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Optimising sampling strategies for emergency response: Soil sampling

Yu. Khomutinin [a], S. Fesenko [b], S. Levchuk [a,*], K. Zhebrovska [c], V. Kashparov [a,d]

[a] Institute of Agricultural Radiology, National University of Life and Environmental Sciences of Ukraine, Kyiv, Ukraine
[b] Russian Institute of Radiology and Agroecology, Obninsk, Russian Federation
[c] State Institution «Institute of Environmental Geochemistry of NAS of Ukraine», Kyiv, Ukraine
[d] CERAD CoE Environmental Radioactivity/Department of Environmental Sciences, Norwegian University of Life Sciences, 1432, Aas, Norway

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ABSTRACT

The novel approach for optimising soil sampling strategies in areas affected by radionuclides is suggested. Major factors influencing the efficiency of soil sampling strategies, including (number of samples, sampling area size, sampling depth and spatial resolution of the sample sites are examined to provide optimisation of the soil sampling plan. The experimental field studies to validate the suggested approach were performed in 25 sampling units ranging from 1.2 × 1.2 m to 60 × 60 m size. The sampling units were selected on arable farmlands, natural meadow and former agricultural land), as well as coniferous and deciduous forests with contamination density of ^{137}Cs ranging from 2.8 kBq·m^{-2} to 24.5 MBq·m^{-2}. The studied areas were contaminated by both the global fallout and the Chernobyl radioactive particles of different types. To determine the values of standard deviation of the log of the soil contamination density of ^{137}Cs, 25 to 256 soil samples were collected with an increment of 0.07–10 m within each sampling unit. It was found that the values of standard deviation of the log of the soil contamination density of ^{137}Cs were not dependent on the mean contamination density, the type of radioactive deposition and the landscape features. The mean value of standard deviation calculated for all sites studied was estimated as 0.44 ± 0.15 and 0.30 ± 0.10 for the sampling area 0.001 m^2 (∅37 mm) and 0.005 m^2 (∅80 mm) at the relative measurement uncertainties lower than 10% (CI = 95%). Concentrations of ^{137}Cs in the soil samples were statistically independent when sampling points were situated at a distance larger than 1 m one from each other. A simple method was developed for assessing minimum sample sizes required for estimation of the median or the geometric mean of radionuclide soil contamination with a relative uncertainty set by the user. The approach was also suggested for estimation of the uncertainty of soil contamination for the case of composite samples. The approach was implemented in the Ukrainian national requirements for assessment of quality of the soil.

1. Introduction

Radionuclides released to the atmosphere after a nuclear or radiological emergency may consist of both gases and particulates (radioactive particles). The deposition of radionuclides on the surface of plant and soil is the first step in their transfer in the terrestrial environments and further migration along the food chains.

Overall, spatial patterns of the radioactive depositions depend on numerous environmental factors resulting in inhomogeneity both at global and local scales. The underlying reasons of the inhomogeneity are temporal and spatial instability of the radioactive clouds and local features of the landscapes: i.e. variations in plant biomass and structure, anthropogenic activities. There are two major types of the deposition that differ in deposition velocity of radionuclides and distribution of radionuclides between soil and plants, namely, dry and wet depositions (IAEA, 1994; Hoffman et al., 1995; Pröhl, 2009). Dry deposition is the explicit capture and adsorption of gases or radioactive particles by plants resulting in plant surface contamination. Wet deposition is removal of radionuclides from the air with precipitations i.e. rain, snow and hail. Radionuclides adsorbed at the plant surfaces can be lost and transferred to soil as result of some natural factors such as wind, rain and similar processes named weathering, although a small fraction can penetrate to the inner tissues of plants because of radionuclide translocation (Shaw et al., 1992; Colle et al., 2009; Hurtevent et al., 2013). Therefore, presence of plants, especially woody plants may result in the formation of local inhomogeneity of the depositions because of plant

* Corresponding author.
E-mail address: slavalevchuk64@gmail.com (S. Levchuk).

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differences in radionuclide interception and because of stem-flow and litter-fall (IAEA, 2009).

Entering the soil, radionuclides are getting involved in the migration processes such as sorption of radionuclides by clay minerals and organic matter, migration in soil profile, plant uptake and others. The migration abilities of radionuclides in soil depend on their properties as chemical elements, and soil properties, which are different in various genetic soil horizons (Fesenko et al., 2001a, 2001b; Iurian et al., 2015). These processes may result in changing of radionuclide inventory in a certain soil layer important for radionuclide transfer to plants.

In case of a nuclear emergency, a decision must be made as to whether a site should be considered as contaminated and which action should be implemented to mitigate the consequences of the contamination (Fesenko et al., 2007, 2009). Such assessments start with measurement of ambient dose or kerma rate ("**k**inetic **e**nergy **r**eleased per unit **mas**s") or even by air sampling. The latter measurements can be made only at the laboratories equipped with relevant tools and analytical supports.

The main objective of the soil sampling is to provide the required information on the radioactive deposition, which should be relevant for the site of interest. This information should also reflect the uncertainty specific for that site. In case of contamination by γ-emitting radionuclides, in field measurements such as aerial surveys, mobile monitoring and in situ gamma spectrometry can provide valuable information in detecting radionuclides and delineating areas with different soil contamination (Martin et al., 2015, 2016; Varley et al., 2017; Connor et al., 2020). Although, such techniques are difficult to apply for radionuclide mapping in case of a complex composition of radionuclides and are not applicable for measuring of betta and alpha emitters, these techniques can provide robust estimates of the mean deposition density avoiding local variations of this parameter.

Because of the Chernobyl accident, a considerable territory of the former Soviet Union, as well as of Western Europe ($>2{\cdot}10^5$ km^2), was contaminated with ^{137}Cs $>$ 40 kBq m^2 (IAEA, 2006). The feature of the Chernobyl accident was a presence of radioactive particles (mainly, in form of fuel material in particulate forms) in the release. The main part of such particles was deposited close to the 4th unit of ChNPP, whereas smaller particles were more widely dispersed (Kashparov et al., 2001, 2003; 2018; Fesenko et al., 2009).

The size of the Chernobyl fuel particles (FP), consisting of uranium oxides of different degrees of transformation, varied from one to hundreds of microns, with a density of about 8–10 g/cm^3. This led to the high rate of gravity deposition of such particles in the near zone of the ChNPP (Salbu et al., 2018).

Specific activity of ^{90}Sr in fuel particles ranged n $\times$ $10^8 \div 10^9$ Bq/g; whereas for ^{144}Ce these values were of n $\times$ 10^{10} Bq/g. The activity concentrations of ^{238,239,240}Pu and ^{241}Am were at the level of n $\times$ 10^6 Bq/g (Kashparov et al., 2003). Areas of high contamination with ^{137}Cs were observed throughout the remote zone, depending primary on rainfall at the time when the radioactive plume passed over that area. The ratio of ^{137}Cs activity to that of ^{90}Sr in the soil was being increased with the distance from the Chernobyl NPP due to the reduction of the fuel particles in the depositions and reached a value of 100–250 at the sites with prevalence of condensed type of deposition (Kashparov et al., 2003). Contamination of the central part of Chernobyl exclusion zone was represented by a superposition of the fuel and condensation component of ^{137}Cs depositions. Most of ^{137}Cs in depositions up to 5 km from the Chernobyl NPP, were bonded within the fuel particle matrix, whereas the condensation component of ^{137}Cs were predominant at the distance large 10 km from the reactor (Kashparov et al., 2001, 2018).

Assessment of the radionuclide deposition data in May 1986 showed that the territory of radioactive contamination, where protection of the population is required goes far beyond of the "circular" 30-km exclusion zone. To optimize the emergency response after the Chernobyl accident, the affected territories were subdivided in zones with different doses to the public and, therefore with different demands in countermeasure application. Radiocaesium was the major dose-forming radionuclide except the time during and immediately after the accident when short-lived and intermediate-lived radionuclides played an important role. Based on this information, the demarcation of a "uncontaminated" area was set at 1 Ci/km^2 of ^{137}Cs (37 kBq m^{-2}) (Izrael et al., 1994). This meant that any area with a contamination density above 1 Ci/km^2 was officially considered as "contaminated". The contamination levels of 1480 (in Russia) and 555 kBq m^{-2} (in Ukraine and Belarus) by ^{137}Cs, 111 kBq·m^{-2} by ^{90}Sr and 3.7 kBq m^{-2} by ^{239,240}Pu were set to identify areas where the population had to be resettled, lands should be abandoned or some restrictions on their use have to be introduced (IAEA, 2006). Further, specific countermeasure options were developed for areas with radiocaesium contamination density 37–185 kBq m^{-2}, 185–555 kBq m^{-2} and 555–1480 kBq m^{-2}. Information on contamination of the affected areas was highly important and was widely used for radiation protection of people and remediation planning both inside and outside 30 km exclusion zone.

