Khomutinin Y.V. , Kashparov V.A., Zhebrovska K.I.

SAMPLING OPTIMISATION
WHEN RADIOECOLOGICAL MONITORING

2004

The results of researches conducted by UIAR during elimination of consequences of catastrophe on ChNPP, concerning basic and urgent problem of radioecological monitoring - representative sampling of soil, plants and milk, and also measurement of samples having an essential volumetric heterogeneity (hot particles) are generalized. The questions of optimum planning of selection representative samples for the samples of the soil, plants and milk are considered. The methods of account of minimum necessary number of samples excerpt for an estimation of average of the controlled quantity with the given relative error are offered.

The book is designed for the scientific employees, radioecologists, post-graduate students and students interested in problems of the environment radioecological monitoring .

ISBN 966-646-034-3

Contents

SYMBOLS USED IN THE TEXT

$N(m;\sigma)$ - The normally distributed random variable with average value m and variance σ^2;

$\bar{x}$ – Sample average for $N(m;\sigma)$;

s^2 – Sample variance for $N(m;\sigma)$;

σ_{av}^2 – Variance of sample average for $N(m;\sigma)$;

n – Amount of sampling;

p – Confidence probability;

$U(p)$ – Fractile of normal law of distribution at the p level;

$t(p,n-1)$– Fractile of the Student probability distribution at the p level with n-1 degree of freedom;

σ_{ln} – Standard deviation of the random variable with logarithmically normal distribution;

C_{so} – Soil density contamination with radionuclide on non-gradient site;

$\bar{C}_{so}$– Average value of soil density contamination with radionuclide on non-gradient site;

$Me_{C_{so}}$ – Median of soil density contamination with radionuclide;

$W_{C_{so}}$ – Coefficient of variation of soil density contamination with radionuclide on non-gradient site;

μ_{so} – Average log of density contamination with ^{137}Cs on non-gradient site;

s_{so} – Standard deviation of log of density contamination with ^{137}Cs on the site;

$s_{1,so}$ Standard deviation of log of density contamination with ^{137}Cs on non-gradient site for sampling by the sampler in 3.7cm diameter (1 injection);

$s_{5,so}$ Standard deviation of log of density contamination with ^{137}Cs on non-gradient site for sampling by the sampler in 3.7cm diameter (5 injections);

$\Delta^-(\Delta^+)$ The lower (upper) absolute error;

$\delta(\delta^+)$ The lower (upper) relative error;

C_p – The specific ^{137}Cs content in plant sampling on non-gradient site;

μ_p – Average log of the specific ^{137}Cs content in plant sampling on non-gradient site;

s_p – Standard deviation of log of the specific ^{137}Cs content in plant sampling on non-gradient site;

δ_{inst} – Ratio error of measurement of radionuclide content in sample at ±σ level;

s_{inst}^2 Variance of log of specific activity of sample stipulated by error of measurement;

$Tf_{so,p}$ – Transfer factor in chain "soil - plant";

Tf_{me} – Median of transfer factor in chain "soil - plant";

k – Average value of log of ^{137}Cs transfer factor in plants;

s_k – Standard deviation of log of ^{137}Cs transfer factor in plants;

$r_{so,p}$ – Coefficient of correlation between log of soil density contamination with ^{137}Cs and log of ^{137}Cs content in plants;

$C_m(t)$ – The specific ^{137}Cs content in milk on personal small farms in the settlement at the moment t;

$\overline{C}_m(t)$ – Average value of the specific ^{137}Cs content in milk on PSF at the moment t during the year;

$\overline{C}_m^{Cs}$ – Average annual value of the specific ^{137}Cs content in milk on PSF in the settlement;

$\mu_m(t)$ – Average value of log of the specific ^{137}Cs content in milk on PSF at the moment t;

$s_m(t)$ – Standard deviation of log of the specific ^{137}Cs content in milk on PSF at the moment t;

$s^2_{n.sa.}$ – Variance of log of the specific activity of sample, stipulated only by volumetric heterogeneity;

$s^2_{sum.sa.}$ – Variance (common) of log of specific activity of sample stipulated by all random factors;

$s^2_{n.sit.}$ – Variance of log of soil density contamination stipulated only by microheterogeneity of contamination on the site;

INTRODUCTION

The unprecedented nuclear accident in the history of humanity, which happened in 1986 on the 4-th block of Chernobyl NPP, caused radioactive contamination of vast territories in different European countries. The most contaminated with radioactive fallout countries are Belarus, Russian Federation and Ukraine. Two types of fallout represent the generated traces of radioactive contamination: a condensation component of volatile highly mobile fission product (represented practically everywhere) and fuel component (concentrated mainly in the 30-km zone of Chernobyl NPP). [1, 2, 3, 4].

Environmental contamination with radioactive particles (hot particles) has become one of the distinctive features of Chernobyl fallout. They are finely dispersed nuclear fuel (fuel particles) or initially inert aerosol particles that turned into condensation centres of volatile highly mobile radioisotopes (condensation particles) during the accident. The fuel particles had fallen in the 30-km zone Ch NPP mainly, but they also had been detected on a large distance from the accident: in Finland, Sweden, Norway, Lithuania, Poland, Germany, Czech, Austria and other countries. [5,6,7,8, 9,10,11].

Since the first days after accident, the behaviour of this radioactive fallout in the environment has been permanently researched and analysed. The radioecological monitoring of the contaminated territories has been carried out. More detailed radioecological researches of different environmental objects have been conducted; the various scientific researches connected with sampling of objects, contaminated with radionuclides have been executed.

The theoretical fundamentals of radioecological monitoring explicitly are represented in the literature, for example, in the publications of Izrael Yu. [12]. Though approaches, stated in the literature are general, they are oriented on contamination of environment at the same time because of the nuclear - fuel cycle enterprises and as a result of global fallout caused by the nuclear weapon tests. The levels of contamination of agricultural products for this radioactive fallout, as a rule, are not high and do not exceed permissible levels. Chernobyl accident has required solving deeper and nicer problems. The vast territories of developed agricultural production have been contaminated with radioactive fallout. The society had to solve the problem of agricultural production on the territory, contaminated with radionuclides. Wide punctation of radioactive Chernobyl fallout is one more feature, which has involved detail inspection of the territories and reveal of radioactive contamination spots. It also demands the evaluation of contamination levels of defined agricultural grounds, agricultural production, and forage for domestic animals. It requires to detailing radioecological monitoring for defined fields, agricultural grounds and products, groups of animals, as its objects. The past emergency experience has shown that a frequent necessity in realising more detail radioecological researches of environmental objects for solving defined tasks in addition to general radioecological monitoring. The realisation of mass radiological control and monitoring of agricultural products and foodstuff is also necessary under conditions of radioactive contamination of vast territories with developed agricultural production. In order to solve all listed problems it is necessary to increase the number of sampling and measurements greatly and to invest great amounts of money into radioecological monitoring as a whole and for individual radioecological researches of the environmental objects. In this connection the problem of optimisation of soil, plants, agricultural products sampling and spectrometric (radiometric) measurements volume has become actual. The works in this direction were begun in 90 and have been proceeding now in UIAR [13,14,15,16,17] the experimental researches are conducted as in the 30-km Zone of Ch NPP as beyond the bounds.

The source experimental information is the basis of any monitoring, including radioecological. Direct observations and measurements for researched object (in situ) or selected samples of researched object are source of this information. The selected samples with any reliability describe actual contamination with radionuclides of researched object. That property of sample population (a sampling) is called the representativeness concerning radioactive contamination of the investigated object. Depending on the representativeness of a sampling the real contamination with radionuclides of the object can be evaluated with any error. Thus the quality and reliability of radioecological monitoring as whole and different radioecological researches of environmental objects are determined by representativeness of the selected samples totality.

It is necessary to know about a source of radioactive contamination, physical-chemical characteristics of radioactive fallout and particularities of radionuclides migration in the environment to realise representative sampling for investigation of different objects of the environment, products of industrial and agricultural activity. These factors have an effect on representativeness of selected soil samples totality especially when radiological inspections of territories contaminated with radionuclides.

The availability of fuel particles in the soil and their consequent dissolution with a different velocity, even on small sites makes soil contamination with radionuclides to extremely inhomogeneous [18, 19, 20]. Microrelief non-uniformity and radionuclides redistribution under effect of biogenic factors essentially influence non-uniformity of selected soil samples contamination with radionuclides.

The fallen out radionuclides migrate deeply into the ground. The intensity of migration is determined by chemical properties of an element, physical-chemical properties of fallout, landscape and soil-climatic particularities (when the man-caused activity is absent) [21]. The radionuclides are uniformly mixed in arable stratum of soil and during the time migrate into subsoil horizon. The neglect of the factor of vertical migration will cause serious errors when evaluating density contamination of the territory with radionuclides.

It is necessary to distinguish two kinds of representativeness: physical and statistical. Correct technique for sampling of single sample within the time and space (for example, it is account of vertical migration of radionuclides when evaluating soil density contamination; it is account of seasonal dynamics when evaluating milk contamination in settlements) determines the physical representativeness of sample. The number of the selected samples (sample volume) and statistical variability of contamination of researched object with radionuclides determine statistical representativeness of sample. However in practice variability (variance) of estimated magnitude is very seldom known a priori. Therefore, the average value or the median and appropriate error is usually evaluated according to experimental data with a prescribed confidential probability. Then the error can be very large that requires additional measurements. To describe the quality of the executed researches, the investigators frequently point out only measurement error of a single sub-sample taken from sample and extend it on the value that describes the whole researched object (density contamination of territory, specific content etc.), which leads to underestimating the true error of investigated magnitude definition and also to incorrect conclusions concerning the researched phenomenon. The presence of fuel particles in the selected soil sample may cause grave errors when measuring its activity. For example, the γ-spectrometric measurement outcome can vary in the range of order of measured magnitude depending on fuel particle position in measuring flask and on flask geometry. When we use small sub-samples for radiochemical analyses, there is a probability for isolated fuel particles to get into a measured sub-sample. In this case the activity value of a sub-sample may not correspond to the activity value of the whole sample.

A lot of techniques and recommended practices for inspecting and evaluating of radioactive contamination of the environment [22,23,24,25,26,27,28] have been developed and offered since liquidation of the consequences of the accident on the Chernobyl NPP. But, despite knowledge accumulated in this area, the problems on optimisation and representativeness of soil sampling are still actual (in particular when specify density contamination of definite sites or grounds with radionuclides).

In connection with generalisation of monitoring information and consequent acceptance of any solutions, a problem on the representativeness of obtained outcomes also appear. Therefore an important part of radioecological monitoring is

- providing with sampling of representative population of samples which describe researched object;
- maintenance of the right sample-processing mode before the measuring (the process of sample preparation);
- reliability of measuring outcomes.

For example, the accuracy of definition of the radionuclides content in researched object and the possibility of averaging of some parameters depend on the representativeness of samples for measuring (the amount of samples, sample mass, square, depth and pitch of sampling) for correct evaluation of density contamination of territory, specific soil or vegetation activity, radionuclides concentration in air etc. The presence of random factors, such as the presence of "hot" particles in the samples (particles with anomalous high activity) may cause grave errors when defining parameters and when extrapolating measuring outcomes from the researched object. The problem of representative sampling is especially actual for complex radioecological monitoring, which is the basis to calculate irradiation of the inhabitants in the settlements located on the territory that underwent radioactive contamination after the accident on Ch NPP [29].

The problem of optimisation of the amount of samples and measurements to evaluate necessary parameters of the environmental objects contamination with a specified error when costs are minimum becomes actual taking into account laborious sampling and high prices of measurements. Therefore the important problem to guarantee the quality of radioecological monitoring is to define the necessary minimum amount of samples for evaluation of controlled parameters with a specified error.

The planning of amount of samples (amount of sampling) is based on the theory of statistical conclusions [30,31,32,33]. Such planning of amount of samples or sub-samples for tests (observations, measurements) has become widely spread and recognised when quality control of production in different branches of industry: military, aircraft, automobile, chemical, metal-working, radio, petroleum, engineering, instrument-making and other. Some of these methods have become classical and are standardised by the appropriate documents [34,35,36,37,38].

The problems of planning and optimisation of soil sampling for radioecological researches are similar to appropriate problems in geology (mineral deposit sampling). Lithochemical searching of geochemical anomalies is the closest analogy with procedure of detection of radioactive by contaminated spots on the territory and evaluation of average density contamination of certain sites (plots) on the basis of sampling and measurement of soil samples activity [39,40,41,42,43,44,45].

Agrochemical research is certain analogy with radiological research of the fields and agricultural lands (concerning a procedure of work and soil sampling) to evaluate average density contamination with radionuclides (median of density contamination) [46,47,48].

Great attention is paid to these issues in the area of environmental protection where general requirements for the soil sampling are also standardised [49,50,51].

The statistical representativeness of sampling when carrying out radioecological researches of different environmental objects is considered in the given publication. The development of methods of optimisation of representative population of soil, plants and milk samples under actual conditions of radioactive contamination on vast territories (for example, the accident on the Ch NPP) is the purpose of this monograph. The samples must ensure the estimation of median levels of contamination with a specified ratio error.

The sites with non-gradient contamination of territory (plots) have the vital importance when study statistical characteristics of soil and plants contamination with radionuclides, and groups of animals with non-uniformity conditions of stockkeeping have the vital importance when study statistical characteristics of milk contamination with radionuclides. The sites with non-gradient contamination are the sites of territory (fields, lands), within the limits of which any trend of contamination is absent and all local deviations of density contamination have a casual character. The group of animals with non-uniformity conditions of the stockkeeping is the group of animals (herd), which in grazing or stabling period of the stockkeeping has on average the same forage resources. These problems will be discussed in more detail in the appropriate chapters of the monograph.

Next major tasks are considered and solved in the presented monograph:

1. To estimate statistical performances of radioactive contamination of soil, vegetation and transfer factors of radionuclides in a link "soil - plant" at the sites with non-gradient contamination. To determine the kind of probability distribution of soil, vegetation contamination and transfer factors at such sites and to estimate their parameters.
2. To estimate the radius of influence of soil and plants sample and to estimate centre to centre distance of sampling, which ensures statistical independence of radionuclides contents in samples at site with non-gradient contamination;
3. To estimate statistical performances of milk specified radiocontamination in private farms in the settlements and its annual dynamics. To determine a kind of probability distribution of milk contamination in the private farms in the settlements in the specific moment, to estimate its parameters and construction of the stochastic model which describes milk contamination in settlements during the year.
4. To estimate statistical performances of specified activity ^{137}Cs in individual sample, which has volumetric contamination non-uniformity (for example, fuel particles).
5. To determine necessary minimum amount of soil, plants and milk samples when radioecological monitoring, that are necessary for estimation of median of radiocontamination of considered objects (including their derivatives, for example, radionuclides transfer factors from soil to plants) with a specified ratio error.
6. To optimise the milk sampling schedule in the private farms in the settlements for estimating annual individual doze of internal exposure with a specified ratio error.
7. To determine necessary minimum amount of measurements for determining the median specified contents of radionuclides in individual soil sample containing fuel particles for measurement sub-samples of different volume and geometry with specified ratio error.
8. To construct the nomograms providing practical use of the offered procedures for planning soil, plants and milk sampling.

