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Evaluation of optimal number of soil samples for detail reconstruction of initial field of ^{137}Cs fallout in Chernobyl affected areas

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Abstract

A Chernobyl-derived ^{137}Cs- fallout was associated with one or two rainfalls Because of that vast areas of the Europe affected by Chernobyl-derived fallout are characterized by non-uniform field of radionuclide contamination. It was assessed after detailed field investigation within few river basins of the Central Russia located in areas with different levels of Chernobyl contamination, that existing maps of radionuclide contamination composed during last two decades are not enough detailed for assessment of initial contamination field transformation by the lateral migration processes of the Chernobyl-derived ^{137}Cs. This problem can be overcomed if additional soil sampling are undertaken in reference locations for correction of exiting radionuclide contamination maps. However it is necessary to evaluate the optimal number of bulk samples which should be taken in each sampling point for receiving statistically correct results of radionuclide concentration. Special investigation was undertaken in few catchments (S= 2-50 km^2) of the Central Russia, located in areas with different levels of initial Chernobyl contamination, for evaluation the optimal number of samples, which should be taken in each sampling point for the determination of Cs-137 concentrations error not exceed 30 % on 95 % confidence level..

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Introduction

Radioactive contamination is believed to be one of the most dangerous consequence of human`s activity. Hence definition of contaminated area and registration of contamination level become essentially important tasks in ecological framework (Dercon et al. 2012). Chernobyl`s disaster in 1986 was the largest man-triggered incident in a history of 20-th century. A derived Cs-137 fallout affected vast territories of Europe and partly the west of Asia (De Cort, 1998). The structure of emerged field contamination can be characterized as a very non-uniform phenomenon. In general distribution of radionuclides within vast regions was determined by processes of tropospheric circulation. That is why two the most “pollution tracks” were formed on the territory of the East Europe (Izrael et al. 1995). Despite this regional trend was assessed in the first years after accident, the problem of large-scale heterogeneity of contamination remains actual. This situation is additionally complicated by lateral migration of radionuclides mainly caused by soil

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erosion. Therefore the knowledge about magnitude of Cs-137 concentration's fluctuation is crucial for works dedicated to relatively small areas. Unfortunately existing radioecological maps is not enough detailed for high-precision investigations. Hence it is necessary to overcome this problem based on additional sampling program.

Sampling program should be elaborated based on exiting knowledge about radionuclide contamination of study catchment, as well as relief features. At the same time obtained data should be statistically reliable. In case of limited resources and time investigator is faced with difficult dilemma. From one hand investigation requires large number of samples to assess local variability of Cs-137 concentrations but on the other hand there is an obvious need for registration of more detailed trend of initial fallout within whole studied area. The way to solve this problem is to find an optimal number of soil samples which can be quite enough for definition of systematic and random spatial variations.

As it mentioned by group of authors (Owens and Walling, 1996; Quine, 1989; Sutherland, 1991; Walling and He, 1999) the correct assessment of Cs-137 inventories for the reference sites is a key for investigation of radionuclide redistribution or lateral migration along the slopes or the entire catchment. One of the main problems for all assessments in is taking into account a spatial variability of Cs-137 inventories and calculating of its mean values. Furthermore, in case of intensive Chernobyl contamination the fallout of radionuclides was associated with a single rainfall event, or to the prevailing winds in case of dry fallout deposition (Golosov, 2003).

Each reference site is supposed to content enough sampling points for correct evaluation of random (local) variations. This variations hypothetically depend on the intensity of contamination. For example the highest variability was registered in 100-km^2 zone around Chernobyl power plant with deposition level of more than 1500 kBq/m^2. Taken samples from area of 100 m^2 showed variations of as much as 3- to 10-fold (Izrael, 1990, 1996). The variability of Cs-137 of interfluve uneroded geomorphological units is represented in Table 1 (Golosov et al., 1999a, 1999b).

Table 1. The variation of contamination level for different geomorphologic units.