The estimation of the contamination density with predefined uncertainty or given error in case of radiological emergency represents a big challenge both at early stage after the accident and in the long-term. It is obvious, that an increase in the number of soil samples, area of the sampler used or mass of the taken samples, may result in reducing the error of the estimation of the contamination density and spatial distribution of radionuclides at the site of interest. However, it is also clear that this comes with increased costs of labour, sampling, sample transport, preparation and analysis (IAEA, 2004; ICRU, 2006; Onda et al., 2015; IAEA, 2019).

To reduce the cost associated with sampling, a number of individual samples can be merged to obtain a composite sample that was considered as a representative one for the study area. The contamination density calculated based on activity of such composite sample differ from the actual contamination density of the site and the assessment of the uncertainty related using the composite sample for estimating contamination density was also a highly important task. Implementation of the sampling program has required a lot of resources. Identification of the minimum number of samples for assessing the contamination density within the predefined uncertainty in assessment of radionuclide concentrations in the soil, justification of required depth and mass of the sampled soil were of high importance. This allowed minimization of associated cost and other resources, while retaining the required accuracy of the environmental impact assessments (Theocharopoulosa et al., 2001).

This paper summarizes lessons learned from planning and implementation of soil sampling programs for estimation of terrestrial contamination density. These sampling programs were performed from 1986 to 2016 by the Ukrainian Institute of Agricultural Radiology (UIAR) and Russian Institute of Radiology and Agroecology in areas contaminated by the Chernobyl fallout, including the Chernobyl Exclusion Zone (ChEZ). Various factors (number of samples, sampling area, sampling depth and spatial arrangement of the samples) were examined to improve efficiency of the soil sampling and optimize the sampling planning.

2. Material and methods

2.1. Statistical justification

The distribution of any contamination within an area can be mathematically described by a continuous function of the locality coordinates $f(x,y)$. Generally, this function has three components (Khomutinin et al., 2001):

- Systematic component - monotonic change (trend) of the contamination density conditioned by the global (in respect to study area) gradient of fallout;

- Spot component - localities with increased or reduced density of contamination against a background of the trend;
- Random component is a variation of the contamination density as function of:
  - microheterogeneity of radioactive deposition in a sampling point,
  - the sample collection method and handling procedures and,
  - the analytical procedure applied for measurement of radionuclide activity concentration.

Each of the items listed above can also be represented by a function of locality co-ordinates. Combining these functions gives a contamination density *f(x,y)* in a specific point of the investigation. It is possible to present *f(x,y)* as the sum of functions describing these components (additive model), and as a product (multiplicative model). As *f(x,y)* is a strictly positive random variable, and the logarithmically normal law of probability distribution describes the probability distribution of the values in the specific point, the multiplicative model used can be described as:

$$f(x,y) = f_{tr}(x,y) \cdot f_{st}(x,y) \cdot f_{ac}, \quad (1)$$

where: $f_{tr}(x,y)$ – function describing monotonic trend of the contamination density; $f_{st}(x,y)$ – function describing spots of the contamination density against the trend; f_{ac}– random component independent of location.

The estimate of the radionuclide contamination density in a point *(x, y)* is a random value. The multiplicative model for *f(x,y)* can be substituted by an additive model for *z(x,y)* applying the simple logarithmic transformation $(Y = \ln(X))$, where:

$$z(x,y) = z_{tr}(x,y) + z_{st}(x,y) + z_{ac} \quad (2)$$

where, the transformed variables have approximately normal probability distributions.

The presentation of contamination density as (1) and (2) is sufficiently general to describe the contamination density of most complex systems. This approach is routinely applied for geological mapping and has also been successfully applied to construct the map of the radioactive contamination within the 30 km Chernobyl exclusion zone (Kasharov et al., 2001, 2003, 2018).

Sampling planning, measurements for radionuclide activity concentrations in samples, and evaluation of the results are based on assumptions concerning the probability distributions of these variables. It is well known that in the Earth's crust, the concentration of elements and their natural radioactivity often follow a log-normal distribution (Krige, 1966; Malanca et al., 1996). The results of this study have showed that the variability of the deposition density of radionuclides were asymmetrical and could be fitted by a lognormal distribution (Stuart and Ord, 1987; Khomutinin et al., 2001; Perevolotskiy, 2006). The probability density of the log-normal distribution can be presented as:

$$f(C_s) = \frac{1}{C \cdot s \cdot \sqrt{2\pi}} e^{-\frac{1}{2}\left[\frac{\ln(C)-\mu}{s}\right]^2} \quad (3)$$

where: C is the soil contamination density; μ is the mean/average log[1] of the soil contamination density and s is standard deviation of the log of the soil contamination density. The parameters of this probability distribution have a well-known physical meaning.

The contamination density median equals:

$$M_C = \exp(\mu). \quad (4)$$

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

which is also known as the geometric mean (GM). The parameter ***s*** is the approximation of the coefficient of variation W_c of the contamination density at the site. This parameter is independent of the half-life of the radionuclide of concern:

$$s \cong \frac{SD}{\overline{C}} = W_C \quad (5)$$

where: *SD* is the standard deviation of the soil contamination density; is the average soil contamination density.

A log-normal distribution is defined as a distribution where the logarithms of the values and not obligatory 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 application of the geometric mean (GM) and geometric standard deviation (GSD). GSD is the exponent of the standard deviation of the natural logarithms of the individual values of the soil contamination density with a radionuclide.

The geometric mean provides a good estimate of the most probable value for the log-normally distributed data, and this parameter are not so much affected by outliers as the arithmetic mean is.

Dispersion of the log of the soil contamination density in samples, s^2, is conditioned by micro-non-uniformity of the site contamination including discrete fuel particles, by the sample preparation method prior to the measurement. This parameter includes the size selection of the measured sub-sampling, the uncertainty associated with the sampling scheme, and radioactivity measurements.

2.2. Sites description

The overall approach taken in the mapping of Chernobyl affected areas was based on selection of the sampling units within areas characterised by random variation in soil contamination. The examples of such sites are given in Table 1. The control sites (S18 and S19) were selected because the ^{137}Cs activity concentrations in the soil (at the deposition density of 2–3 kBq m^{-2}) were primarily of global fallout origin. Seventeen of the study sites (S1–S16 and S23) were located inside of the Chernobyl exclusion zone and only seven sampling sites (S2' and S17-22) were selected outside. The depositions were presented by the condensed component of the Chernobyl fallout formed as a result of the condensation of volatile fission products at nine sites. Substantial contributions of the fuel particles to the total activity (above 25%) were observed at the rest twelve sites. The latter sites were selected largely within the 10-km ChNPP zone in the areas where the total contamination density with ^{137}Cs ranged from 0.38 to 30 MBq m^{-2}. In terms of land use, the sites constituted of: natural meadow (five sites), former agricultural land (eight sites) arable land (seven sites) and five forests sites (Table 1).

The sampling sites on natural meadow S1, S2' and S14 were located on flooded sites of the rivers Pripyat and Povch, sampling sites S2 and S16 on dry meadows, covered by natural perennial grass with plants of 20–40 cm of height with prevalence of wood small-reed (*Calamagrostis epigejos*). At the time of the Chernobyl depositions (26.04–06.05.1986), at the study sites there were only deep litter and a stand of dry grass of 1985, whereas new grasses at the site just started to grow.

After the evacuation of the population from the Chernobyl exclusion zone vast arable lands areas were abandoned and because of the succession process these fields began to overgrow with wild-growing herbs (papaya creeping, oatmeal red, etc.) and tree understorey. At the time of the Chernobyl depositions, these lands were either completely without vegetation after ploughing or covered by winter cereals. At the time of sampling, i.e. in 2016, the lands were covered with natural perennial grasses.

The sampling sites were also selected on arable lands that were a part of the crop rotation and had been ploughed several times within experimental studies both inside of the Chernobyl exclusion zone (S9,

Table 1
Major characteristics of sampling sites.