Realised researches based on the systematic and comprehensive approach to a problem of soil, vegetation and milk representative sampling when conducting radioecological monitoring allowed to estimate and systematise statistical performances of radiocontamination objects of monitoring under various conditions and to develop procedures of account of necessary minimum amount of samples. The regularities and nomograms obtained during this project are used now in UIAR to develop the techniques of soil and agricultural products sampling on the radiocontaminated territory, and also to plan the research engineering in the 30-km Zone of Chernobyl NPP. Researches data are the basis of territory mapping of the 30-km Zone of Ch NPP on a density contamination with ^{90}Sr and transuranium elements (maps are on the book cover) [52, 53]. The methodical approaches, used in this work on the development of the optimum schedule of milk sampling have an independent practical importance for estimating annual individual doze of an internal exposure co-ordinating the amount of samples and sampling time.

The present edition is recast with due regard for comments, appeared after publication of the monograph in Ukraine. It contains corrected and supplemental new data obtained by the authors. According to this certain estimations of statistical parameters indicated in the present publication, differ a little from the values obtained before [16]. However this distinction is in error limits of estimation of these parameters.

ACKNOWLEDGEMENTS

Many people have contributed in a variety of ways in the preparation of this book.

At UIAR we would like to express our gratitude to Prof. Y.Ivanov, PhD N.Lazarev, S.Lundin, S.Zvarych, PhD V.Yoshchenko, PhD S. Levchuk, V.Protsak, A.Kadygrob for their constant support and help.

Our sincere thanks go to Prof. B.Prister and Prof. I.Los, who brought their professionalism and expertise to guiding the book in its various stages, especially for comments and suggestions that have resulted in many important improvements.

We could not have produced this book without financial support provided by the Ministry of Emergencies of Ukraine and State Department of the Administration of the Exclusion Zone and the Zone of Absolute Resettlement. Our special thanks are due to V.Kholosha and M.Proskura for their care in organising experiments in the Zone.

Also we would like to thanks to Ferris I.G. (FAO) and Voigt G. (IAEA) for organisational and financial support our book in English; and to N. Zhebrovska, A.Ischenko, for the help when translation this monograph.

1. General information about the errors of measurement

The general information from the theory of probability, mathematical statistics and theory of errors that is necessary for understanding of outcomes and statistical conclusions, presented in the monograph, are indicated briefly in the given section. The particulars of errors can be found in special [54,55,56,57] and in the classical literature on a theory of probability and mathematical statistics [30–32].

1.1 Main concepts of the theory of errors

The quantitative expression of this or that property of investigated object (physical magnitude) is characterised by some numerical value. Up to a measurement it is known, that this value exists.

True value of physical magnitude — this is value, perfectly reflecting properties of the given object, as in quantitative and in a qualitative sense. It is objective and it also depends on neither our consciousness, nor means of a measurement used for experimental determination of it. As a result of experimental determination of value of physical magnitude the findings are different from the true value.

The absolute error of measurement Δ – is a divergence of an outcome of a measurement from the true value of measuring magnitude. It can be determined by the difference between outcome of measurement x and true value of measuring magnitude θ:

$$\Delta = |x - \theta| \qquad (1.1)$$

The ratio error of measurement δ is the magnitude:

$$\delta = \frac{\Delta}{\theta} = \frac{|x - \theta|}{\theta} \;, \qquad (1.2)$$

evaluated in relative units or in percentage.

The real value of physical magnitude – the value that has been found experimentally θ_r, and is so similar to the true one, that can be used for the given purpose instead of it. Then the errors of measurement are determined as the following:

$$\Delta = |x - \theta_r| \;; \qquad \delta = \frac{|x - \theta_r|}{\theta_r} \qquad (1.3)$$

As a rule, the outcomes of observation have different values during the experimental determination of the value of physical magnitude and repetition of the experiment. In any series of measurement the outcomes of single measurement are always different, that is caused by many reasons. The problem is to determine, which evaluation is best for measuring magnitude, and which error is possible in the following measurement under the same conditions of measuring.

Two kinds of errors of measurement components: random error and systematic one can be selected at analysing errors of this or that magnitude concerning the true value.

The appearance of *random errors* has a random character, and methods of mathematical statistics and theory of probability can describe the errors and their distribution. Among the outcomes of measurement there are the ones, which considerably differ from average value of measuring magnitude in the given experiment (distinctly distinguished outcomes). They are caused by either sudden modifications of measurement conditions or by the observer's (operator's) mistakes. The problem is to determine, whether questionable outcomes refer to bad mistakes and have to be excluded from outcomes of observations or they are regular with the definite probability.

The systematic errors are same for all series of observations or they are some functions of time. The systematic error can be eliminated by corrective action. The outcomes of measurement after taking corrective action are called *corrected*. The problem on corrective action is getting complicated when the error correction is determined with some error. In this case corrective action can appear unsuitable, as it does not result in increase of precision measurement. The systematic errors are divided into *constant ones* and *variable ones*. The errors of measurement that don't change through the whole series of measurements are called *constant systematic errors*. For example, error connected with equipment calibration or the ones caused by errors in determinations of characteristics of standards. The errors of measurement that change through the process of measurements are called *variable systematic errors*. For example there is the temperature drift of measurement instrumentation. They are divided into progressive and periodic. If systematic error decreases or increases monotone through the measurement it is called *progressive*; if it is the periodic function of time it is called *periodic*.

The error of measurement method — is a component of error of measurement, which happens because of measurement method imperfection. It could be an error of theoretical approach laid in a method or made by simplification. The errors of method also appear with extrapolation of the property, measured on limited part of some object to the whole object if the one doesn't have any homogeneity of measured property.

While doing indirect measurement the error of the unknown magnitude is calculated on the base of errors of measurement magnitude, connected with the unknown magnitude and determined by direct measurement. In this case the precise knowledge about these magnitudes, their interdependency or independence is necessary.

The researcher can come across the problem of using the outcomes of several investigations received by means of various accuracy with different number of measurement (observation). In this case it is necessary to decide whether it is possible to use the outcomes of all experiments to make general conclusion improving final results.

Let's consider random errors more in details.

Any random variables are described by distribution functions of probabilities: integral and differential.

The probability that the outcome of observation x_i in *i*-test will have less value than specified value x is called *integral distribution function of probabilities* of outcomes of observations:

$$F(x) = P\{x_i \le x\}, \qquad (1.4)$$

where *P* — is a symbol of probability of an occurrence indicated in curly braces.

The integral distribution function of probabilities is no decreasing function of argument. The value of integral function is changed from 0 up to 1.

The differential distribution function of probabilities of a random variable *x* — is the function, which derivatives from integral function on its argument:

$$f(x) = dF(x)/dx \qquad (1.5)$$

The differential distribution function of probabilities *f(x)* is often called distribution density of probability of random variable *x*, and its graphic presentation — a probability curve. Integral distribution function and differential distribution function of probabilities are connected the proportion:

$$F(x) = \int_{-\infty}^{x} f(x)dx \qquad (1.6)$$

The distribution density of probability answers the following conditions:

$$\begin{aligned} &1) \quad f(x) \geq 0 \\ &2) \quad \int_{-\infty}^{+\infty} f(x)dx = 1 \end{aligned} \qquad (1.7)$$

The dimensionality of distribution density of probability of a random variable *x* is magnitude $\mathbf{x}^{-1}$. Probability of hit of an outcome of observation *x* in a specified interval (a; b) is determined as

$$\int_{a}^{b} f(x)dx = F(b) - F(a) \qquad (1.8)$$

1.2 Distribution Laws of random variables

The uniform distribution. If the random variable can take any value on an interval (a; b) with the same probability, then that one is described by the uniform distribution law of probability with distribution density of probability:

$$f(x) = \begin{cases} \dfrac{1}{b-a}, & a \leq x \leq b \\ 0 \quad , & x < a;\ x > b \end{cases} \qquad (1.9)$$

The exponential distribution. The random variable *x* has an exponential probability distribution with parameter λ > 0, if

$$f(x) = \begin{cases} \lambda e^{-\lambda x}, & x \geq 0 \\ 0 \quad , & x < 0. \end{cases} \qquad (1.10)$$

Poisson distribution. A probability distribution of a discrete random variable x which takes value n = 1, 2, 3, …, with probabilities

$$P(x = n) = \frac{a^n e^{-a}}{n!} = f(n,a) \qquad (1.11)$$

is called the law of Poisson distribution with parameter a. It should be noted, that the number of radioactive particles (decay), measured by detector for the certain period in button taken for a measurement, is subject to the law of Poisson distribution.

The normal law of distribution (law of Gauss distribution). It is one among most extended laws of errors distribution. The normal law with parameters (m; σ) has the following expression for a differential distribution function:

$$f(x)=\frac{1}{\sqrt{2\pi\sigma}}e^{-\frac{1}{2}\left(\frac{x-m}{\sigma}\right)^2} \qquad (1.12)$$

It should be noted that despite being widely adopted the normal law of distribution is only a model of objective distributions. In particular it differs from zero on all infinity of axles. Therefore normal distribution supposes, though and with small probabilities, that measured physical magnitude can take negative meaning. This assumption is nonsense for measurements of radionuclides and other substances contents in samples.

Logarithmically normal (lognormally) distribution. The random variable x has logarithmically normal probability distribution, if its the logarithm has a normal probability distribution with parameters (μ; $\sigma_{\ln}$). The expression for its distribution density is:

$$f(x)=\frac{1}{\sqrt{2\pi}\cdot x\cdot\sigma_{ln}}e^{-\frac{1}{2}\left(\frac{ln(x)-\mu}{\sigma_{ln}}\right)^2} \qquad (1.13)$$

In description of outcomes of radionuclides and the other substances activity measurements, the logarithmically normal probability distribution is free from the defect mentioned above, however it supposes the occurrence of samples with unfairly high volume.

1.3 Distribution parameters of random variables and its assessment

The description of random variables with assistance of distribution function is the most universal and correct. However it is sufficient to know one or a few numerical performances of random variable in certain tasks, in particular for normalisation of errors. These performances are: ordinary and central moments.

The ordinary moment of order of r of random variable x is magnitude

$$a_r=\int_{-\infty}^{+\infty}x^r\cdot f(x)dx\ . \qquad (1.14)$$

The ordinary first moment is called *mathematical expectation* of random variable:

$$a_1=\int_{-\infty}^{+\infty}x\cdot f(x)dx=m_x=m \qquad (1.15)$$

The central moment of order r is called magnitude:

$$\alpha_r = \int_{-\infty}^{+\infty} (x-m)^r \cdot f(x)dx \ . \qquad (1.16)$$

The central second moment called variance is the second important parameter of probability distribution, its numerical performance:

$$Dx = \alpha_2 = \int_{-\infty}^{+\infty} (x-m)^2 \cdot f(x)dx \qquad (1.17)$$

The variance is a performance of dispersion of random variable according to mathematical expectation. It is the basis for valuation errors of measurement. The variance has dimensionality of random variable quadrate. The other magnitudes are often used as measure of dispersion of random variable according to mathematical expectation:

- *standard deviation* which is positive magnitude of the square root of variance $\sigma = \sigma_x = \sqrt{Dx}$;
- *coefficient of variation w* equals the relation of standard deviation to mathematical expectation and expressed in relative units or in percentage terms.

The central third moment is used for evaluating asymmetry with the coefficient of skewness $k_a = m_3 / \sigma^3$. For symmetrical distributions the central moment of any odd order equals zero. The distribution has positive asymmetry if $k_a > 0$ and negative one if $k_a < 0$.

The central fourth moment is used for evaluating flat-toppedness and peakedness of curve of probability distribution with help of coefficient of excess. It is true $m_4 = 3\sigma^4$ for the normal law of distribution therefore coefficient of excess is given by $k_\varepsilon = m_4 / \sigma^4 - 3$. For the peaked distribution – $k_\varepsilon > 0$ and for the flat-topped distribution – $k_\varepsilon < 0$.

Shown performances of random variable assume the distribution law and its parameters are known. However the performances are always determined on the basis of experimental data by mathematical statistics methods. In practice the random variable is represented by sample of amount n $(x_1, x_2, ..., x_n)$, on the basis of which it is necessary to evaluate a kind of distribution, as well as its parameters. First of all mathematical expectation and variance (standard deviation) are estimated parameters. Estimation of parameter is called point estimate, if one number expresses it. Quality of point estimates is characterised by the following concepts:

- The point estimate is called unbiased if its mathematical expectation coincides with the true value of estimated parameter;
- The point estimate is called consistent, if it difference from estimated parameter can be made indefinitely small when the amount of observations (sampling amount) increases;
- The point estimate is called effective, if its variance is less than the variance of any other evaluation of the given parameter.

Unbiased, consistent and effective estimation of mathematical expectation is sample average of observations outcomes $\overline{\mathrm{x}}$:

$$\bar{x}=\frac{1}{n}\sum_{i=1}^{n}x_i \tag{1.18}$$

Consistent and effective valuation of variance is

$$\bar{s}^2=\frac{1}{n}\sum_{i=1}^{n}(x_i-\bar{x})^2 \tag{1.19}$$

Unbiased and consistent estimate of variance is sample variance

$$s^2=\frac{1}{n-1}\sum_{i=1}^{n}(x_i-\bar{x})^2 \tag{1.20}$$

Point estimate of standard deviation is the sample deviation

$$s=\sqrt{\frac{1}{n-1}\sum_{i=1}^{n}(x_i-\bar{x})^2}\ . \tag{1.21}$$

If the investigated random variable *z* is differentiable function of several other random variables with known performances $z=f(y_1,y_2,\dots,y_n)$, then the point estimate of it variance can be calculated on the basis of expansion of this function in Taylor series in the midpoint of the function to the second-order term. In case of statistical independence of magnitudes $y_1,y_2,\dots,y_n$ the expression for point estimate of variance is given by

$$s_z^2=\sum_{i=1}^{n}\left(\frac{\partial f}{\partial y_i}\right)^2_{y_i=\bar{y}_i}\cdot\sigma_{y_i}^2 \tag{1.22}$$

Thus, the error of sought quantity can be evaluated if indirect measurement on the basis of errors of magnitudes measurement is connected with sought quantity and determined by direct measurement.

1.4 Determination of confidence intervals for true values of mathematical expectation and for variance of the normally distributed random variable

Any point estimate determined on the basis of experimental data is their function and therefore it is random variable itself with distribution dependent on distribution of basis random variable and on number of tests. Let outcomes of measurements (observations) x_i have normal probability distribution. Let's consider two cases.

The variance of distribution σ^2 is known. In this case sample average $\bar{x}$ also has normal distribution with mathematical expectation $\bar{x}$ and variance σ^2/n. Probability of position *p* of unknown mathematical expectation (which is necessary to be evaluated) could be determined in specified interval:

$$P\left\{\bar{x}-\frac{u_p\sigma}{\sqrt{n}}\le m\le\bar{x}+\frac{u_p\sigma}{\sqrt{n}}\right\}=p, \tag{1.23}$$

where u_p is appropriate fractile of normal distribution, and defined from the tables. Thus obtained interval for unknown value *m* is called confidential one and appropriate value *p* is confidence probability. The error value $\Delta_p = \frac{u_p \sigma}{\sqrt{n}}$ is called the confidential boundary of error of measurement outcome and the estimation of mathematical expectation (outcome of measurement) is noted down as $m = \bar{x} \pm \Delta_p$.