Contamination level, kBq/m^2	Geomorphological unit	Coefficient of variation, %
40 – 1000	Interfluve	25
5 – 40	Interfluve	47
< 5	Upper part of cultivated slope	20.5

As declared by Sutherland the correct evaluation of Cs-137 activity at 95% confidence level with an allowable error of ±10% should not be less than 12 (Sutherland, 1991). The inventories of reference sites are believed to be constituted due only to one random process – rainfall.

Following investigation were undertaken to figure out the optimal number of samples desired to make correct assessments of ^{137}Cs fallout with low spendings of resources and time. The main emphasis was made on empirically observed concentrations of radionuclides in Chernobyl-affected areas.

Material and Methods

The objects of the study were four catchments in Chernobyl-affected area within East-European Plain (Figure 1). All chosen catchments are located in the steppe or forest-steppe zones and have different contamination level in range7,5 - 356 kBq/m^2. The relief of all chosen catchments is characterized by upland with a very dense network of gullies and valleys. The interfluve slopes of catchments are rather gentle with typical length 400-500 meters. The slope gradient of surface is rarely exceeds 5^o. Every studied catchment has area less than 50 km^2 and at the moment fallout the most part of their area was arable lands. Cs-137 fallout was mostly mixed in cultivated layer of soil. Below cultivated layer 137Cs concentration instantly decreases to incomparably low value. Therefore during further investigation only cultivated layer (30 cm from surface) was examined.

The process of sampling was undertaken by conducting reference sites on interfluve positions where erosion rates are very low or process of erosion is actually absent. 12 samples were taken in each reference location. Every sample was taken in such way that all thickness of contaminated layer was less than or equal to the lower limit of sampling. In this case such depth was 30 cm – the thickness of ploughing layer of soil where all provided radionuclides were mixed.

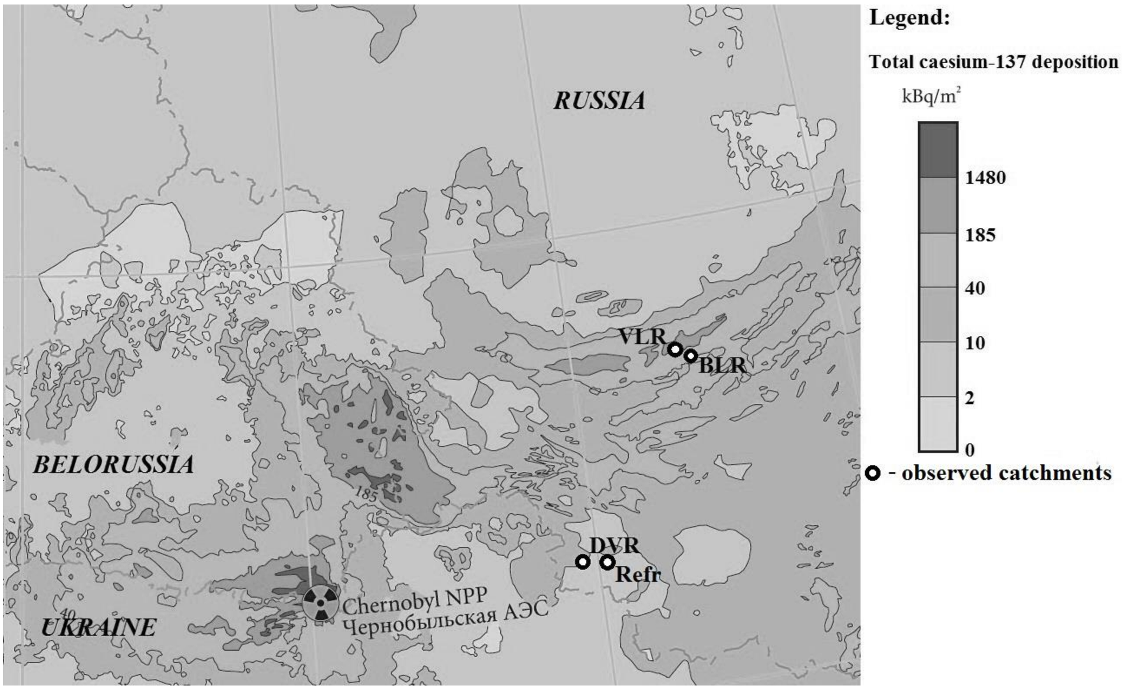

Figure 1. Location of observed catchments (De Cort, 1998).