Site #	Year	Coordinates		Fraction of fuel particles[a]	Land use	Sampling parameters				Geometric mean of contamination density[b], kBq/m^2
		Latitude N°	Longitude E°			*Sampling pitch, m*	*Area, m^2*	*Unit size, m*	*Quantity of samples*	
Global fallout sites										
S18 [c]	2003	50.632	32.619	$\Delta FP = 0\%$	Fallow	X1 = 10	0.005	50 × 40	30	2.8·(1.4) $^{\pm 1}$
						X2 = 2.5	0.005	10 × 10	21	
S19	2003	50.600	32.540	$\Delta FP = 0\%$	Arable land	X1 = 10	0.005	50 × 40	30	3.2·(1.3) $^{\pm 1}$
						X2 = 2.5	0.005	10 × 10	21	
Chernobyl deposition sites: condensed deposition										
S2	1991	51.250	29.360	ΔFP<5%	Meadow	X1 = 5	0.015	45 × 45	100	250·(1.2) $^{\pm 1}$
S2′	1991	51.137	28.621	$\Delta FP = 0\%$	Meadow	X1 = 5	0.015	35 × 60	104	520·(1.3) $^{\pm 1}$
S13[c]	1999	51.153	30.353	ΔFP<10%	Arable land	X1 = 10	0.001 and 0.004	60 × 60	98	110·(1.4) $^{\pm 1}$
						X2 = 2	0.001 and 0.004	10 × 8	58	
						X3 = 0.1	0.001	0.9 × 0.9	100	
S15[c]	1999	51.303	29.642	ΔFP<10%	Arable land	X1 = 10	0.001 and 0.004	60 × 60	98	8600·(1.8) $^{\pm 1}$
						X2 = 2.5	0.001	10 × 10	21	
						X3 = 0.15	0.001	0.9 × 0.9	49	
S16[c]	1999	51.289	29.853	ΔFP<10%	Meadow	X1 = 10	0.001 and 0.004	40 × 40	50	37·(1.8) $^{\pm 1}$
						X2 = 2.5	0.001	10 × 10	21	
						X3 = 0.1	0.001	0.3 × 0.3	24	
S17 [c]	1999	51.182	29.492	ΔFP<2%	Arable land	X1 = 10	0.001 and 0.004	60 × 60	98	770·(1.2) $^{\pm 1}$
						X2 = 2	0.001 and 0.004	8 × 8	24	
						X3 = 0.2	0.001	1.2 × 1.2	48	
S20[c]	2013	51.140	28.600	$\Delta FP = 0\%$	Forest	X1 = 5	0.001	50 × 50	121	190·(1.4) $^{\pm 1}$
S21[c]	2013	51.110	28.430	$\Delta FP = 0\%$	Forest	X1 = 5	0.001	40 × 40	81	180·(1.4) $^{\pm 1}$
S22[c]	2016	51.231	29.430	$\Delta FP = 0\%$	Forest	X1 = 5	0.001	30 × 50	77	2720·(1.7)
Chernobyl deposition sites: fuel particles										
S1	1991	51.450	30.064	ΔFP≈30%	Meadow	X1 = 5	0.015	45 × 50	110	6250·(1.2) $^{\pm 1}$
S3[c]	1999	51.377	30.036	ΔFP≈40%	Fallow	X1 = 2	0.001	8 × 8	25	23000·(2.0) $^{\pm 1}$
						X2 = 0.2	0.001	1.2 × 1.2	48	
						X3 = 0.07	0.001	0.2 × 0.2	12	
S4	1991	51.377	30.036	ΔFP≈40%	Fallow	X1 = 5	0.015	45 × 40	90	24500·(1.1) $^{\pm 1}$
S5	1999	51.371	30.031	ΔFP≈40%	Forest	X1 = 2	0.001	8 × 8	25	6600·(1.4) $^{\pm 1}$
S6	1991	51.291	29.681	ΔFP≈25%	Fallow	X1 = 5	0.015	45 × 45	100	2200·(1.2) $^{\pm 1}$
S7[c]	1999	51.341	30.103	ΔFP≈25%	Fallow	X1 = 2	0.001	8 × 8	25	2000·(2.1) $^{\pm 1}$
						X2 = 0.2	0.001 and 0.015	1.2 × 1.2	98	
						X3 = 0.07	0.001	0.2 × 0.2	12	
S7′	1991	51.341	30.103	ΔFP≈25%	Fallow	X1 = 5	0.015	45 × 50	110	2100·(1.6) $^{\pm 1}$
S8	1999	51.341	30.100	ΔFP≈25%	Forest	X1 = 2	0.001	8 × 8	25	2300·(1.4) $^{\pm 1}$
S9[c]	1999	51.340	30.126	ΔFP≈25%	Arable land	X1 = 10	0.001 and 0.004	60 × 60	98	1570·(2.0) $^{\pm 1}$
						X2 = 2	0.001 and 0.004	8 × 10	58	
						X3 = 0.1	0.001	0.9 × 0.9	99	
S10	1991	51.290	30.100	ΔFP≈30%	Fallow	X1 = 5	0.015	50 × 45	110	1100·(1.6) $^{\pm 1}$
S11[c]	1999	51.264	30.096	ΔFP≈25%	Fallow	X1 = 2	0.001	8 × 8	25	530·(1.4) $^{\pm 1}$
						X2 = 0.2	0.001 and 0.015	1.2 × 1.2	98	
						X3 = 0.07	0.001	0.2 × 0.2	12	
S12[c]	1999	51.263	30.090	ΔFP≈25%	Forest	X1 = 2	0.001	8 × 8	25	380·(1.6) $^{\pm 1}$
S14	2000	51.471	30.016	ΔFP≈25%	Meadow	X1 = 10	0.001	50 × 30	24	1460·(1.9) $^{\pm 1}$
						X2 = 2.5	0.001	10 × 10	21	
S23[c]	2016	51.434	30.109	ΔFP≈25%	Forest	X1 = 5	0.001	30 × 30	49	2510·(1.4) $^{\pm 1}$

[a] The values of ***ΔFP*** are given for 1999 (Kashparov et al., 2003).
[b] At the date of sampling.
[c] The conjugate plant samples were taken together with soil samples at these sites.

S13, S15) and beyond it. On the sites, which were outside of the Chernobyl exclusion zone (S17, S19) the traditional farming practice, was used for planting of plants. Thus, at the time of sampling, rye, oats and wheat were planted at the sites of S9, S13 and S17, whilst lands of sites of S13 and S19 were freshly ploughed.

The forests sites were selected both in coniferous forest (S5, S8, S12, S20, S23), presented mainly by Scots pine (*Pinus sylvestris* L.), and in deciduous forests (S21 and S22) presented by silver birch (*Betula pendula (L.) Roth.*). The forest sites of S20-22 were selected outside of the Chernobyl exclusion zone within the western spot of the Chernobyl condensed depositions. At the time of Chernobyl depositions in 1986, the pine's age at experimental sites ranged from 20 to 35 years that of birch from 8 to 15 years. At the time of soil sampling, the height of the pine trees was 15–30 m with a diameter at the height of 1.3 m of 10–30 cm, and the height of birch was 15–25 m with a diameter at the height of 1.3 m of 8–23 cm.

2.3. Soil sampling

The migration rate of radionuclides in the soil profile was considered for making decision on the most appropriate sampling depth to measure the total deposition density of radionuclides. In the first years after the Chernobyl accident, soil sampling was predominantly performed by ring (⌀ 14 cm, 0.015 m^2) to a depth of h = 5 cm in accordance with the sampling guide that was adapted at that time (IAEA, 1992). Subsequently, due to vertical migration of radionuclides because of natural processes and possible redistribution of radionuclides in the soil profile made by wild animals or by human activities, the depth of soil sampling was increased to 30 cm. Therefore, cylindrical core sampler (⌀ 3.7 cm, 0.001 m^2, h = 30 cm) and ring (⌀ 14 cm, 0.015 m^2, h = 5 cm) were used for soil sampling in the current research (see Table 1).

Additional surveys, which had been performed that time by the Ukrainian Institute of Agricultural radiology (UIAR), showed that a sampling depth of 30 cm was mainly sufficient for representative estimates of deposition densities of the main long-lived radionuclides of the Chernobyl fallout, including ^{137}Cs, ^{90}Sr and transuranic elements. The exception was low-humus sand soils, where the depth of vertical migration of ^{90}Sr could exceed 30 cm) (Shestopalov et al., 2003). Therefore, the sampling depth of 30 cm was recommended for the soil sampling and was used for the sites presented in Table 1.

Sampling was carried out by a rectangle grid with a spacing distance X1 = 2, 5 or 10 m (Table 1). Grid soil samples were taken at intersection points between grid cells, consisting of 1 and 4 or 5 composite cores taken within a 20-cm radius (see Fig. 1). Each core was measured as an individual sample. Four or five cores were combined forming a composite sample for measurement. In 1991 the ring sampler (⌀ 14 cM, h = 5 cM) was used for soil sampling at the sites S1, S2, S2′, S4, S6, S7' and S10 (Table 1).