The variance of distribution σ^2 is unknown. In this case random variable $t = \frac{m - \bar{x}}{s\sqrt{n}}$ is followed to Student distribution with n-1 degree of freedom and the random variable $\chi^2 = \frac{s^2}{\sigma^2}(n-1)$ has χ^2-probability distribution with n-1 degree of freedom. Hence confidential intervals for mathematical expectation and variance are created by analogy:

$$\begin{aligned} &\bar{x} - \frac{t_{p,n-1} \cdot s}{\sqrt{n}} \le m \le \bar{x} + \frac{t_{p,n-1} \cdot s}{\sqrt{n}} \\ &\frac{s^2 \cdot (n-1)}{\xi_{n-1,(1+p)/2}} \le \sigma^2 \le \frac{s^2 \cdot (n-1)}{\xi_{n-1,(1-p)/2}} \end{aligned}, \qquad (1.24)$$

where $t_{p,n-1}$ and $\xi_{n-1,(1+p)/2}$ ($\xi_{n-1,(1-p)/2}$)are appropriate fractile s of Student distribution and χ^2-distribution with n-1 degree of freedom determined from the tables.

2. Statistical performances of soil, vegetation and agricultural products contamination with radionuclides

Some territory (field, site) contamination with any radionuclide can be mathematically described by some continuous function of the locality co-ordinates $f(x,y)$. Generally this function has three components:

- Trend of contamination - monotonic component of density radioactive fallout conditioned by global (with respect to controlled territory) gradient of fallout;
- Spot of contamination - localities with increased or reduced density contamination against a background the trend;
- Random component, describing microheterogeneity of radioactive fallout in a point conditioned by technique and process of soil sampling, preparation for measurement, technique and process of measurement.

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

$$f(x,y) = f_{tr}(x,y) \cdot f_{st}(x,y) \cdot f_{ac} \, , \qquad (2.1)$$

where $f_{tr}(x,y)$ – function describing monotonic trend of density contamination;

$f_{st}(x,y)$ – function describing spots of density contamination against trend;

f_{ac} – random component independent on co-ordinates of point on investigated territory.

Such presentation is convenient when the multiplicity model for $f(x, y)$ is substituted by an additive model for $z(x, y)$

$$z(x,y) = z_{tr}(x,y) + z_{st}(x,y) + z_{ac} \, . \qquad (2.2)$$

taking the logarithm $z(x,y) = ln\big(f(x,y)\big)$. Later on if it is not agreed specially, all researches and statistical analysis will be made for logs of density contamination $z(x,y) = ln\big(f(x,y)\big)$.

The representation of density contamination as (2.1) and (2.2) is sufficiently general. The density contamination practically of any complexity can be described with such model. This idea has given good account when mapping geological indications [58] and was used in UIAR for constructing a map of contamination with ^{90}Sr on the 30-km zone of the Ch NPP [52].

When we normalise density contamination of arbitrary plot (site) on the trend of general form $z'(x,y) = z(x,y) - z_{tr}(x,y) + z_{st}(x,y)$, we receive uniformly contaminated (non-gradient) site ($f'(x,y)$=1) with respect to the normalised density within the limits of which all divergences of density contamination have a random character.

Thus, researching statistical performances of density contamination, for example experimental one, on uniformly contaminated sites (*f(x,y)* = const), (speaking differently - non-gradient), gets fundamental importance. The statistical conclusions, obtained for these sites, are the basis of similar conclusions for sites that have trend of radioactive contamination of arbitrary form $f_{tr}(x,y)\cdot f_{st}(x,y)$. That fact, that any plot (site) of non-uniformly contaminated territories by any way can be separated in practice on quasi-non-gradient sites in contamination, indicates the importance of statistical performances research of density contamination on non-gradient sites. Therefore uniformly contaminated (non-gradient) sites of territory are the subjects of further consideration in the present work.

The systematic researches for study the features of statistical performances of soil, plants, and agricultural products contamination with radionuclides and for determining regularities and their statistical connections with the features of landscape and kinds of radioactive fallout have been conducted in UIAR for 10 years.

2.1 Survey of observations, sampling and measurement

The experimental sites on fuel or condensation traces were selected according to two types of Chernobyl radioactive fallout (**Fig. 2.2**, Fig. 2.3). Two control sites were also selected on the territory, where the soil radioactive contamination was generated mainly by global fallout (the radioactive Chernobyl fallout is practically absent). Soil radioactive contamination on these sites is background in comparison with the Chernobyl fallout (further - background sites). The soil density contamination with ^{137}Cs on these sites is 2-3 kBq/m^2 (Fig. 2.3, sites **S-P 18** and **S 19**). The sites contaminated mainly with a fuel component of radioactive fallout were selected in the 30-km zone of the Ch NPP, and with condensation component both in the 30-km zone, and outside it - on adjoining territories. The experimental sites cover a broad spectrum of levels of soil contamination with ^{137}Cs - from 50 kBq/m^2 up to 30 MBq/m^2. The sites were selected for both types of fallout on the virgin grounds (or fallow) and on the arable grounds, which have been used in agriculture till now. The experimental sites include three groups of landscapes: flood plains, flats and watersheds, and also sites located in the woods.

An experimental site is usually in the shape of a square, rarely – a rectangle. The method of the embedded sites was used for sampling on experimental site. Inside the large (main) site (with a large pitch of sampling, 10m) the middle site (with a pitch of sampling 2 m), was selected inside the middle the small site (with a pitch of sampling 0,1-0,05 m) was selected, as it is shown in the diagram (Fig. 2.1). The internal sites were selected in localities with the most developed vegetative cover.

The general diagram of location and numeration of experimental sites is indicated below in the figures (**Fig. 2.2**, Fig. 2.3). The marking of sites, where only soil samples were taken, starts with the letter "S" in the diagrams. The plant samples were taken along with soil samples under the same diagram on some sites.

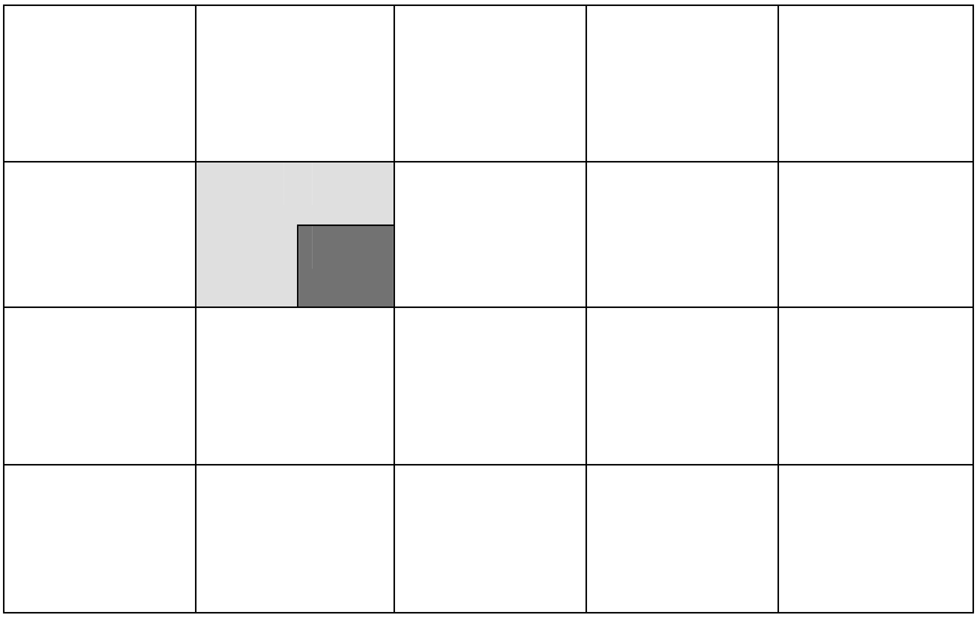


Fig. 2.1. The scheme of soil and plants sampling

The statistical performances of different (cultivated and growing wild) plants contamination with radionuclides, which are typical for the radioactive contaminated territory of Ukrainian Polissia, were evaluated and analysed. The species diversity of plants includes the main agricultural plants triticum (clurum vulgare), secale (cereale), avena (sativa); the typical representatives of meadows and pastures: elytrigia repens (L) Nevski, calamagrostis epigeios (L) Roth and oenothera biennis (L) - wild animals forage. The marking of sites, where the soil - plant conjugate samples were taken starts with letters combination "S-P" in the indicated diagrams.

The settlements, where there the basic radiological posts of UIAR located, were selected to research the statistical performances of the specific contents ^{137}Cs in milk (Fig. 2.3). The settlements, where the milk was sampling, are marked as the sites starting with letter "M" on the indicated map.

The Kiev area: Ragivka (**M1**), Lugovyky (**M2**) Polis'ke region (≈ 50 km from the Ch NPP), Kovshovate (**M3**) Tarascha region (≈ 220km from the Ch NPP).

The Zhytomyr area: Gladkovychi (**M4**) Ovruch region (≈ 90km from the Ch NPP), Lypnyky (**M5**) Lugyn region (≈ 120km from the Ch NPP), Selets' (**M6**) Narodychi region (≈ 80km from the Ch NPP).

The Chernigiv area: Pakul' (**M7**) Chernigiv region (≈ 50km from the Ch NPP).

The Rivne area - Myliachi (**M8**) Dubrovytsia region (≈ 230km from the Ch NPP).

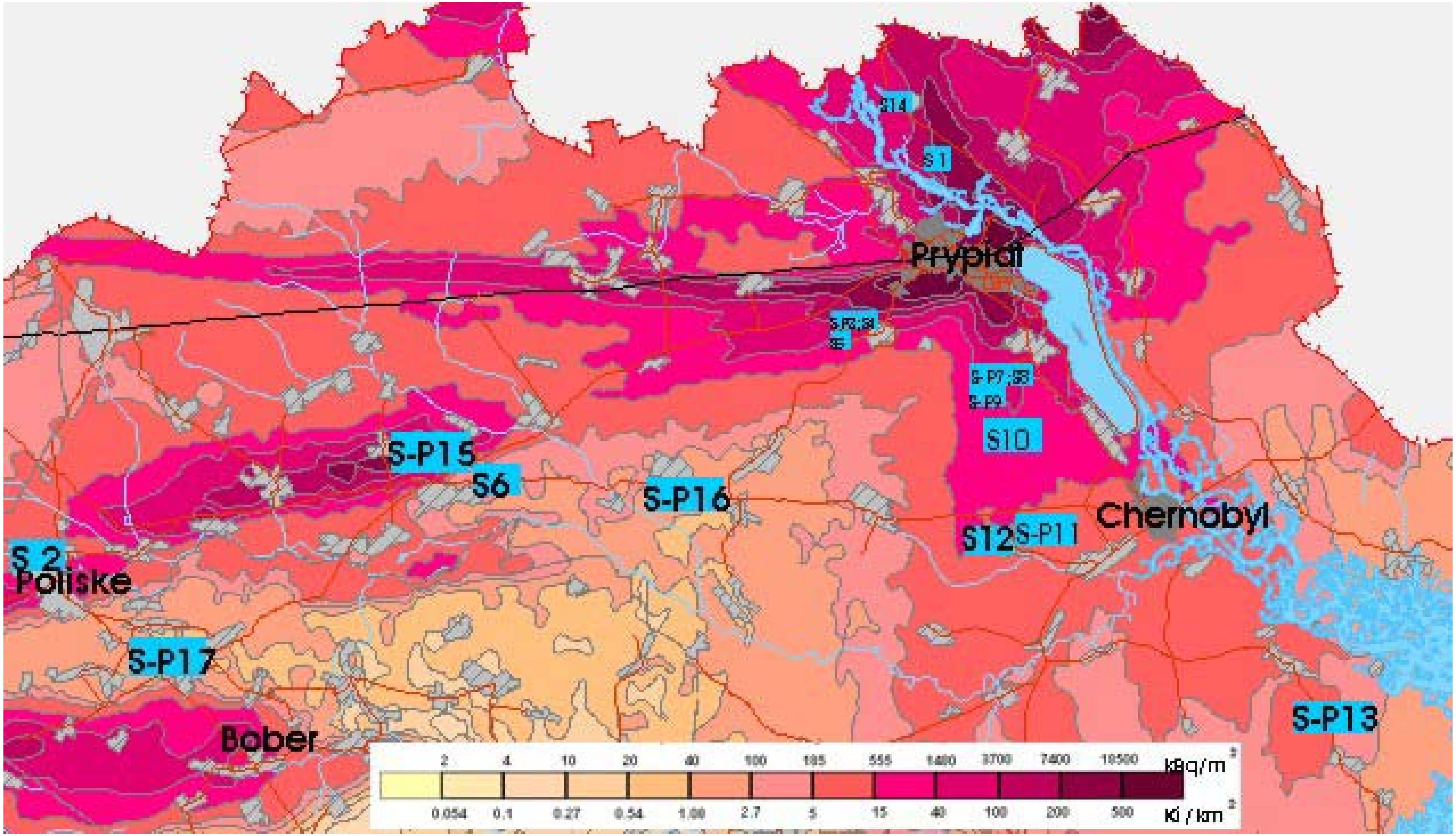


Fig. 2.2. The scheme of experimental sample sites position in the 30-km zone and adjacent areas (^{137}Cs density of contamination [59]).