Subsequent laboratory processing of the 137Cs samples involved oven-drying at 105°C, grinding, sieving to<2mm and homogenization of sub-samples for gamma-analysis. The 137Cs activity was measured at 661.66 keV, using a high-resolution, low-background, hyperpure germanium coaxial gamma-ray detector. Count times were typically in the range of 0.5–24 h depending on 137Cs concentration in each individual sample, providing a maximum relative error of the activity determination of _5–10%. Sample preparation, treatment and 137Cs activity measurements were carried out at the Laboratory of Soil Erosion and Fluvial Processes, Faculty of Geography, Lomonosov Moscow State University.

The mean value calculated from twelve points was believed to be reliable (Sutherland, 1991). Another question has been araised: is it possible to use less number of sampling points to get close to given value? Hence three hypotheses were suggested: how close will be assessment based on 3, 4 and 5 random sampling points to mean value, which was received based on 12 sampling points for each reference location. It was analyzed the whole number of combinations for corresponded number of samples taken from twelve-point selection. The volume of observed data is determined by combinatorial formula (Table 2).

Table 2. Number of 3,4 and 5-sample combinations

Number of samples	Number of combinations
3	220
4	495
5	792

$$C_r^n = \frac{n!}{r!(n-r)!} \qquad (1)$$

C_r^n – number of combinations, n – number of sampling points, r – number of points taken for averaging.

When the desired combinations were obtained the calculation of difference between mean value of each combination and mean value of 12 samples were conducted. Than the ratio of taken difference to the mean value of 12 samples were found and converted to percent. This final value was argued as a inaccuracy of assessment made on corresponded number of samples.

The estimation of confidence level was conducted as calculation of fate corresponded to combinations which wean value was less than definite mistake (Figure 2).

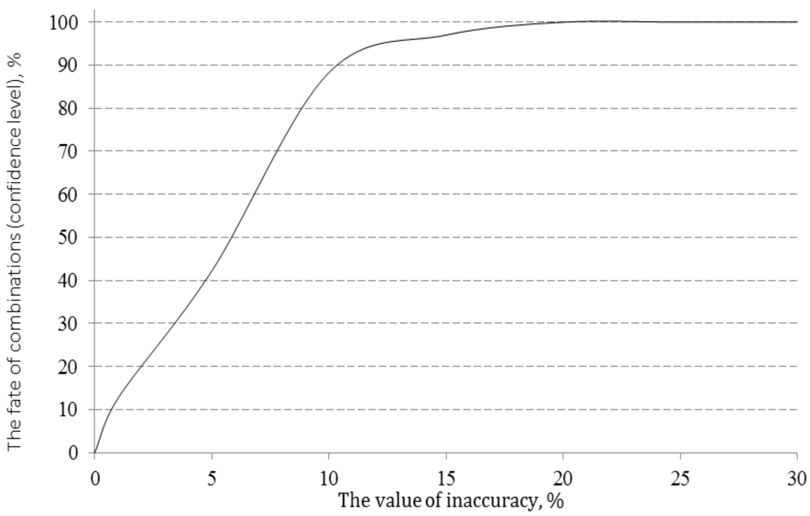

Figure 2. The cumulative curve of the value of inaccuracy on the reference site VLR-1. Assessment was made with 4-sample averaging.

After that all the values of inaccuracy with corresponded confidence level were put into the table and analyzed.

Results and Discussion

Studied reference sites were divided into groups according to level of contamination. Therefore three groups were distinguished: with level of 0 – 10, 10 – 100 and >100 kBq/m^2 (in re-calculation for the May of 1986).

Area with level of radionuclide contamination 0 – 10 kBq/m^2.