To estimate the influence of size of the sampling pitch on the standard deviation of the logarithms of soil contamination density of ^{137}Cs the method of embedded sites was used for sampling on some experimental site. Inside the large grid a cell was divided into a medium grid with a spacing distance of X2 = 0.2, 2 or 2.5 m (Table 1). Soil samples were taken at interception point in the same way as it was done for the large grid. Inside the medium grid a cell was chosen and divided into a fine grid with X3 = 0.07, 0.1 or 0.15 m pitch of sampling (Table 1), as shown in Fig. 1. In each node of the fine grid one soil core was selected and measured as an individual sample.

2.4. Measurements

Activity concentrations of ^{137}Cs 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*) and a passive protection device, and was operated using the *OMNIGAM* software. Polyethylene cylindrical containers (130 cm^3) and Marinelli beakers (1000 cm^3) were utilized for the measurements. Calibration of the spectrometer was conducted using certified standards (soil matrix multi-nuclides standard, V.G. Khlopin Radium Institute, Russia).

QC/QA procedures included regular monitoring of the system performance, control 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 internal validation results obtained through gamma measurements of IAEA CRM (IAEA-375, "Radionuclides and trace elements in soil") showed good agreement with the certified values. Relative total combined uncertainties of measurements (for 95% confidence level) did not exceed following values: 10% for sites contaminated with Chernobyl deposition, and 25% for sites where radionuclide contamination had mostly global fallout origin.

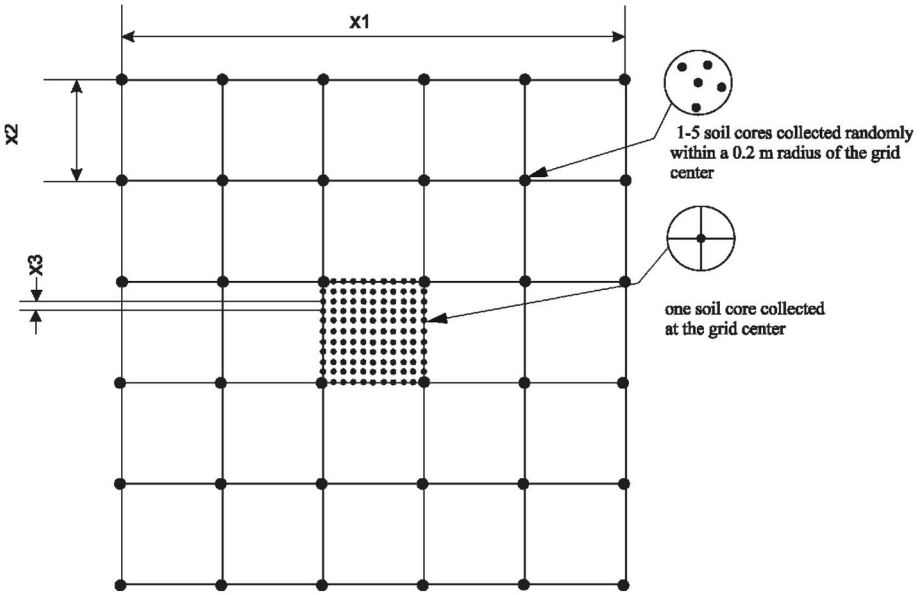

Fig. 1. The embedded sites scheme for soil sampling. X1-a large grid cell, X2-a medium grid cell, X3-a fine grid cell.

Activity concentration of ^{90}Sr in soil samples taken in 1999 from sampling unit S3, S5, S7, S8, S11 and S12 with size of unit 8 × 8 m (Table 1) (sampling pitch was 2 m, sampling area – 0.001 m^2, quantity of samples −25) was measured by non-destructive beta-spectrometric method (Courti et al., 2002) using a beta-spectrometer (SEB-01-70, AKP, Ukraine) with thin scintillation plastic detector.

3. Results and discussion

The results obtained show that values of the mean ^{137}Cs soil contamination density calculated based on samples, taken at the distance of a few meters one from another, can differ more than one order of magnitude (Fig. 2). The high inhomogeneity can be illustrated by the following example. During soil sampling at the forest sampling unit S20 (Fig. 2a), the samples of fruit bodies of some mushroom species were also sampled. The results of the measurements for ^{137}Cs concentrations in these samples have shown that activities of the samples of *Boletus badius*, *Russula paludosa Britz*, *Suillus bovinus* and *Collybia dryophilia ghb* with the fresh weight of 50–100 g ranged from 1.7 to 2.7 kBq for ^{137}Cs. The decomposition of these mushrooms can lead to local variation of the ^{137}Cs contamination density from $190 \cdot (1.4)^{\pm 1}$ kBq m^2 (Table 1) to 460 kBq m^2. This effect alone can already lead to large errors in the determination of the mean contamination of sites when selecting a small number of samples.

The evaluation of the probability distributions of ^{137}Cs concentrations in samples taken in all sample units presented in Table 1 has shown that all of them follow the lognormal distribution. Typical histogram of ^{137}Cs soil contamination density (in kBq m^{-2}) and its lognormal approximation are shown in Fig. 2.

Standard deviation and coefficient of variation are normally used to characterize the dispersion. Therefore, it was also assumed that soil contamination density had a log-normal probability distribution. In that case, the standard deviation normalized on the mean value characterizes relative dispersion of values of soil contamination density (the coefficient of variation or relative standard deviation).

An important feature of the dispersion of the logarithms of soil contamination density was that this parameter does not depend on time (on the decay of radionuclides) and can be considered as some time invariant (in contrast, for example, to the coefficient of variation of the logarithm of soil contamination density). Therefore, the standard deviation of the logarithms of soil contamination density (s) was selected as a basic parameter for estimating the variability of the density of soil contamination on non-gradient sites. Thus, one of the objectives of our research was to obtain estimates of this parameter for areas of ^{137}Cs on the sites located on different landscapes, with different physicochemical 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 soil ^{137}Cs contamination density at various sampling sites measured for different sampling parameters including: different sampling pitches, sampling areas, and quantity of samples taken at the site (Table 1). Since the values of standard deviation (s) were estimated based on a limited quantity of individual samples, they follow the χ^2-distribution (Stuart and Ord, 1987), which determines the non-symmetry of the errors (Δ- and Δ+) presented in Tables 2 and 3.

The mean values of standard deviation (s) of the logarithms of the ^{137}Cs soil contamination density of 0.44 ± 0.15 (Table 2) and that of ^{90}Sr of 0.44 ± 0.13 (Table 3) are not statistically different ($t(37) = 2.0$, $p > 0.05$) for sampling sites with size of 8 × 8 m, sampling pitch of X1 = 2 m, sampling area of 0.001 m^2 and the number of samples taken at the site of 25 (typical sampling parameters for sites S3, S5, S7, S8, S11 and S12). The mean values of standard deviation (s) of the logarithms of the soil contamination densities were decreasing from 0.44 ± 0.15 to 0.24 ± 0.11 with the increase of the sampling area from 0.001 m^2 to 0.015 m^2 (see Table 2 and Fig. 3). It was also found that the standard deviation is not statistically different ($t(31) = 2.0$, $p > 0.05$) for sites with condensed