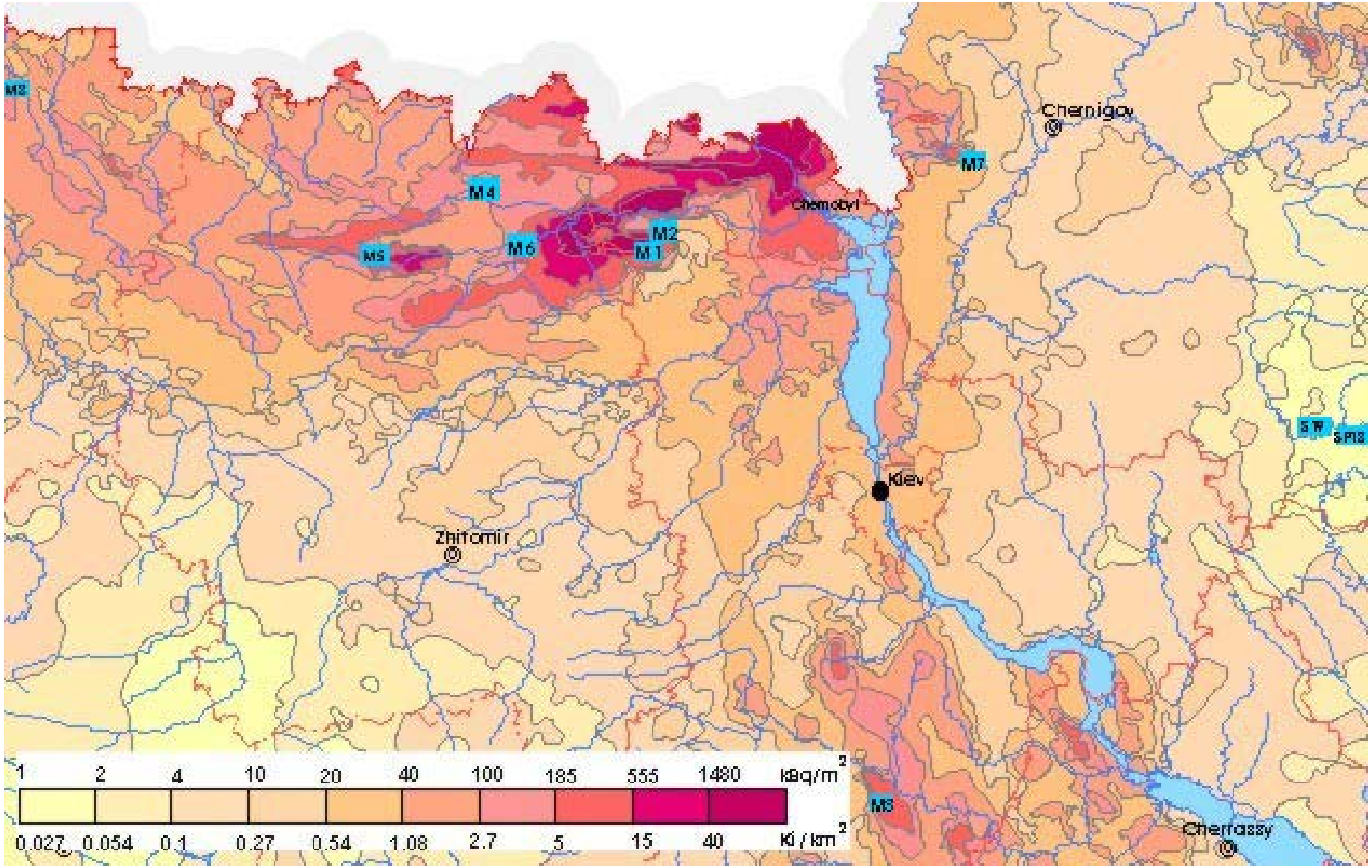


Fig. 2.3. The scheme of experimental sample sites position on the areas with global radioactive fallouts and base radiological station of UIAR (^{137}Cs density of contamination).

The following had to be taken into account when evaluate density soil radioactive contamination as the analysis of research outcomes conducted in UIAR, and literary data [52] evidences:

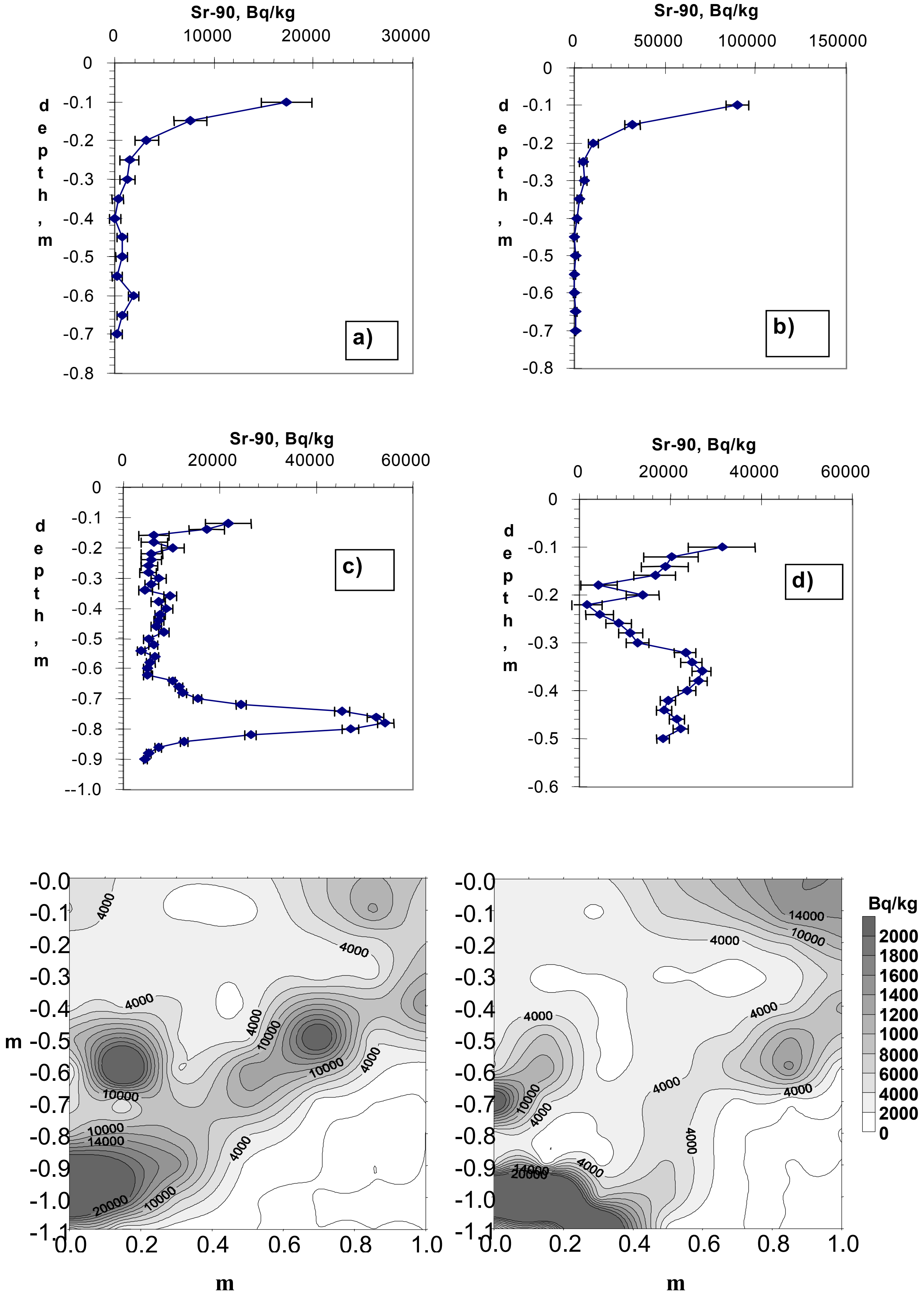
- particularities of landscape, where the density contamination is evaluated;
- statistical performances of contamination;
- intensity of the radionuclide vertical and horizontal migration;
- value of sampling pitch;
- sampling square.

The long-term observations of the vertical migration of ^{137}Cs and transuranium elements show, that their main mass on the territory radioactively contaminated in consequence of accident on the Ch NPP is in the upper 5-10 cm soil layer now. Thus, it is necessary to sample into 10-cm depth on the virgin sites and it is enough to evaluate the soil density contamination with these radionuclides. However, during the time the situation will vary. The consequences of accident at the enterprise "Maiak" in 1957 evidence it. The main reserve of ^{137}Cs in flood-plain soils of the river Tech' has already located in the layer 0-10, 0-20 cm depending on the landscape [60,61].

The detailed measurements of profiles of vertical distribution ^{90}Sr into the depth to 110 cm was conducted in the 30-km zone of the Ch NPP on the right bank of the river Prypiat', on sandy plateau (the soils with small humus contents) (Fig. 2.4).

The obtained outcomes show, that the main reserve of ^{90}Sr in some sampling points can be concentrated in a 30-cm layer (Fig. 2.4 a, b), or is much deeper than 30 cm (Fig. 2.4 c, d). The pit measuring 1m x 1m x 1.1m for detailed soil sampling was dug to study in detail vertical distribution of ^{90}Sr on one of those sites with characteristic sandy soil. The characteristic soil contamination with ^{90}Sr for two vertical cuts is shown in figure (Fig. 2.4). The distance between cuts is 70 cm. The obtained outcomes are the evidence of large volumetric non-uniformity of distribution ^{90}Sr on sandy soils with small humus content, realised in consequence of its vertical migration [21].

The higher velocity of ^{90}Sr migration can be observed not only in sandy soils, consequences of the accident at the enterprise «Maiak» are the evidence about it [60,61]. Both ^{90}Sr and ^{137}Cs migrate on larger depth in remoisten flood-plain soils than on non-flooded sites of terraces. If in there are up to 90 % ^{90}Sr and ^{137}Cs in upper 0-10 cm layer, sod-meadow soil of non-flooded terrace then in alluvial-stratified soil near a channel, the maximal contents ^{137}Cs is displaced on 10-20 cm depth, and ^{90}Sr - 20-40 cm.



cut 1 cut 2

Fig. 2.4. – Example of ^{90}Sr distribution in a structure of sandy soils

On the arable lands radionuclides are mixed even more of less uniformly in arable layer (depth 15-20 cm) and migrate into subsoil horizon. No more than 10-20 % of the total content of radionuclides percolated in subsoil layer before 1999 on data obtained on experimental allotments [62]. The similar measurements were conducted in the 30-km zone of the Ch NPP on site **S-P 15**. The points, where the sampling was conducted level-by-level with pitch into depth 5 cm, were selected in random manner on the field ploughed every year till 1991. The outcomes of this research are shown in Figure (Fig. 2.5).

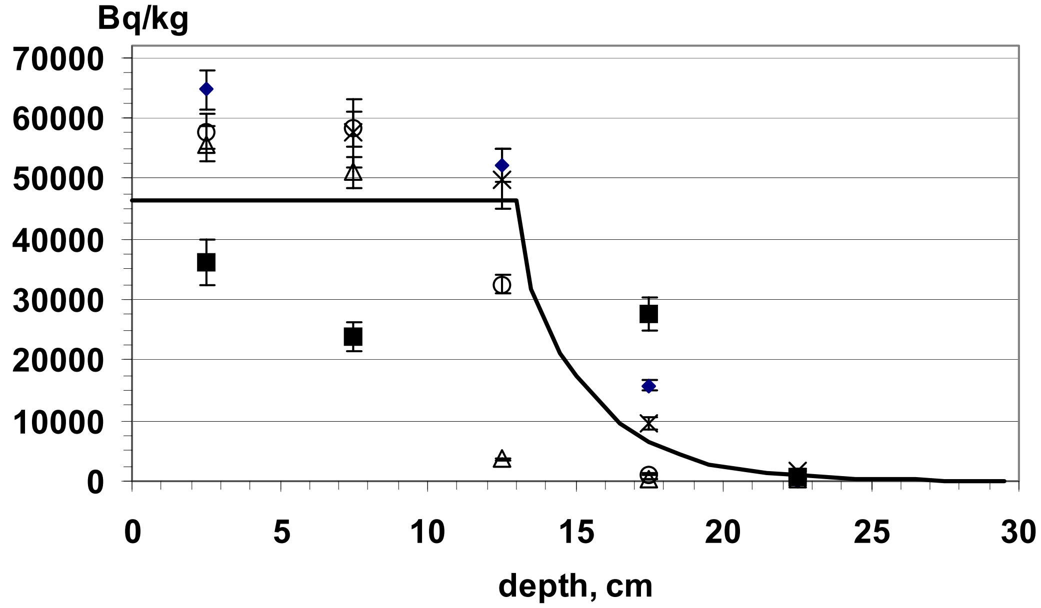


Fig. 2.5. – ^{137}Cs migration in subsoil on the sample station **S-P 15**.

As it is obvious in figure the depth of arable layer is 13-15cm under production conditions. 14 % of activity ^{137}Cs is below 13 cm, and more than 6 % is lower than 15 cm.

Thus, it was enough to sample into 30 cm depth on the whole contaminated with ^{137}Cs territory to define soil density contamination with ^{137}Cs till 2000. The depth of sampling with 30 cm was also sufficient to determine the soil density contamination with ^{90}Sr, except sand poor in humus. There the sampling depth has to be determined in every specific case after appropriate monitoring of ^{90}Sr migration depth.

Two types of sampler were used:

- ring with diameter 13.2 cm, height 5 cm;
- cylindrical sampler with diameter 3.7 cm, height 30 cm.

The ring was used on those sites, where the main part of radionuclides is into 5-cm soil layer. The cylindrical sampler was used, as a rule, on all sites and first of all on arable and fallow lands and also in the woods.

The soil sampling was conducted in the every network node (Fig. 2.1). The sampling with ring into depth 5 cm was conducted on the standard technique [26]. A ring drives into soil and then accurately dug out. The soil sampling with sampler was conducted into depth 30 cm with pitch 10 m and 2 m, as a rule, this way: in every point (network node) 5 samples (injections) were taken by the envelope with the side 5-10 cm. Central sample was packed into separate polyethylene bag; the other four were united and packed into the other package. It allowed (node) having three sampling squares (0.001 m^2; 0.004 m^2 and 0.005 m^2) in every point. Sometimes only one sample (one injection) has taken in network node. On sites **S-P 18** and **S 19** sampling square was 0.005 m^2 (five injections).

The samples of plants were selected in the same nodes of regular network. The pitches of regular network for agricultural cultures (arable fields) were 10 and 2 meters and for growing wild plants - 10, 2 and 1 meter depending on uniformity of vegetative cover. The plant sample square was is shape of circle with diameter 0.2 - 1 m depending on density of plant growth.

The milk samples were taken from each cow monthly in the private farms. The milk samples were taken separately from each herd if the animals of the settlement were pastured on different pastures (tracts), in summer period.

The radionuclide contents in soil, plants and milk samples were measured with γ-spectrometer ADCAM-300 with the semiconductor detector GEM-30185. As a rule, only one sub-sample with volume 100 cm^3 (cylinder with diameter 6.8 cm, height 3 cm) was measured from the sample. The sub-samples taken on the sites **S-P 18** and **S 19** were measured in two geometric shapes: the cylinder 100 cm^3 (diameter 6.8 cm, height 3 cm) and the Marinelle flask 1000 cm^3. The measuring average error of ^{137}Cs activity in soil samples was ≤10% (±2σ), and on global fallout - ≤20% (±2σ) in the Marinelle flask and ≤25% (±2σ) in cylindrical vessel 100 cm^3 for sites located on Chernobyl traces. The measuring average error of ^{137}Cs activity in plant samples was from ≤10% to 30 % depending on average sample activity on site at level ±2σ. The measuring error of ^{137}Cs activity in milk did not exceed 15% (±2σ).

2.2 Statistical performances of soil contamination

The research of statistical performances of soil radioactive contamination, describing random component of contamination variability of the territory, was conducted on basis of analysis ^{137}Cs and ^{90}Sr contents in soil samples taken on the experimental sites. The geostatistical image of the density contamination of two typical sites located on different fallout traces is shown in figures (Fig. 2.6–Fig. 2.7).

The local non-uniformity (randomness) of soil contamination within the limits of site is corollary of micro-non-uniformity of radionuclide fallout on site caused by the influence on their local sedimentation of atmosphere fluctuations, microcontour and vegetation features, and also consequent radionuclide redistribution. The fuel particles presence in original radioactive fallout makes soil contamination more heterogeneous even on small sites. Besides the significant influence on contamination heterogeneous of soil high layer introduces consequent anthropogenous effect of soil organisms, wild animals and people. Inevitable errors of determining of the radionuclide contents in the soil sample are also added to it.