In this group four reference sites were used for analysis. The results are presented in Table 3.

Table 3. The results of combination calculations with corresponded value of inaccuracy for the reference sites DVR-1–4, %.

	The value of inaccuracy, %						
	>1	>5	>10	>15	>20	>25	*>30*
				3 samples			
DVR-1	8,7	33,8	57,5	78,1	93,2	98,2	99,1
DVR-2	7,3	44,3	78,1	95,4	99,1	99,5	99,5
DVR-3	13,2	50,7	85,4	98,6	99,5	99,5	99,5
DVR-4	8,9	50	89	99	99,5	99,5	99,5
				4 samples			
DVR-1	7,9	40,0	71,3	90,7	98,4	99,8	99,8
DVR-2	9,7	51,3	87,1	99,2	100,0	100,0	100,0
DVR-3	15,2	62,8	94,5	100,0	100,0	100,0	100,0
DVR-4	13,9	62,2	94,3	100,0	100,0	100,0	100,0
				5 samples			
DVR-1	8,7	46,6	79,9	95,7	99,6	100,0	100,0
DVR-2	13,8	60,7	93,9	100,0	100,0	100,0	100,0
DVR-3	17,9	72,5	97,9	100,0	100,0	100,0	100,0
DVR-4	16,8	70,7	98,5	100,0	100,0	100,0	100,0

As it can be seen from Table 4 in the majority of cases (more than 95%) all values don`t exceed deviation of 20 %. This fact urges to declare that the precision of 20-% inaccuracy and 95% confidence level for this particular catchment can be achieved if only 3 samples were taken. In case of inaccuracy of 15 % 5 samples are required for averaging. It is interesting to mention that almost all conducted assessments don`t have value of inaccuracy of 30 %.

Area with level of radionuclide contamination 10-100 kBq/m^2.

Six reference site linked to two catchments with corresponded level of contamination were evaluated in this group (Table 4).

Table 4. The fate of combinations with corresponded value of inaccuracy for the reference sites BLR-1–3, Refr-1–3, %.

	The value of inaccuracy, %						
	>1	>5	>10	>15	>20	>25	>30
				3 samples			
BLR-1	8,2	39,3	90,4	99,5	99,5	99,5	99,5
BLR-2	19,2	86,3	99,5	99,5	99,5	99,5	99,5
BLR-3	8,2	39,3	79,5	99,1	99,5	99,5	99,5
Refr-1	16,9	73,5	98,2	99,5	99,5	99,5	99,5
Refr-2	2,7	21,9	42,0	64,8	78,1	87,2	95
Refr-3	8,7	46,6	79,5	94,5	99,1	99,5	99,5
				4 samples			
BLR-1	17,0	70,1	97,2	100,0	100,0	100,0	100,0
BLR-2	24,4	93,7	100,0	100,0	100,0	100,0	100,0
BLR-3	7,1	41,2	85,7	100,0	100,0	100,0	100,0
Refr-1	16,0	84,0	100,0	100,0	100,0	100,0	100,0
Refr-2	6,1	24,8	51,9	71,7	87,9	96,8	99,6
Refr-3	12,1	56,0	88,3	99,2	100,0	100,0	100,0
				5 samples			
BLR-1	19,2	77,9	99,1	100,0	100,0	100,0	100,0
BLR-2	33,5	98,4	100,0	100,0	100,0	100,0	100,0
BLR-3	5,4	39,6	90,5	100,0	100,0	100,0	100,0
Refr-1	16,9	93,7	100,0	100,0	100,0	100,0	100,0
Refr-2	4,7	29,4	58,1	82,1	95,6	99,9	100,0
Refr-3	13,8	62,8	94,2	99,9	100,0	100,0	100,0

The values represented above tell us that usage of 3-samples averaging is quite enough for estimation with value of inaccuracy less than 30 % with probability more than 95 %. Usage of 4 samples can improve precision of assessment to 25 % inaccuracy. 5-samples averaging propose to have inaccuracy of 20 % with confidence level more than 95 %.

Area with level of radionuclide contamination >100 kBq/m².