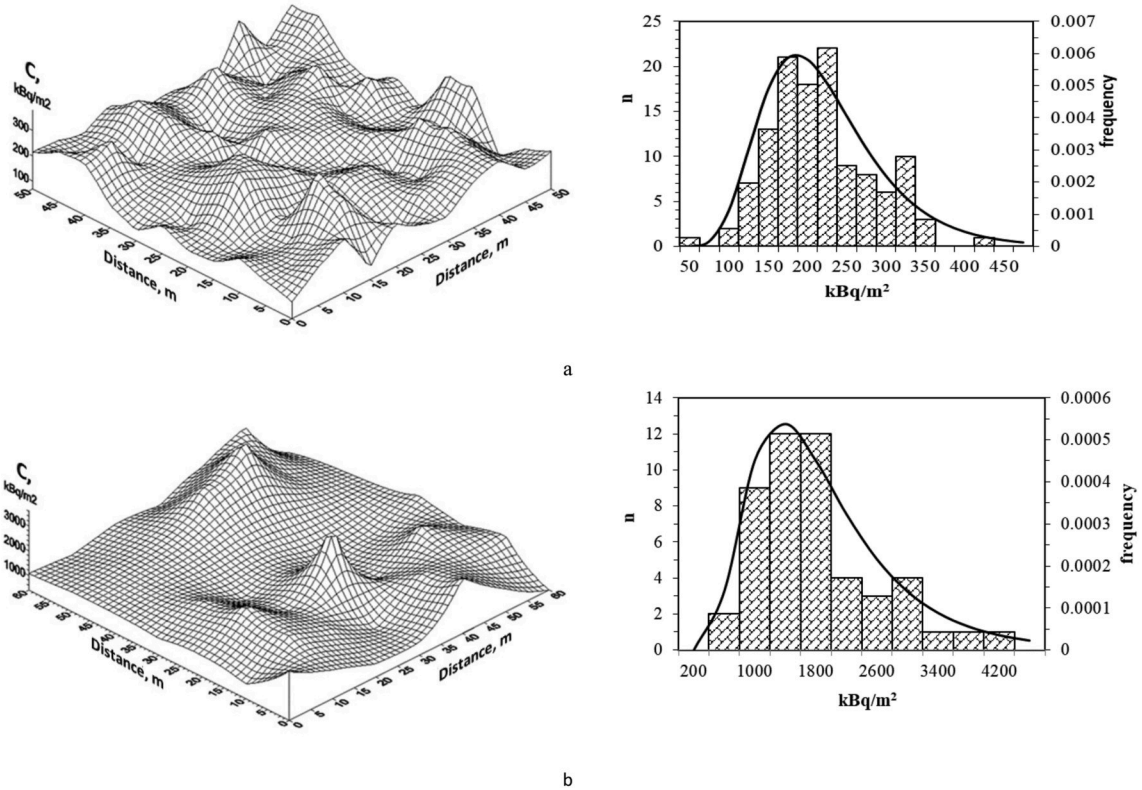

Fig. 2. Distributions of probability of soil contamination density of ^{137}Cs (C) on a uniformly contaminated site (n-quantity of samples): a – S20 (condensed fallout, forest) and b – S9 (fuel depositions, arable land).

Table 2
The values of standard deviation (s) of the log of the soil contamination density with ^{137}Cs at various experimental sampling units.

Site #	Sampling increment, m	Standard deviation of the log of the soil contamination density (s)											
		Sampling area of 0.001 m^2			Sampling area of 0.004 m^2			Sampling area of 0.005 m^2			Sampling area of 0.015 m^2		
		s	Error		s	Error		s	Error		s	Error	
			Δ^-	Δ^+		Δ^-	Δ^+		Δ^-	Δ^+		Δ^-	Δ^+
S1	X1 = 5										0.22	0.03	0.02
S2	X1 = 5										0.15	0.02	0.02
S2′	X1 = 5										0.27	0.04	0.03
S3	X1 = 2	0.70	0.22	0.13									
	X2 = 0.2	0.56	0.12	0.08									
	X3 = 0.07	0.46	0.30	0.13									
S4	X1 = 5										0.10	0.01	0.01
S5	X1 = 2	0.55	0.17	0.10									
S6	X1 = 5										0.33	0.04	0.03
S7	X1 = 2	0.74	0.24	0.14									
	X2 = 0.2	0.46	0.09	0.07							0.28	0.06	0.04
	X3 = 0.07	0.47	0.21	0.11									
S7′	X1 = 5										0.18	0.02	0.02
S8	X1 = 2	0.37	0.12	0.07									
S9	X1 = 10	0.49	0.10	0.07	0.32	0.07	0.05	0.30	0.06	0.04			
	X2 = 2	0.52	0.15	0.09	0.47	0.06	0.05	0.44	0.06	0.05			
	X3 = 0.1	0.70	0.10	0.07									
S10	X1 = 5										0.46	0.06	0.05
S11	X1 = 2	0.34	0.11	0.06									
	X2 = 0.2	0.36	0.08	0.06							0.21	0.05	0.03
	X3 = 0.07	0.28	0.17	0.07									
S12	X1 = 2	0.45	0.15	0.09									
S13	X1 = 10	0.35	0.05	0.07	0.33	0.07	0.05	0.31	0.07	0.05			
	X2 = 2	0.36	0.06	0.04	0.21	0.06	0.04	0.21	0.06	0.04			
	X3 = 0.1	0.25	0.04	0.03									
S14	X1 = 10	0.64	0.21	0.12									
	X2 = 2.5	0.63	0.20	0.12									
S15	X1 = 10	0.56	0.12	0.08	0.45	0.09	0.06	0.42	0.09	0.06			
	X2 = 2.5	0.49	0.08	0.07	0.47	0.15	0.09	0.37	0.12	0.07			
	X3 = 0.15	0.50	0.11	0.07									
S16	X1 = 10	0.55	0.17	0.10	0.48	0.15	0.09	0.39	0.12	0.07			
	X2 = 2.5	0.61	0.12	0.08	0.52	0.17	0.10	0.45	0.15	0.09			
	X3 = 0.1	0.23	0.07	0.04									
S17	X1 = 10	0.20	0.04	0.03	0.18	0.04	0.03	0.16	0.03	0.02			
	X2 = 2	0.16	0.06	0.03	0.20	0.06	0.04	0.16	0.06	0.03			
	X3 = 0.2	0.15	0.03	0.02									
S18	X1 = 10							0.22	0.06	0.04			
	X2 = 2.5							0.34	0.11	0.06			
S19	X1 = 10							0.24	0.08	0.05			
	X2 = 2.5							0.19	0.06	0.04			
S20	X1 = 5	0.34	0.05	0.04									
S21	X1 = 5	0.33	0.06	0.04									
S22	X1 = 5	0.50	0.10	0.07									
S23	X1 = 5	0.35	0.09	0.06									
Mean ± STD		**0.44 ± 0.15**			**0.36 ± 0.13**			**0.30 ± 0.10**			**0.24 ± 0.11**		

Table 3
The values of standard deviation (s) of the log of the soil contamination density of ^{90}Sr at various experimental sampling units 8 × 8 m (sampling pitch was X1 = 2 m, sampling area – 0.001 m^2, quantity of samples was 25).

Site #	s	Error	
		Δ^-	Δ^+
S3	0.56	0.18	0.11
S5	0.48	0.15	0.09
S7	0.52	0.17	0.10
S8	0.26	0.09	0.05
S11	0.30	0.09	0.06
S12	0.55	0.18	0.10
Mean ± STD	0.44 ± 0.13		

and fuel types of the Chernobyl depositions (Fig. 3).

A soil sample taken by a sampler with a large working sampling area can be considered as a set of samples (n) taken by a sampler with a smaller sampling area where sampling points are close each to others. The radionuclide concentrations in such samples will be statistically dependent values. Thus, the radionuclide activity concentrations in a soil sample taken by a sampler with a large sampling area can be considered as a rough estimate of the radionuclide activity concentrations in statistically dependent samples taken by a sampler with a smaller sampling area. As discussed above (see section 2.1), a random variable representing the logarithm of soil contamination density ^{137}Cs may be described by a normal probability distribution. Thus, the dispersion of the mean value of n statistically dependent random variables in assumption that the variables have an equal standard deviation (s_1) can be presented as follows (Stuart and Ord, 1987):

$$s^2 = \frac{{s_1}^2}{n^2} \cdot \left(n + 2 \cdot \sum_{i=1}^{n} \sum_{j=1}^{i-1} r_{i,j} \right), \quad (6)$$

where s –the standard deviation of the mean value of n dependent random variables; s_1 – the standard deviation of the dependent random variables; n – number of the variables; $r_{i,j}$– the corresponding correlation coefficients.