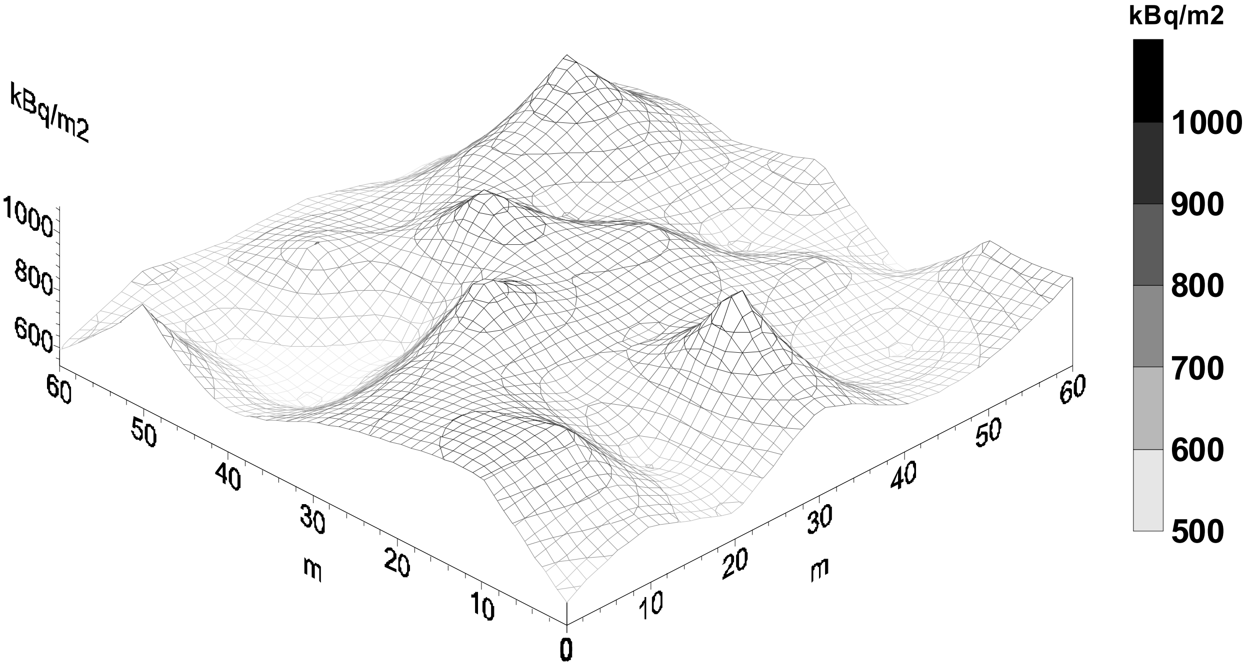


Fig. 2.6– Geostatistical image of the experimental site **S-P 17** ^{137}Cs contamination.

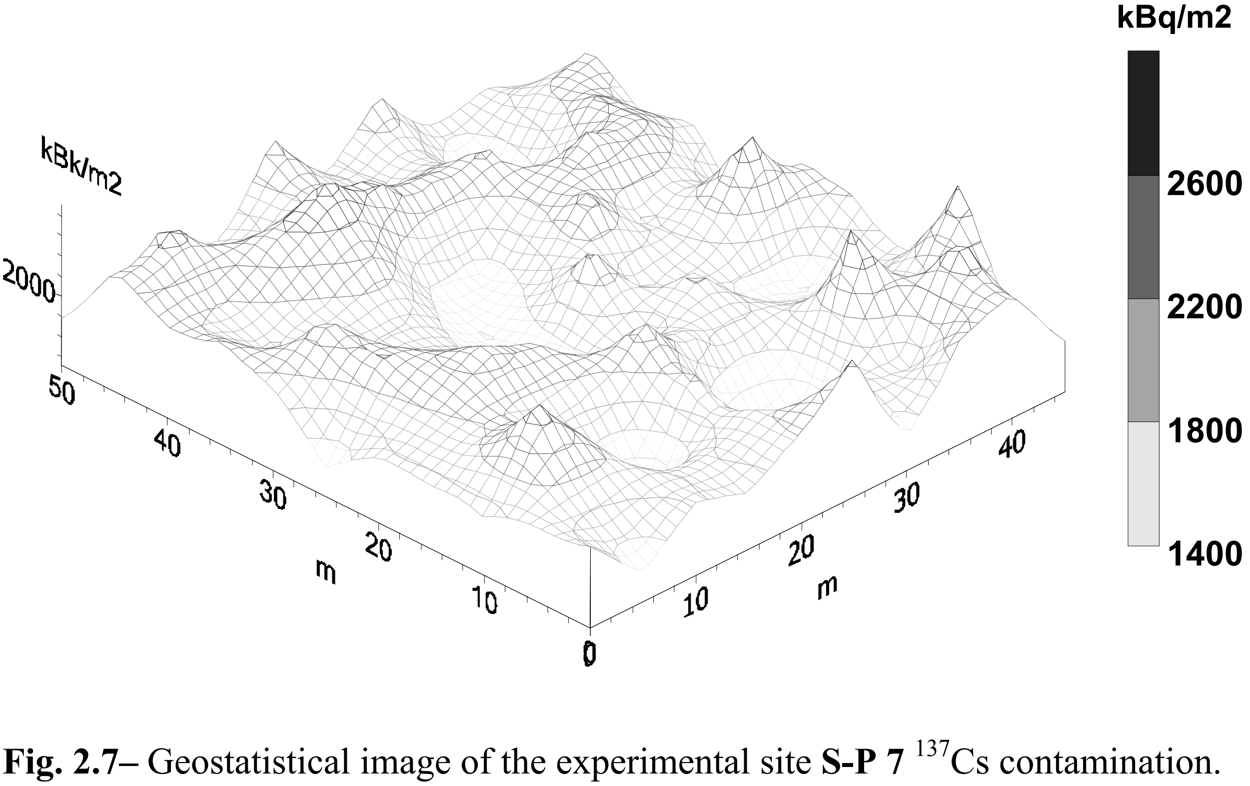


Fig. 2.7– Geostatistical image of the experimental site **S-P 7** ^{137}Cs contamination.

The theory of statistical conclusions [30–33] allows us to consider as some random variable the values of soil density contamination within the limits of some site when global gradient of radioactive fallout obtained of the basis of soil sampling and measuring, is absent. Any random variable is completely described by suitable to its probability distribution law, which is approximated in practice by predicted probability distribution. The choice of this or that predicted probability distribution is an important stage of statistical analysis to describe investigated random variable. When choosing the predicted probability distribution of random variable it is necessary to take into account its physical essence and available a priori information. Such information exists in the considered case. Firstly, density radioactive soil contamination is especially positive magnitude. Secondly, the sampling for evaluating density soil contamination is analogue, in its essence, with geochemical sampling of barren geochemical anomaly or territories polluted with chemical compounds and heavy metals. The content of different elements in minerals and content of minerals in different rocks is described with the logarithmically normal law of probability distribution in most cases very well, which is shown in publications devoted to sampling of mineral deposits and geochemical anomalies [39–45]. Thirdly, this magnitude has a significant spread in values and right-handed asymmetry (Fig. 2.8) as it is shown in the publications [63, 64, 65], where the probability performances of soil density contamination are analysed. The statistical treatment and analysis of obtained measurement outcomes of soil density contamination with ^{137}Cs and ^{90}Sr for selected experimental sites also vindicate this data. In this case the logarithmically normal law of probability distribution is the simplest predicted probability distribution to describe non-gradient sites [30–32]. This probability distribution becomes:

$$f(C_{so}) = \frac{1}{\sqrt{2\pi} \cdot C_{so} \cdot s_{so}} e^{-\frac{1}{2}\left(\frac{ln(C_{so}) - \mu_{so})}{s_{so}}\right)^2} \qquad (2.3)$$

where:

C_{so} – soil density contamination with the radionuclides;

μ_{so} – average log of radionuclides density contamination of the site;

s_{so} – standard deviation of log of radionuclides density contamination of the site.

The parameters of this probability distribution law have quite defined physical sense /63,64/. Density contamination median equals:

$$Me_{C_{so}} = exp(\mu_{so}), \qquad (2.4)$$

which is also called geometric average. The parameter $\boldsymbol{s_{so}}$ is the approximate evaluation for coefficient of variation $W_{C_{so}}$ of density contamination of the site:

$$s_{so} \cong \frac{S_{C_{so}}}{\overline{C}_{so}} = W_{C_{so}} \qquad (2.5)$$

it does not depend on half-life period values of radionuclides.

Dispersion of log of the density soil contamination in samples $s^2_п$ is conditioned by micro-non-uniformity of the site contamination (including fuel particles), by process of sample preparation for measurement and by the choice of measured sub-sampling, error of standard pattern and error of measurement.

The typical histograms of ^{137}Cs and ^{90}Sr density soil contamination in kBq/m^2 and their approximation by the logarithmically normal law of probability distribution on experimental sites when sampling with ring (diameter 13.2 cm, height 5 cm) and with sampler (diameter 3.7 cm, height 30 cm) for different landscapes and radioactive fallout traces are indicated in figure (Fig. 2.8).

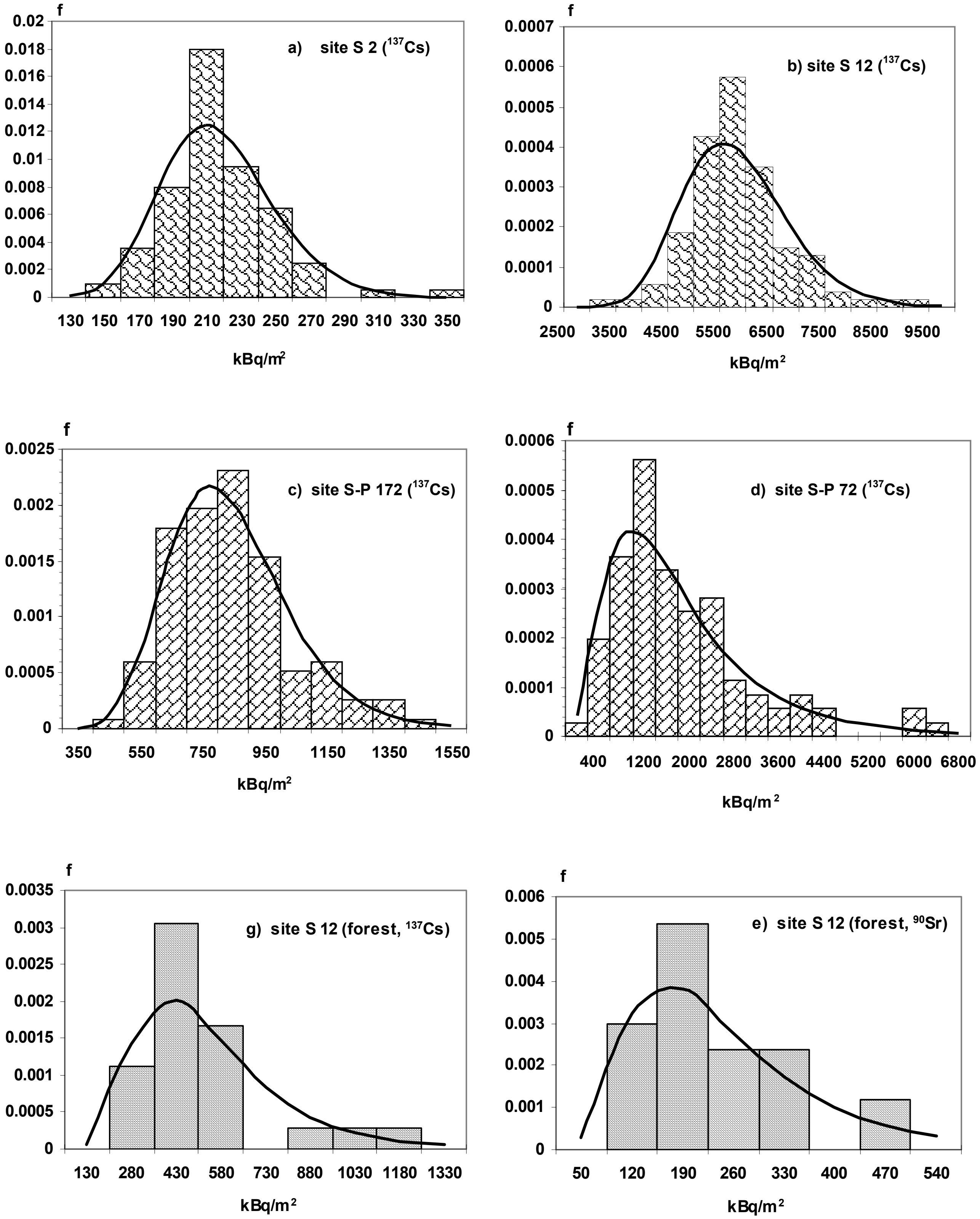


Fig. 2.8.- Distributions of probability of soil density contamination with radionuclides on experimental sample stations when sampling by a ring: diameter 13.2cm, height-5cm (a, b); by a sampler: diameter 3.7cm, height -30cm (c, d, g, e).

Available outcomes of determination of soil density contamination with ^{137}Cs and ^{90}Sr for non-gradient experimental sites according to Kolmogorov's criterion of consent (goodness-of-fit test) (significance level 0.05) [63,64] do not contradict the logarithmically normal law of probability distribution, as examination of the outcomes has shown.

Thus, the outcomes of advanced statistical analysis indicate satisfactory approximation of ^{137}Cs and ^{90}Sr density contamination on non-gradient sites with the logarithmically normal law of probability distribution.

2.2.1 Evaluation of variability of soil density contamination on non-gradient sites

Various performances of variability of random variable are used for random variables analysis and in statistical conclusions. The list of used performances of the spread can be found in the literature on mathematical statistics, for example [30–34,66,67,68]. In practice such performances as spread and dispersion, and also performance calculated on the basis of the last one the standard deviation and coefficient of variation, are used most frequently. In our work a considered random variable (the density soil contamination) has logarithmically normal probability distribution. As mentioned above in this case dispersion of distribution (standard deviation) characterises relative spread of values of the density soil contamination (coefficient of variation). The important distinction of variance of log of density soil contamination is that it does not depend on time (radionuclide decay) and in this sense it is a certain invariant (in contrast, for example, to coefficient of variation of log of density soil contamination). Therefore we have selected parameter s_{so} - standard deviation of log of density soil contamination to study variability of the density soil contamination and its description on non-gradient contamination sites. To find the estimations of this parameter for non-gradient contamination sites located on different landscapes, traces of Chernobyl radioactive fallouts and statistical analysis to reveal the most important factors having influence on their significance is the purpose of the present chapter. The measurement outcomes obtained in the cylindrical flask with volume 100 cm^3 will be mainly used for indicated estimations. The values of estimations obtained when the sub-samples were measured in Marinelle flask will be noted separately.

2.2.1.1 The influence of radionuclide fallout composition

The ^{137}Cs and ^{90}Sr contents were determined in soil samples for some experimental sites located on fuel traces of fallout. The samples preparation for measuring was conducted in the same technique and the ratio error of measurements of radionuclides activity in soil samples was maintained at the same level (≤10 %).