The group of the highest level of radionuclide contamination consists of six reference sites located within the one catchment (Table 5).

Table 5. The results of combination calculations with corresponded value of inaccuracy for the reference sites VLR-1–6, %.

	The value of inaccuracy, %						
	>1	>5	>10	>15	>20	>25	>30
				3 samples			
VLR-1	11,0	40,2	76,3	91,3	96,8	98,6	98,6
VLR-2	15,1	69,4	97,7	100,0	100,0	100,0	100,0
VLR-3	5,0	30,1	54,3	75,8	88,6	95,4	99,1
VLR-4	9,1	41,6	74,0	90,9	98,6	100,0	100,0
VLR-5	8,7	42,0	76,3	93,6	99,1	100,0	100,0
VLR-6	8,7	36,5	63,0	85,4	93,2	98,2	98,2
				4 samples			
VLR-1	12,7	42,2	88,3	97,0	100,0	100,0	100,0
VLR-2	15,8	77,6	99,8	100,0	100,0	100,0	100,0
VLR-3	6,9	34,5	64,4	84,6	95,4	98,6	100,0
VLR-4	10,9	50,3	82,0	97,4	100,0	100,0	100,0
VLR-5	9,7	49,3	86,5	98,8	100,0	100,0	100,0
VLR-6	11,1	35,8	72,7	94,9	100,0	100,0	100,0
				5 samples			
VLR-1	19,2	55,6	88,6	100,0	100,0	100,0	100,0
VLR-2	23,2	87,6	100,0	100,0	100,0	100,0	100,0
VLR-3	9,3	41,3	74,1	91,3	98,6	100,0	100,0
VLR-4	12,2	56,8	90,8	99,7	100,0	100,0	100,0
VLR-5	11,4	57,2	93,6	100,0	100,0	100,0	100,0
VLR-6	7,8	44,3	85,9	99,1	100,0	100,0	100,0

According to the Table 5 above 3-samples averaging is suitable for assessments with inaccuracy of 25 % and confidence level more than 95%. 4 and 5-samples averaging allows to receive values which inaccuracy doesn`t exceed 20 % and confidence level is higher than 95 %.

In general required number of samples for assessment of different precision is presented in Table 6.

Table 6. Required number of samples for assessments mean values of radionuclide contamination in reference locations with different precision (confidence level 95 %).

Level of contamination, kBq/m^2	Value of inaccuracy, %		
	>20	*>25*	*>30*
0-10	4	3	3
10-100	4	4	3
>100	4	3	3

It was found that four samples is an optimal number for assessments with value of ^{137}Cs inventory in reference locations with inaccuracy less than 20 %. Taking into account that considered coefficient of variation of ^{137}Cs inventories is close to 20 % estimations made with mentioned precision can be approved.

Triple sampling is suitable only in case of rough estimations when acceptable value of inaccuracy doesn`t exceed 30 %. Fivefold sampling is believed to be surplus on two grounds. The first one is that assessment made with value of inaccuracy higher than 20 % can be conducted less number of samples. Obtained data also performed that collection of five samples can`t the precision of assessment with confidence level of 95 %.

Conclusion

1. Field of radionuclide contamination of ^{137}Cs can be examined with a sampling of limited number of samples in reference locations. Acceptable value of inaccuracy should not exceed 20 %. The optimal number of samples is 4. Reduction of samples number lead to decreasing precision and reliability of assessment. Averaging made with 5 samples is not effective. Number of samples should be considerably increased if investigation requires much higher precision of estimation.

2. It was shown that sampling of small volume are suitable for rough estimations. In case of high precision survey they can be used to figure local particularities of ^{137}Cs fallout and to arrange sampling sites of bigger volumes.

3. There is still no absolutely precise spatial pattern of ^{137}Cs fallout. It`s impossible to create it in analytical way and at the same time take into account all possible factors and particularities. The only they to improve our knowledge is collecting more new empirical data and it statistical analysis. Small volume sampling is one the resource-saving way to obtain desired information.

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