In the case all the samples are located very close each other, the values of $r_{i,j}$ are rather close and can be approximately considered equal

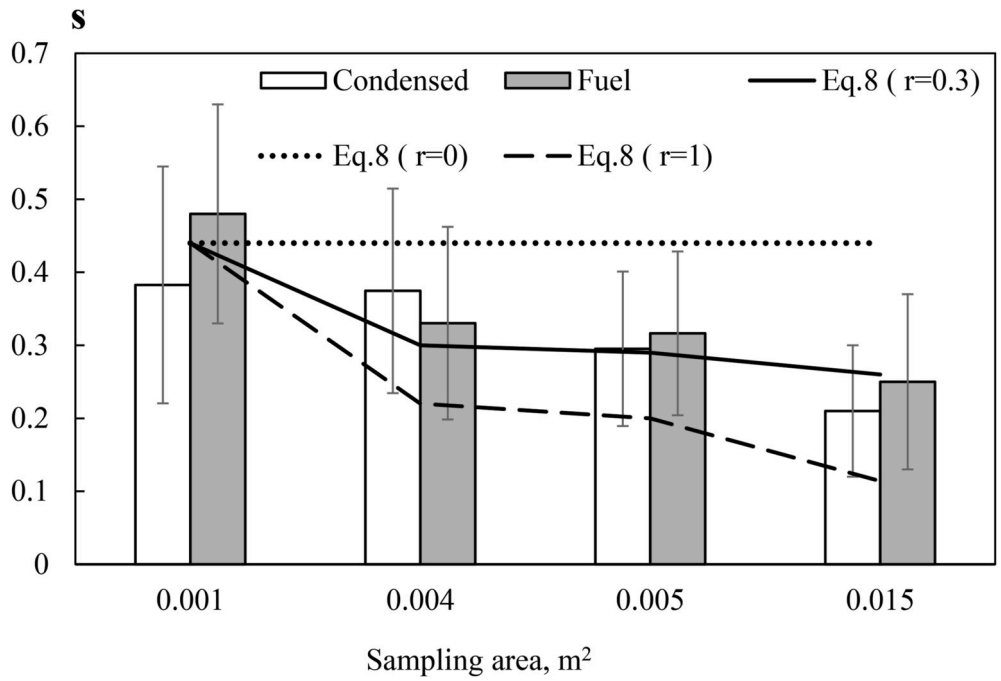

Fig. 3. Variation in average standard deviation of the logarithms of soil contamination densities of ^{137}Cs (mean ± SD) with sampling area size the.

to some effective correlation coefficient (r). Then the expression (6) can be simplified as follows.

$$s^2 = \frac{s_1{}^2 \cdot (1 + r \cdot (n-1))}{n} \tag{7}$$

For $r = 0$ (statistically independent random variables) we obtain the expression well known in statistics (Stuart and Ord, 1987):

$$n = \frac{S}{S_1},$$

where S is the working area for any arbitrary sampler; $S_1 = 0.001$ m^2 - the area of the used sampler. Then the dispersion of the logarithms of soil contamination density at the study site is dependent on the working area of the sampler:

$$s^2 \frac{0.001 \cdot s_1^2}{S} \left[1 + r \cdot \left(\frac{S}{0.001} - 1\right)\right] \tag{8}$$

where s_1 is the standard deviation of the logarithm of soil contamination density when sampling a cylindrical sampler with a diameter of ∅ 3.7 cm (S = 0.001 m^2).

The average standard deviations of the logarithms of soil contamination densities for sampling sites studied complied the relationship given by Eq. (8) for the correlation coefficient (r) value of 0.3 (Fig. 3).

Most of the values used to assess a standard deviation of the logarithms of the soil contamination densities for various sampling sites were obtained with the aid of the cylindrical core sampler ∅37 mm, 0.001 m^2, h = 30 cm). The values of the standard deviation of the logarithms of ^{137}Cs contamination densities obtained at different sites (s) for the samplers with working areas above 0.001 m^2 were recalculated (standardized) to the sampler working area of 0.001 m^2 (∅37 mm).

The analysis of the data given in Table 2 and Figs. 4 and 5 has shown that the influence of the mean ^{137}Cs contamination density, type of deposition, size of the pitch and landscape type on the standard deviation of the logarithms of ^{137}Cs contamination density (parameter (s)) was not statistically significant. In particular, the values of ^{137}Cs contamination density and s were found to be weakly correlated, r(42) = 0.18, p = 0.26. A one-way between subjects ANOVA was conducted to compare the effect landscape type on the standard deviation of the logarithms of ^{137}Cs contamination density in arable land, fallow, forest and meadow conditions. There were no statistically significant differences between landscape group means at the $p < 0.05$ level for the four conditions (F(3,18) = 0.75, p = 0.55). Also, an analysis of variances was conducted to compare the effect of size of the sampling increment (in the range of 5–10 m) on s. Results indicated that the effect was non-significant at the $p < 0.05$ level (F(2,19) = 0.80, p = 0.47). Therefore, for each site, the obtained values of the mean standard deviation of the logarithms of ^{137}Cs contamination densities were averaged considering their weighting coefficients (uncertainties of measurements) (Table 3). The obtained values amounted to 0.44 ± 0.15 and 0.30 ± 0.10 for the sampling areas of 0.001 m^2 (∅37 mm) and 0.005 m^2 (∅80 mm), both for the relative measurement uncertainties lower than 10% (CI = 95%). The weight of samples amounted to 0.2–0.4 kg and 1–2 kg for soil sampling for the depth of 20–30 cm with sampling areas of 0.001 m^2 (∅37 mm) and 0.005 m^2 (∅80 mm), respectively.

An increase of quantity of samples, sample area, mass or volume was accompanied by reduction of the uncertainty in estimating of radionuclide concentrations in the soil. However, the cost for the sample transportation and the time of sample preparation for measurements was also increased. The choice of these parameters is a part of optimisation of the sampling programme.

It was assumed that the activity concentrations of radionuclides are greater than the minimum detectable activity (MDA) for the measurement techniques used in the study. The detection limits for environmental samples analysis depend on the objectives of the sampling and the requirements of the end user. In the presented study these were typically of the order around n × 10^0 Bq for gamma- and beta-emitting radionuclides (^{54}Mn, ^{60}Co, ^{65}Zn, ^{90}Sr, ^{95}Zr,-^{95}Nb, ^{134,137}Cs, ^{144}Ce etc.) and about n × ·10^{-3} Bq for alpha-emitting radionuclides (^{238}U, $^{238,239-240}$Pu, ^{241}Am etc.) after chemical extraction (L'Annunziata, 2012). To achieve these detection levels, the dry weight of sampled material usually amounted to 0.1–1 kg for gamma- and beta-emitting radionuclides, and only a few grams for alpha-emitting radionuclides. For example, density of global fallout deposition of ^{137}Cs in the northern hemisphere was at the time of the research 2–3 kBq m^{-2}. Therefore, a soil sample of 0.001 m^2 area had activity of 2–3 Bq, which was higher than the MDA. As the sample volume was about 200 cm^3, ^{137}Cs activity concentration was about 10 Bq kg^{-1} if sampling depth was 20 cm.