The ratio between the density soil contamination with ^{137}Cs and ^{90}Sr in 30-km zone of the Chernobyl NPP was explicitly studied when constructing the map of 30-km zone of the Ch NPP contamination with ^{90}Sr [52]. The relation ^{137}Cs/^{90}Sr in the 30-km zone has a complicated structure and is determined by ratio of fuel and condensation components of radioactive fallout at each point. However, it is possible to consider this relation as a constant one within the limits of small non-gradient sites. The observable ratio between these radionuclides obtained by the results of sampling in 1999, has turned out at first approximation identical for investigated sites and it is shown below in Figure (Fig. 2.9).

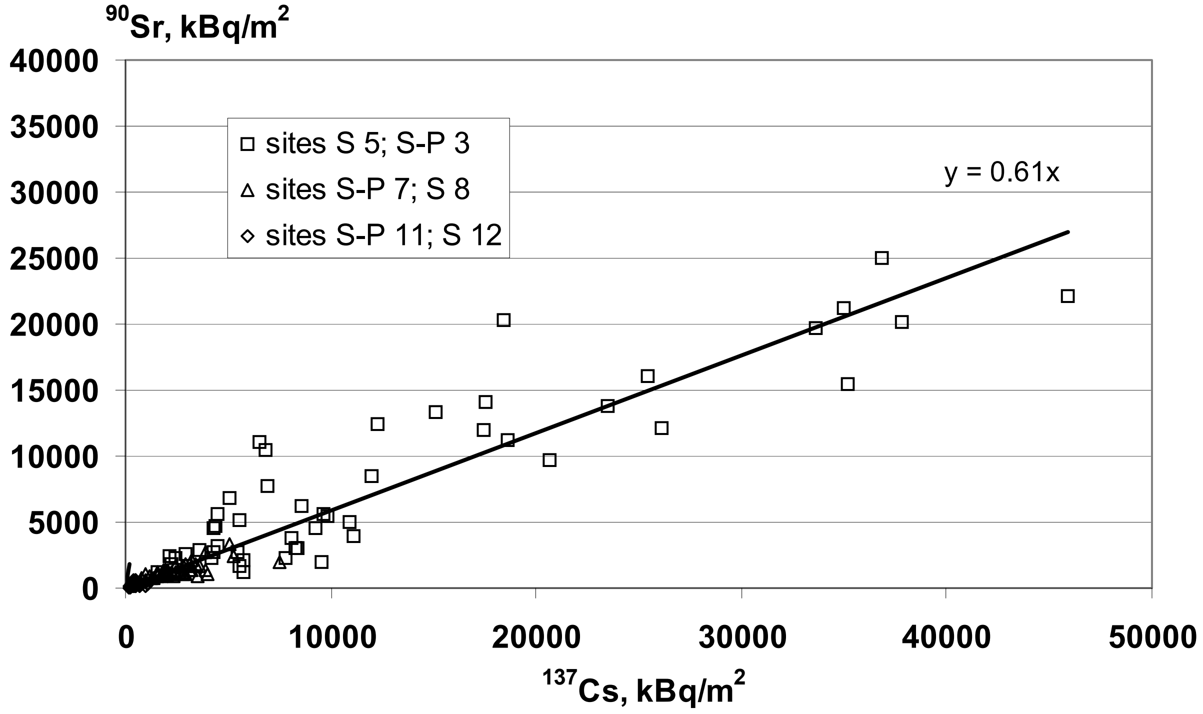


Fig. 2.9.- Ratio of ^{137}Cs to ^{90}Sr content in soil on different fuel traces in 1999.

Therefore, the relative spread in values of density soil contamination with ^{137}Cs and ^{90}Sr (coefficients of variation) will be equal on the same site with non-gradient contamination and for at first approximation identical relative error of measurement. This fact assumes equality of variances of log of ^{137}Cs and ^{90}Sr density contamination of non-gradient sites depending on ratio (2.5).

The equality of variances of log of ^{137}Cs and ^{90}Sr density contamination on non-gradient sites is confirmed by the outcomes of statistical analysis of direct measurements conducted by our researchers (Fig. 2.10).

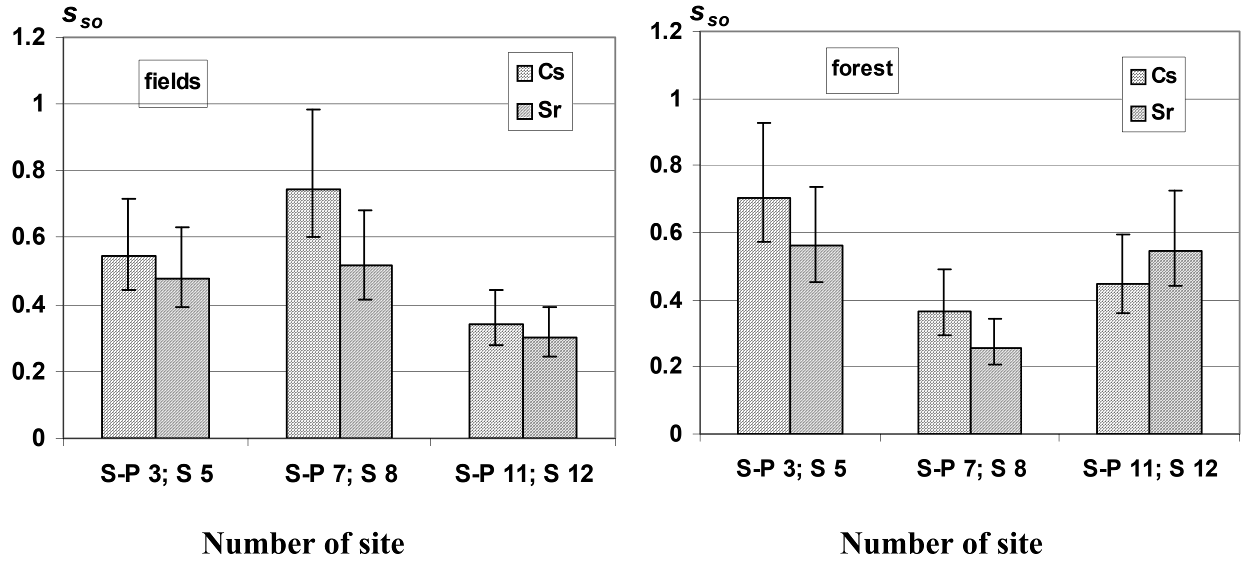


Fig. 2.10.- Magnitudes of standard deviation of log of soil contamination density with ^{137}Cs and ^{90}Sr values on different landscapes and fuel traces.

It is visible in the given Figure that values of standard deviation of log of density contamination with these radionuclides practically do not differ under the condition of close errors of determination of the ^{137}Cs and ^{90}Sr content in soil on different fuel traces and landscape sites.

As on the fuel traces of fallout the ^{90}Sr content in soil are mainly caused by its presence in fuel component, then this statement is correct for any radionuclide of fuel component. It means that with the first approximation we can get the evaluation of standard deviation of log of soil density contamination with any radionuclide of fuel component s_{so}^{Rn}, on fuel traces of fallout if we use obtained evaluations for standard deviation of log of soil density contamination with ^{137}Cs:

$$s_{so}^{Rn} \cong \sqrt{(s_{so}^{Cs})^2 + 0.01 \cdot (b^2 - 1)} \qquad (2.6)$$

where b=1, if the error of measurement of radionuclide activity in soil sub-sample does not exceed 10 %;

$b=10\times\delta$, if the error of measurement of radionuclide activity in soil sub-sample exceeds 10 %;

δ is relative error of measurement of radionuclide activity in a soil sub-sample.

Thus, later on the evaluation of influence of the other factors on spread in values of density contamination on non-gradient sites will be conducted only in relation to variance (standard deviation) of log of soil density contamination with ^{137}Cs.

2.2.1.2 Dependence of variance value on Sampler Square

As it is mentioned above, two types of samplers: ring (diameter 13.2 cm, height 5 cm) and cylindrical sampler (diameter 3.7 cm, height 30 cm) were used for soil sampling. While sampling, the sampling square had four values: 0.00107m^2, 0.0043 m^2, 0.0054m^2 (cylindrical sampler 1, 4 and 5 injections) and 0.014m^2 (ring). The values of variance of log of density contamination correspond to these squares on each experimental site. These differences are quite explained and have statistical nature.

It is possible to consider a soil sample selected with sampler of large square as some number of sample (***n***) selected with sampler of smaller square, next to each other. The values of radionuclide content in these samples will be statistically dependent ones. It is possible to consider the radionuclide content in soil sample selected with sampler of large square at first approximation as average value of the radionuclide content in statistically dependent samples selected by a sampler with a smaller square. As it shown above, the random variable representing log of soil density contamination with ^{137}Cs, has normal distribution probability. The variance of average value of these ***n*** of statistically dependent random variables is [30,32]:

$$\sigma_{av}^2 = \frac{\sigma^2}{n^2} \cdot (n + 2 \cdot \sum_{i=1}^{n} \sum_{j=1}^{i-1} r_{i,j}) , \qquad (2.7)$$

where $r_{i,j}$ – appropriate correlation coefficients.

Since for discussed situation all the samples are located compactly, next to each other, the values $r_{i,j}$ are close and at first approximation consider them equal r - some effective correlation coefficient. Then expression (2.7) is simpler:

$$\sigma_{av}^2 = \frac{\sigma^2 \cdot (1 + r \cdot (n-1))}{n} , \tag{2.8}$$

For $r = 0$ (statistically independent random variables) we have expression well known in statistics [30,32]. In this case $n = \frac{S}{S_1}$, where S – sampling square for any sampler; $S_1 = 0.00107\ m^2$ – sampling square of cylindrical sampler 3.7 cm in diameter (one injections).

Then the ratio of variances of log of soil density contamination on experimental site gets the dependence on working sampler square

$$\frac{s_{so}^2}{s_{1,so}^2} = \frac{0.00107}{S}\left[1 + r \cdot (\frac{S}{0.00107} - 1)\right] , \tag{2.9}$$

where $s_{1,so}^2$ - variance of log of soil density contamination for sampling with cylindrical sampler (diameter 3.7 cm).

The effective correlation coefficient for experimental sites, where the sampling square had various values, had been evaluated on the basis of expression **(2.9**) (Table 2.1).

Table 2.1 Значение эффективного коэффициента корреляции для различных площадок.

Conventional sign of a site	Type of fallout	Land use	Effective correlation coefficient, r
S-P 3	fuel	fallow	**0.06**
S-P 7	fuel	fallow	**0.17**
S-P 9(large)	fuel	arable	**0.23**
S-P 9(small)	fuel	arable	**0.23**
S-P 11	fuel	fallow	**0.38**
Average value			**0.21±0.11**
S-P 13(large)	condensate	arable	**0.85**
S-P 13(small)	condensate	arable	**0.74**
S-P 15(large)	condensate	arable	**0.60**
S-P 15(small)	condensate	arable	**0.56**
S-P 16(large)	condensate	fallow	**0.71**
S-P 16(small)	condensate	fallow	**0.36**
S-P 17(large)	condensate	arable	**0.78**
S-P 17(small)	condensate	arable	**0.57**
Average value			**0.65±0.11**

Conducted variance analysis of obtained effective correlation coefficient has shown, that statistically significant factor is the *Type of fallout*. The value of average effective correlation coefficient on fuel traces of radioactive fallout equals 0.21 and it proves actual absence of dependence for the ^{137}Cs content in samples selected next to each other.

The values of standard deviation of log of soil density contamination with ^{137}Cs s_{so} obtained with each sampling square were normalised on values $s_{1,so}$ to exclude the influence of individual features of the site. The outcome of conducted normalisation for different experimental sites is shown in figure (Fig. 2.11). There are theoretically (approximated) generalised dependencies **(2.9)** of this ratio on working sampler square for fuel and condensation traces of fallout.

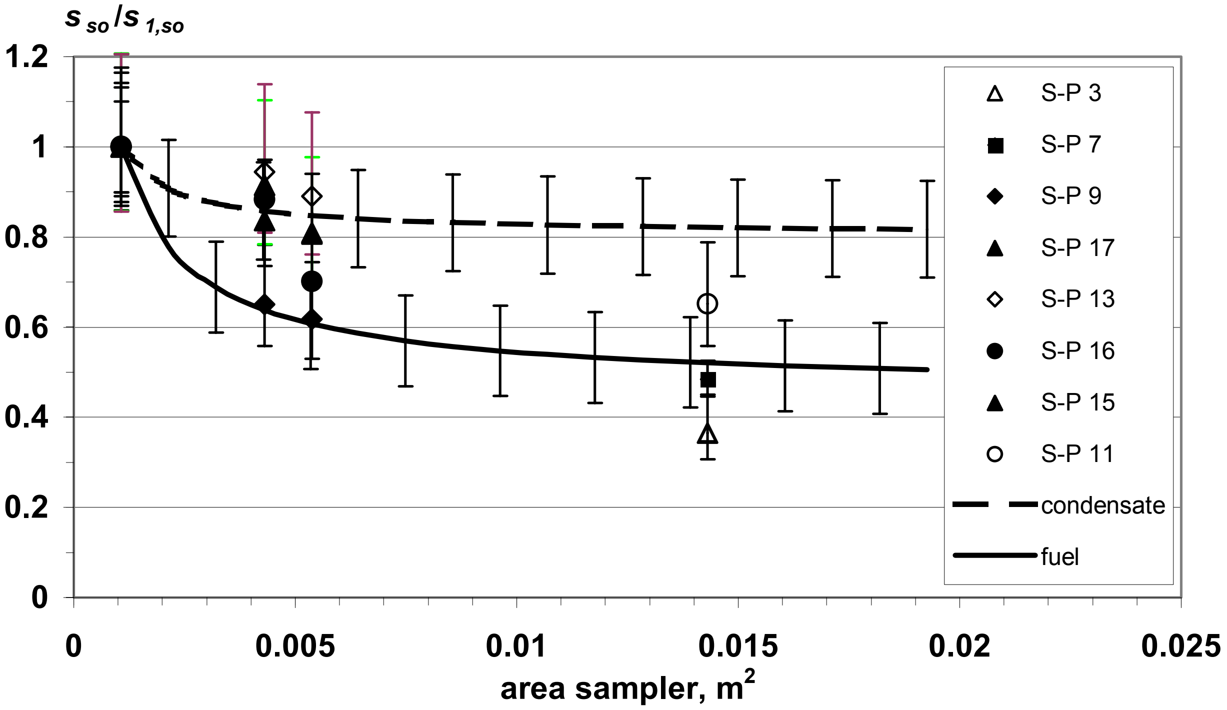


Fig. 2.11.– The dependence of the normalised standard deviation of log of soil density contamination on the sampler square.

These dependencies are very important to analyse the observable values of variance (standard deviation of log) of soil density contamination that are obtained for different experimental sites and for sampling with samplers of different working square. They allow recalculating obtained values on the same sampler square. The fact, that these dependencies differ on fuel and condensation traces of fallout is the evidence of dependence of variance of log of soil density contamination on the type of radioactive fallout. This problem will be considered in more detail.

2.2.1.3 Influence of the other factors on variance of log of soil density contamination with ^{137}Cs

In correspondence with expression **(2.9)** all values of standard deviation of log of soil density contamination with ^{137}Cs, obtained on different sites (s_{so}), were recalculated (normalised) for common sampler square (in to magnitude $s_{1,so}$). The cylindrical sampler with 3.7cm in diameter was taken as standard sampler.

