag}) can be estimated based on $s_{T_{ag}} = 0.5 \pm 0.1$.

It was shown that about 5 conjugated vegetation and soil samples should be taken at each sampling site, if the reduction factor value was approximately 2. But, when the difference between T_{ag} and T'_{ag}, determined for two sites, is only 20%, then the minimum number of samples that must be taken at each sampling site is increased up to 80 (Khomutinin et al., 2001).

4. Conclusion

The activity concentration of ^{137}Cs in plants sampled at the homogeneously contaminated sites, located in both the Chernobyl affected areas and in the regions contaminated by the global fallout may be satisfactorily approximated by the log-normal probability distributions. The standard deviations of the logarithms of activity concentration of ^{137}Cs in the vegetation were not statistically different for the sampling sites with the different contamination density and the type of radioactive deposition, different landscape and the composition of vegetation species. The same conclusion can be drawn in respect of aggregated transfer factors estimated for the study sites. The values of the average standard deviation of the ^{137}Cs activity concentration logarithms in plants and the corresponding transfer factors for the vegetation sampling area $\geq$1 m^2 and the relative activity measurement uncertainties $\leq$10% were estimated as 0.4 $\pm$ 0.1 and 0.5 $\pm$ 0.1, respectively (CI = 95%). These findings were used for estimating the minimum necessary number of plant and conjugate soil–plant samples to evaluate the geometric mean of ^{137}Cs activity concentrations in vegetation and aggregated transfer factors in a uniformly contaminated site to a given relative error at a confidence probability of 95%.

Declaration of competing interest

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

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