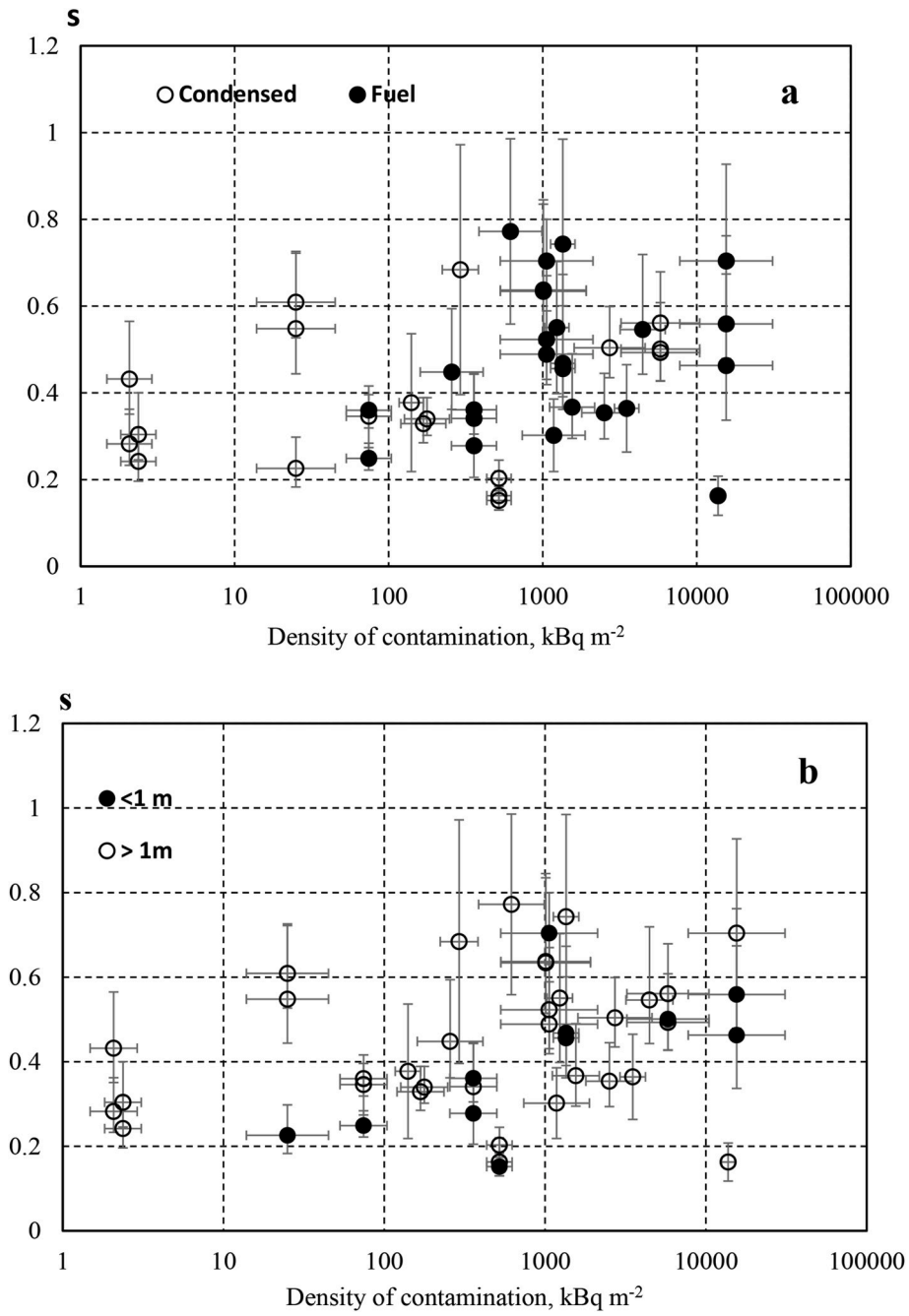

Fig. 4. Variation in standard deviation of logarithms of soil contamination density of ^{137}Cs with the deposition density and the size of the sampling increment. The vertical and horizontal bars are the errors of the standard deviation of the logarithm of ^{137}Cs soil contamination density and the geometric standard deviations of ^{137}Cs soil contamination density, respectively.

When undertaking field sampling, selected samples of soil should be statistically independent and representative for the contaminated sites. Statistical independence between samples can be achieved by taking samples with sufficiently large distances between samples to avoid a correlation in the radionuclide concentrations in the samples. This can typically be assessed from a semi-variogram or variogram analysis. Statistical independence can be achieved at distances beyond which the maximum semi-variance or variance is reached, i.e. the point at which the variogram becomes constant. Samples for ^{137}Cs concentration in soil, including composite samples consisting of several closely related samples, taken by a sampler with working area of 0.001–0.01 m^2 are considered to be statistically independent if collected more than 1 m apart (Khomutinin et al., 2001).

Based on these results the minimum number of samples that guarantees the required (predefined) precision of the GM estimate for a given dispersion of radionuclide contamination density may be calculated. The theory of interval estimates of unknown parameters was used (Kendall and Stuart, 1973) to approach this issue. In case of the normal distribution of a random variable X with parameters m and σ, where the σ value is known, the statistics U (Eq. (9)), follows the standard normal probability distribution with zero expectation and the standard deviation equal to one, i.e., *N (0; 1)*.

$$U = \frac{\overline{X} - m}{\sigma} \cdot \sqrt{n} \tag{9}$$

where, $\overline{X}$ is the sample mean; m is the expectation of the distribution; σ is

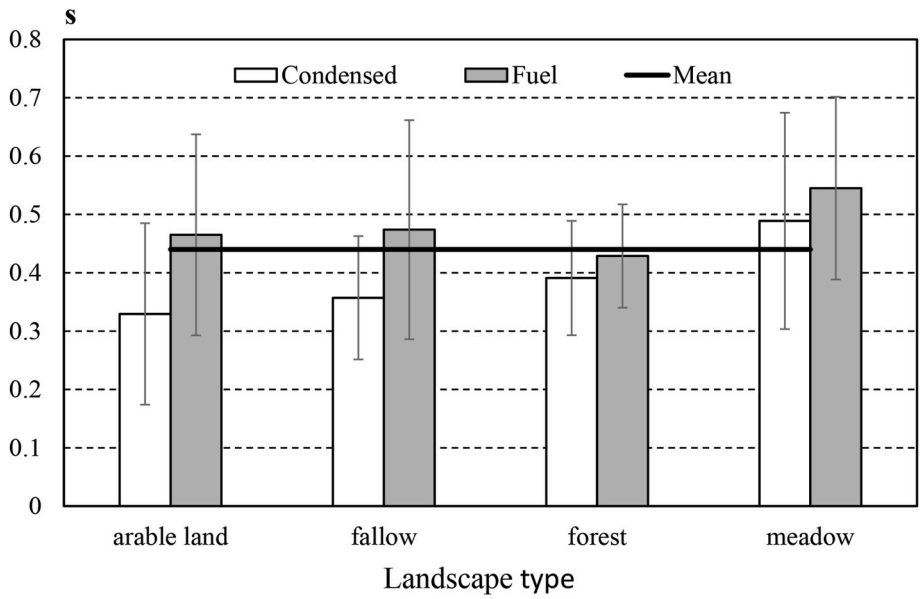

Fig. 5. Variations of standard deviation of the logarithms of soil contamination density of 137Cs when the sampling area is S = 0.001 m2 depending on type of fallout and land use. (mean ± SD).

the standard deviation of the distribution; n is the sample size.

Applying this statistical deduction to the logarithm of the contamination density, we can find the confidence interval where the GM of the density of contamination is, with the confidence level of p subordinates to the following inequality.

$$exp\left(\overline{\mu} - \frac{U_p \cdot s}{\sqrt{n}}\right) \le GM \le exp\left(\overline{\mu} + \frac{U_p \cdot s}{\sqrt{n}}\right) \quad (10)$$

U_p is the quintile of the normal distribution corresponding to the level p. This relationship allows to find the minimum number of soil samples required to determine the GM of the contamination density with a given relative error for the confidence probability level of p:

$$n \ge s^2 \cdot \left\{\frac{U_p}{\mathrm{In}(1+\delta)}\right\}^2, \quad (11)$$

where $\delta = exp\left(\frac{U_p \cdot s}{\sqrt{n}}\right) - 1$ – is the upper level of relative error of the GM of the contamination density.

The choice of an appropriate sampling design is a crucial step in the sampling planning for radiological assessments. The soil sampling can rely on both single (point) and composite samples. In this study, a single sample was one soil core, or a uniform array of soil cores collected from one specific location (within 0.2 m radius of the grid node) within the sampling site. A composite soil sample consisted of several single samples that were merged to create one composite sample for the further analysis. The point samples, forming composite sample, were collected at such a distance between samples that allows the assumption that activity concentration of radionuclides in the samples were statistically independent. Usually, the merging of point samples to obtain composite samples may improve soil sampling representativeness, if there is no strong contamination gradient.

Data on variability of the radionuclide deposition density obtained in this study were used to assess the number of samples to be taken required to fit to the acceptable uncertainty level. Using the interval estimate (Eq. (11)) a two-dimensional diagram was designed (Fig. 6). The nomogram allows the estimation of the number (n) of required single samples to obtain a geometric mean value of soil contamination density of ^{137}Cs on a uniformly contaminated site with a specified relative error. Vertical bars represent possible errors of n for confidence interval of 68%. Variability of the standard deviation of the log of soil density contamination results the error of the parameter n.

Thus, if predefined relative error of the GM of the soil contamination is $\delta_{\gamma=0.95} = 25\%$ and relative error of measuring ^{137}Cs activity of sample $\delta_{meas.} \le 10\%$, it is required to take 12 composite samples with sampler of working area of 0.001 m^2 (Fig. 6a) or 5 composite samples with area 0.005 m^2 (Fig. 6b). If only 3 samples with sampling area 0.001 m^2 or one sample with sampling area 0.005 m^2 were taken, then for any precision of the radionuclide measurements, the relative error of assessment of geometric mean contamination density ($\delta_{\gamma=0.95}$) is 50% (Fig. 6).

It was recently reported (Khomutinin et al., 2001) that when 5 independent samples, each of sampling areas of 0.001 m^2, are collected on a uniformly contaminated site and analysed, there is only a small difference in the standard deviation of the log of soil contamination density when compared with a combined or single composite sample (sampling area is 0.005 m^2).

As a result of these findings, the official standard of Ukraine for the determination of radioactive contamination by soil sampling is to use a combined soil sample of 5 single samples, where the distance between point soil samples at the test site should be at least 1 m. In this case, if a relative measurement uncertainty does not exceed 10% at the ±2σ level, the relative error of the geometric mean assessment of contamination density on uniformly contaminated sites does not exceed 30% (Khomutinin et al., 2001).