The conducted statistical analysis for brought to common sampling square of values of standard deviation $s_{1,so}$ has shown, that average fallout density with ^{137}Cs (on the site), sampling pitch (in limits of 0.2-10 m) and square of the experimental site for level of value α=0.05 are not statistically significant factors (Fig. 2.12).

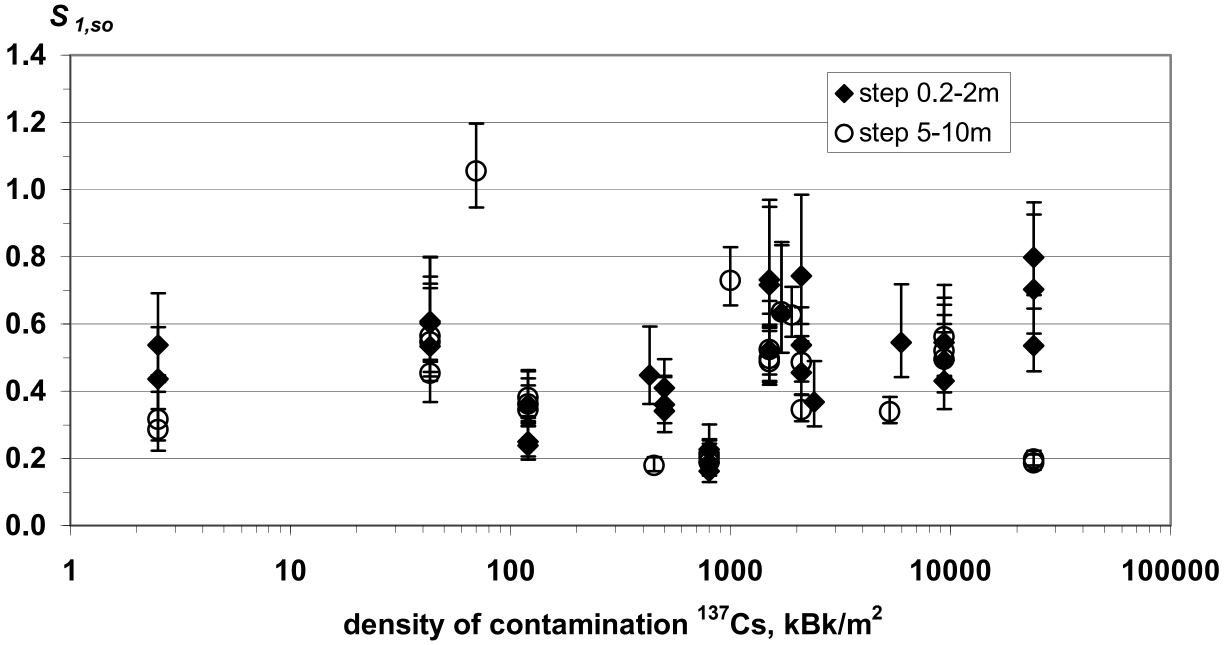


Fig. 2.12 – The influence of density fallouts and sampling pitch on standard deviation of log of soil density contamination with ^{137}Cs.

Thereby values $s_{1,n}$ with ^{137}Cs were averaged due regard for their weighting coefficients (errors of determination) for each experimental site brought to common sampling square and are shown in one table (Table 2.2).

Table 2.2 - Generalised values of standard deviation of log of soil density contamination with ^{137}Cs for different sites

Conventional sign of site	Type of fall-out	Land use	$s_{1,so}$	Error $s_{1,so}$	
				Δ^-	Δ^+
S 1	fuel	grassland	0.339	0.034	0.043
S 2	condensate	grassland	0.180	0.019	0.024
S-P3; S 4	fuel	fallow	0.484	0.072	0.109
S 5	fuel	woodland	0.545	0.103	0.173
S 6	fuel	fallow	0.627	0.065	0.084
S-P 7	fuel	fallow	0.513	0.075	0.113
S 8	fuel	woodland	0.367	0.071	0.123
S-P 9	fuel	arable	0.580	0.097	0.154
S 10	fuel	fallow	0.730	0.076	0.098
S-P 11	fuel	fallow	0.370	0.059	0.091
S 12	fuel	woodland	0.447	0.086	0.146
S-P 13	condensate	arable	0.323	0.051	0.078
S 14	fuel	grassland	0.635	0.120	0.204
S-P 15	condensate	arable	0.507	0.085	0.135
S-P 16	condensate	суходол	0.551	0.104	0.176
S-P 17	condensate	arable	0.197	0.033	0.053
S-P 18	condensate	fallow	0.277	0.043	0.0612
S 19	condensate	arable	0.249	0.038	0.0522

Statistical analysis of generalised values of standard deviation of log of soil density contamination with ^{137}Cs (Fig. 2.13) has confirmed the difference between the average values $s_{1,so}$ on fuel and condensation traces of fallout for sampling with sampler (diameter 3.7 cm, sampling square - 0.00107 m^2) at the moment of sampling (1999).

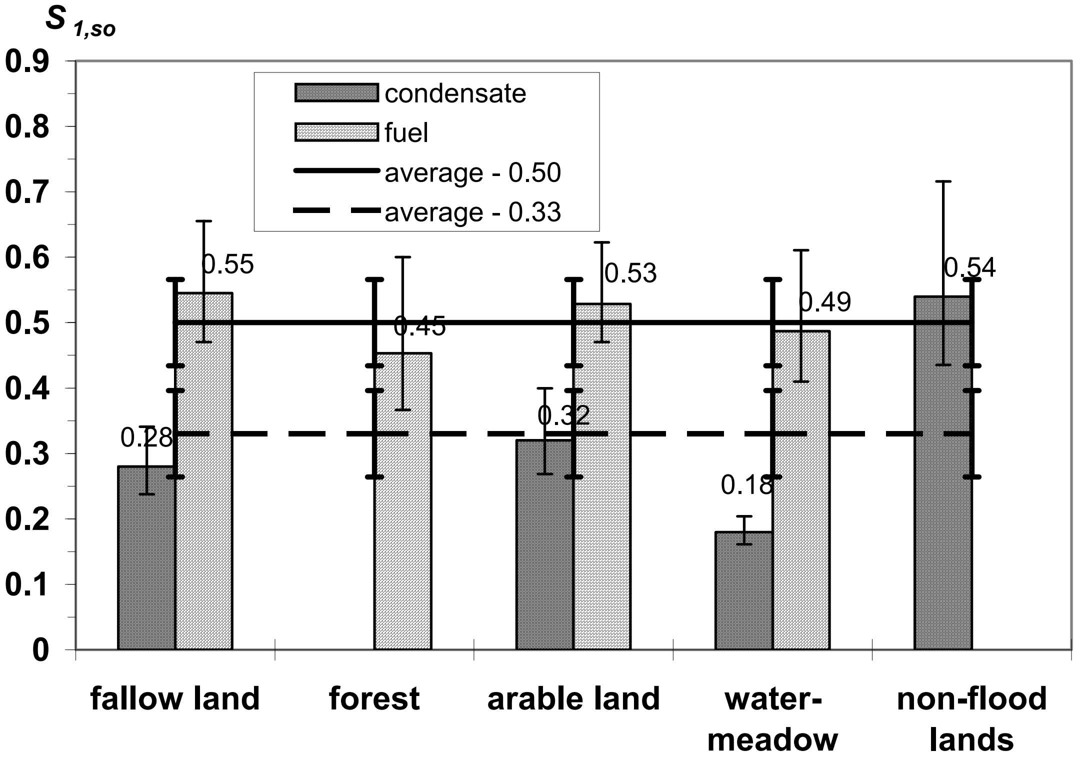


Fig. 2.13– The influence of the kind of fallouts and some landscape particularities of on standard deviation of log of soil density contamination with ^{137}Cs when sampling square is of $S_1 = 0.00107 m^2$.

The presence of different dependencies for standard deviation of log of soil density contamination with ^{137}Cs on fuel and condensation traces of fallout on sampling square allows to analyse the ratio $q = \frac{s_{so}^{fuel}}{s_{so}^{cond}}$. It is seen in the given figure (Fig. 2.14) the difference between standard deviations of log of soil density contamination on considered traces of fallout is practically absent for sampling squares ≥ 0.005 m^2. The fact that the considered ratio decreases to less than 1 while sampling square increases, is the evidence of approximation of the obtained dependence (2.9).

Thus, it is convenient to accept the value 0.0054 m^2 as base sampling square, and the integrated sample received in the result of 5 injections with sampler (diameter: 3.7 cm) or with the other sampler (sampling square is 0.0054 m^2) as a base soil sample. Since values of magnitudes s_{so}^{fuel} and s_{so}^{cond} can only approach during the time in accordance to dissolution of fuel particles, the selected sampling square guarantees the equality of variance of soil density contamination with ^{137}Cs on non-gradient sites for all traces of radioactive fallout even after 1999.

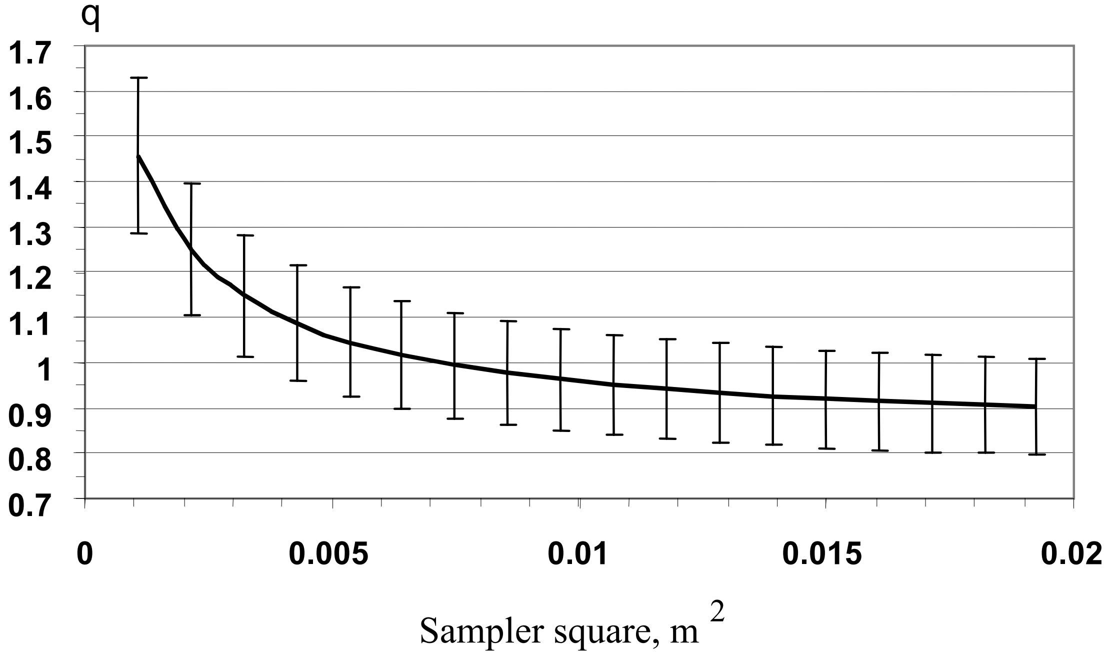


Fig. 2.14– The relation between standard deviations of log of soil density contamination with ^{137}Cs depending on sampling square on fuel and condensation traces of fallouts.

Depending on stated above, outcomes, generalised for the different sites, ((Table 2.2), were recalculated on sampling square 0.0054 m^2 (in magnitude $s_{5,so}$ – Table 2.3).

Table 2.3 - Generalised values of standard deviation of log of soil density contamination with ^{137}Cs for different experimental sites brought to sampling square 0.0054 m^2

Conventional sign of site	Type of fallout	Land use	$s_{5,so}$	Error $s_{5,so}$	
				Δ^-	Δ^+
S 1	fuel	grassland	0.206	0.021	0.026
S 2	condensate	grassland	0.153	0.016	0.020
S-P3; S 4	fuel	fallow	0.294	0.044	0.066
S 5	fuel	woodland	0.331	0.062	0.105
S 6	fuel	fallow	0.380	0.039	0.051
S-P 7	fuel	fallow	0.311	0.046	0.069
S 8	fuel	arable	0.352	0.059	0.094
S-P 9	fuel	woodland	0.222	0.043	0.075
S 10	fuel	fallow	0.443	0.046	0.060
S-P 11	fuel	fallow	0.225	0.052	0.088
S 12	fuel	woodland	0.271	0.052	0.088
S-P 13	condensate	arable	0.274	0.043	0.066
S 14	fuel	grassland	0.385	0.073	0.124
S-P 15	condensate	arable	0.430	0.072	0.115
S-P 16	condensate	waterless	0.468	0.089	0.149

		valley			
S-P 17	condensate	arable	0.167	0.028	0.045
S-P 18	condensate	fallow	0.235 0.204*)	0.036 0.029	0.052 0.041
S 19	condensate	arable	0.211 0.20*)	0.031 0.030	0.044 0.043
Average value $s_{5,so}$			**0.30**		
Standard deviation $s_{5,so}$			**0.09**		

*) the Marinelle flask 1000 cm^3.

The outcomes of conducted variance analysis of values of standard deviation of log of soil density contamination with ^{137}Cs for different sites brought to sampling square 0.0054 m^2 are shown in figure (Fig. 2.15).

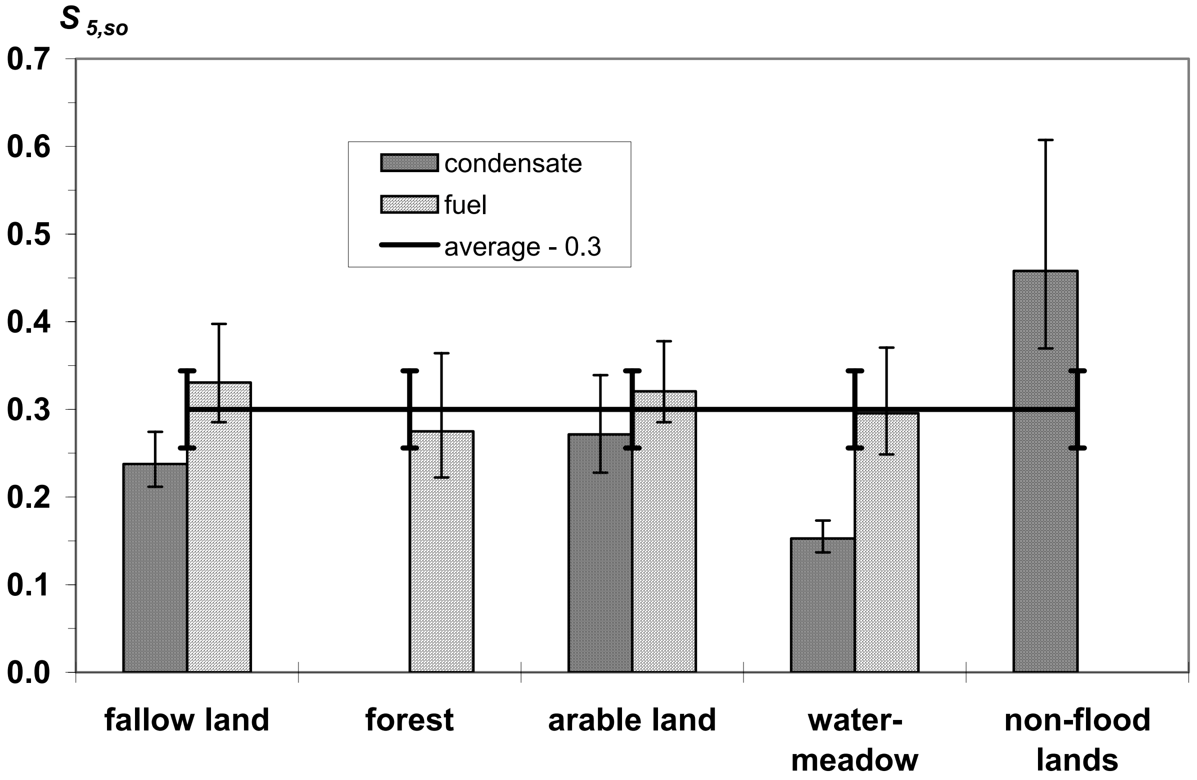


Fig. 2.15– The influence of the kind of fallouts and some landscape particularities on standard deviation of log of soil density contamination with ^{137}Cs when sampling square is of S_1 = 0.0054 m^2.