The numbers of soil samples required to estimate the geometric mean of the density of radioactive contamination of soil with predefined uncertainty is presented in Table 4. The predicted number of samples depends on the values of $\delta_{\gamma=0.95}$ (the relative error of geometric mean of soil density contamination) and δ_{meas} (the relative uncertainty of measurement of radionuclide activity in a sample of soil at 95% confidence level). Table 4 shows how the estimate on the contamination density is a function of the number of samples and the measurement uncertainty.

Thus, for relative uncertainty of measuring ^{137}Cs activity of sample $\delta_{meas.} \le 20\%$, if 5 composite samples with area 0.005 m^2 were sampled, we will have relative error of geometric mean of soil contamination estimation $\delta_{\gamma=0.95} = 20\%$ (Table 4).

The more samples are taken from the sampling site, the more accurate and reliable are estimates the contamination density. However, a detailed sampling campaign involves a large volume of samples and requires a high cost. Additionally, excessive accuracy of estimates of contamination density is not normally required, although the overall uncertainty (δ) should ensure the conclusions made based on these data.

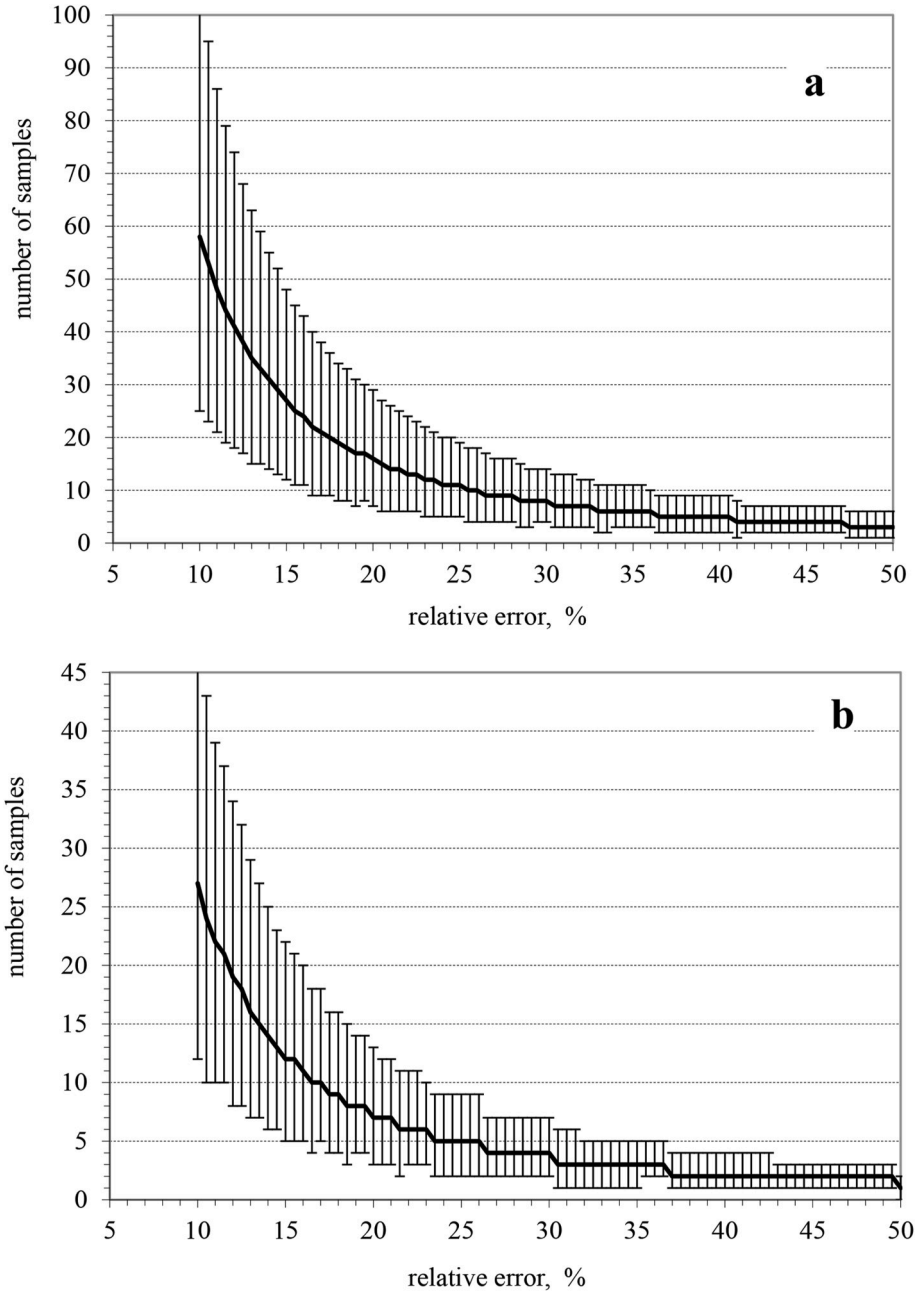

Fig. 6. Nomogram for calculation of the number of the single samples required to obtain a geometric mean of soil contamination density of ^{137}Cs on a uniformly contaminated site with a specified relative error, with uncertainties of measurement of the radionuclide activity in soil samples ≤10% (p = 0.95): a – $s = 0.044 \pm 0.15$ for sampling areas 0.001 m^2; b - $s = 0.030 \pm 0.10$ for sampling areas 0.005 m^2.

Therefore, determination of the minimum number of sampled samples that provide the specified uncertainty (δ) of estimating the contamination density is highly valuable. Conversely, having information on the number of soil samples, it is important to know what uncertainty in assessments of the contamination density can be anticipated with the allocated funding for the sampling campaign. The proposed approach for optimising the number of the soil samples taken for monitoring of the contaminated soils allows proper planning of sampling at contaminated sites.

4. Conclusions

The ^{137}Cs contamination density of uniformly contaminated sites, located on both in the Chernobyl accident affected areas and territories contaminated only by the global fallout may be satisfactorily approximated by the log-normal probability distributions. The standard deviation of the logarithms of ^{137}Cs contamination density (s) is not dependent on the mean contamination density, the type of radioactive deposition and the landscape features. The mean values of standard deviation of the logarithms of the ^{137}Cs contamination density tended to be reduced with an increase of the sampling area up to 0.015 m^2. Based on these findings the number of soil samples required to achieve a predefined uncertainty for estimating the soil contamination density was obtained. The approach and methods outlined in the paper allow planning the number of sampled samples and conducting radiological monitoring of the soils contaminated with ^{137}Cs, ^{90}Sr and ^{239,240}Pu with optimal cost of the sampling campaign. These methods are not only applicable on sampling in areas affected by the Chernobyl accident. They can be also used in some other contamination sceneries; such as soil contamination by nuclear accidents or assessment of the

Table 4
Required number of soil samples to assess the GM of radioactive contamination density at the sample site.

Sample type	S, m^2	Relative uncertainty of measuring ^{137}Cs activity, $\delta_{meas.}$, %			
		10	20	30	40
Relative error of geometric mean of soil contamination estimation $\delta_{\gamma=0.95} = 10\%$					
Single sample	0.001	64	66	70	75
	0.002	54	56	60	65
	0.003	50	52	56	61
	0.004	48	51	54	59
	0.005	47	50	53	58
Composite sample	0.003	21	23	27	32
	0.004	16	19	22	28
	0.005	14	16	20	25
Relative error of geometric mean of soil contamination estimation $\delta_{\gamma=0.95} = 20\%$					
Single sample	0.001	18	18	19	21
	0.002	15	16	17	18
	0.003	14	15	16	17
	0.004	14	14	15	17
	0.005	13	14	15	16
Composite	0.003	6	7	8	9
	0.004	5	5	6	8
	0.005	4	5	6	7
Relative error of geometric mean of soil contamination estimation $\delta_{\gamma=0.95} = 30\%$					
Single sample	0.001	9	9	10	10
	0.002	8	8	8	9
	0.003	7	7	8	9
	0.004	7	7	8	8
	0.005	7	7	7	8
Composite sample	0.003	3	4	4	5
	0.004	3	3	3	4
	0.005	2	3	3	4
Relative error of geometric mean of soil contamination estimation $\delta_{\gamma=0.95} = 40\%$					
Single sample	0.001	6	6	6	6
	0.002	5	5	5	6
	0.003	4	5	5	5
	0.004	4	5	5	5
	0.005	4	4	5	5
Composite sample	0.003	2	2	3	3
	0.004	2	2	2	3
	0.005	2	2	2	2

contamination of radiation legacy sites.

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