For magnitude $s_{5,n}$, the conducted analysis has not shown statistically significant differences on fuel and condense traces of radioactive fallout (as it was expected), and also the influence of landscape performances of sites (as well as for magnitude $s_{1,п}$). The observable differences have a casual character. Obtained evaluation ($s_{5,so}$=0.30±0.09) is the average one on different traces of radioactive fallout. This evaluation can only decrease during the time after dissolution of fuel particles and decrease of volumetric heterogeneity of the sample. Therefore it is possible to use this evaluation at the first approximation, as conservative evaluation, in any next year after the accident on the Ch NPP.

2.3 Statistical performances of plant contamination

The statistical aspects of plant contamination with radionuclides, on non-gradient with contamination sites, fields, meadows, fields and other grounds (hereinafter they are sites) are being considered in the present section. As it was mentioned before the plants had been sampled on the same experimental sites as the soil samples. The typical space structure of vegetation contamination on experimental sites is shown below in Figures (Fig. 2.16- Fig. 2.20).

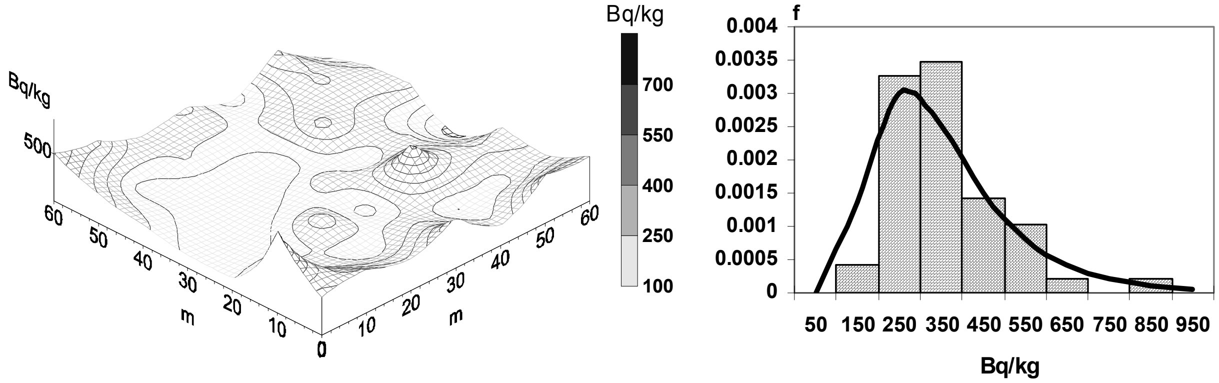


Fig. 2.16.- The spatially-statistical structure of secale (cereale) contamination with ^{137}Cs on the experimental site **S-P 9**.

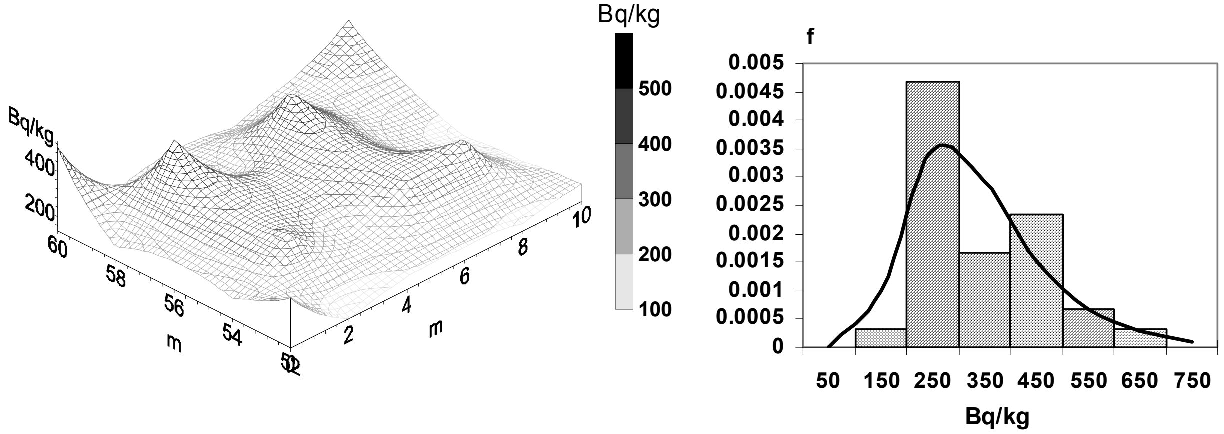


Fig. 2.17.- The spatially-statistical structure of secale (cereale) contamination with ^{137}Cs on a small experimental site **S-P 9**.

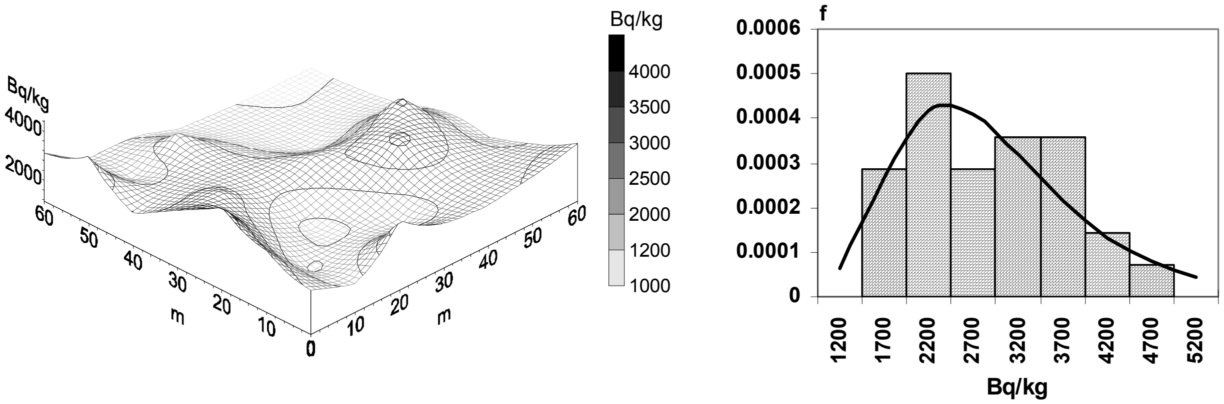


Fig. 2.18.- The spatially-statistical structure of oenothera biennis (L) contamination with ^{137}Cs on the experimental site **S-P 15**. Donkey grass did not grow in the upper corner of the site

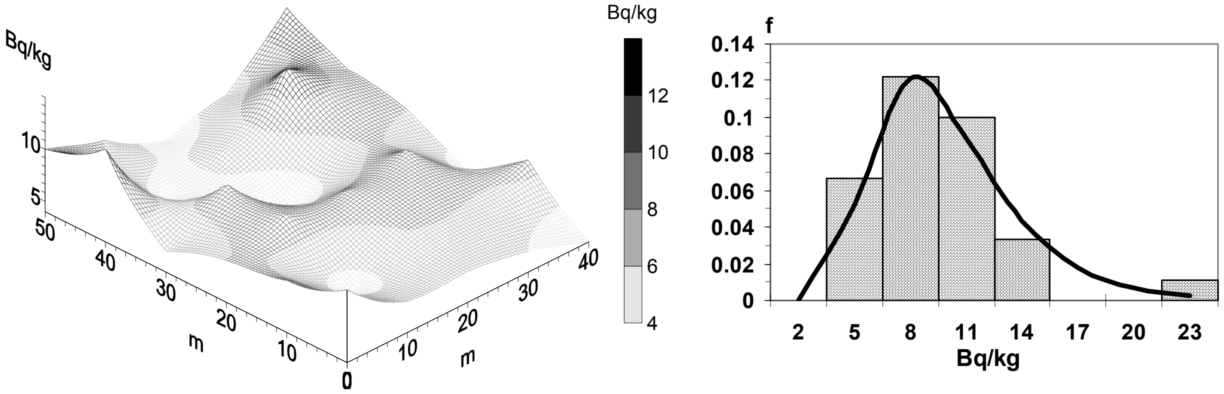


Fig. 2.19.- The spatially-statistical structure of calamagrostis epigeios (L) Roth contamination with ^{137}Cs on a small experimental site **S-P 18**.

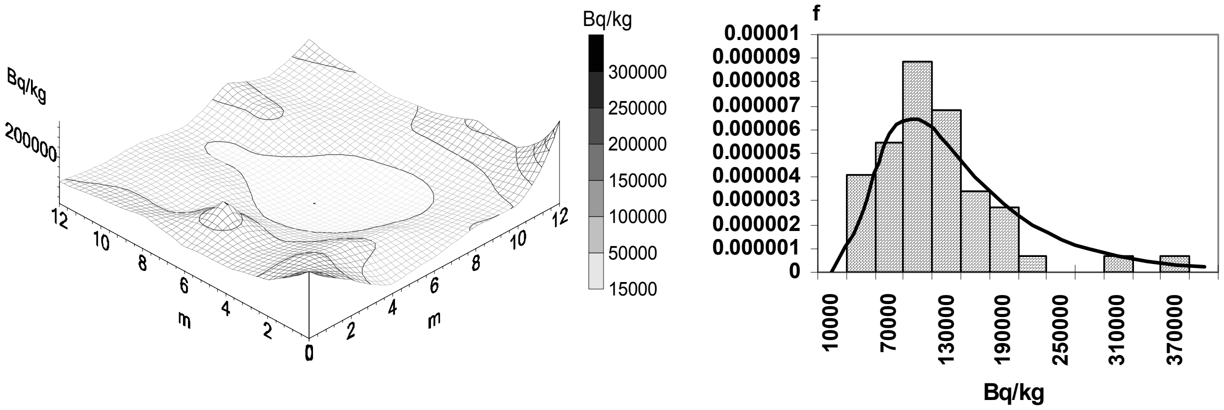


Fig. 2.20.- The spatially-statistical structure of elytrigia repens (L) Nevski contamination with ^{137}Cs on the experimental site ^{137}Cs **S-P 3**.

The non-uniformity of vegetation contamination is the consequence of the site's microheterogeneity contamination, natural soil microheterogeneity, and its fertility and physical-chemical properties, i.e. function of uncontrolled random variables. The inevitable errors of determination of the ^{137}Cs content in plant samples are also imposed on it. It allows to consider plant contamination with ^{137}Cs on non-gradient sites as a random variable [63,64], which is described by its own law of probability distribution.

The ^{137}Cs specific content in plant samples C_p is especially positive magnitude. Literary data [63], and also the statistical processing and analysis of outcomes of ^{137}Cs concentration measurement in the selected samples of various plants prove, that this magnitude has significant spread in values and right-hand asymmetry (Fig. 2.16-Fig. 2.20). As well as for the soil density contamination, the simplest theoretical probability distribution to describe the ^{137}Cs specific contents in plant samples selected on non-gradient sites, is the logarithmically normal law of probability distribution:

$$f(C_p) = \frac{1}{\sqrt{2\pi} \cdot C_p \cdot s_p} e^{-\frac{1}{2}\left(\frac{ln(C_p)-\mu_p)}{s_p}\right)^2} \qquad (\,2.10)$$

where μ_p - average log of the ^{137}Cs specific contents in plants selected on non-gradient site;

s_p - standard deviation of log of the ^{137}Cs specific contents in plants selected on non-gradient site.

The approximation of vegetation contamination on non-gradient sites gives quite satisfactory outcomes by this law of distribution probability (Fig. 2.16 - Fig. 2.20).

2.3.1 Variability of the ^{137}Cs contents in plant samples and the various factors influence on it

As well as in soil samples the variance of log of the ^{137}Cs specific content of plants s_p^2 on non-gradient sites is stipulated by microheterogeneity contamination of the site, natural soil microheterogeneity, process of sampling preparation and measuring error. Only measuring error is controlled magnitude among the listed factors. For example it is mainly determined by the time for sub-sample measuring when using γ-spectrometric method. The measuring of the ^{137}Cs contents in vegetative samples selected on different sites (with different average activity) was conducted with different error because of different methodical and organisational reasons (objective and subjective). Average value of a ratio error of ^{137}Cs measurement in plant samples selected on the the same site equals from ≤ 10% up to 30 % at the level $\pm 2\sigma$ depending on the density contamination of the site. Average value of a ratio error of ^{137}Cs measurement for all sites equals 8.8 % at the level $\pm\sigma$ (Table 2.4). To make it possible under these conditions to compare correct spread indexes of the specific ^{137}Cs content in samples for different sites we have accepted: the base ratio error of measurement should not exceed 10 % at the level $\pm\sigma$. Taking into account this assumption and ratio (2.5) оценки для $\boldsymbol{s}_p$ the evaluations for $\boldsymbol{s}_p$ on experimental sites S-P 13 and S-P 18 were corrected according to the expression:

$$s_p = \sqrt{s_{0,p}^2 - \left(\frac{\delta_{inst}}{100}\right)^2 + 0.01} \qquad (2.11)$$

where $s_{0,p}$ - initial standard deviation of log of the specific ^{137}Cs content in plants;

δ_{inst} - ratio error of measurement of the specific ^{137}Cs content in vegetative samples at the level ±σ.

The evaluations of parameters of the logarithmically normal law of probability distribution obtained according to described above position are also listed in table (Table 2.4). These evaluations and conditions will be considered as basic ones.

Table 2.4.- The evaluations of parameters of the logarithmically normal law of probability distribution of vegetation contamination with ^{137}Cs on different sites.

Conventional sign of site	**Type of vegetation**	**Median value ^{137}Cs contents in plants, Bq/kg**	δ_{inst}**, %**	**Value s_p**
S-P 3	**elytrigia repens (L) Nevski**	87060	2.3	0.62
S-P 7	**calamagrostis epigeios (L) Roth**	520	5.7	0.22
S-P 9 (big)	**secale (cereale)**	280	8.6	0.45
S-P 9 (small)	**secale (cereale)**	270	9.2	0.36
S-P 11	**calamagrostis epigeios (L) Roth**	250	5.6	0.38
S-P 13 (big)	**avena (sativa)**	90	16.1	0.30
S-P 13 (small)	**avena (sativa)**	60	14.1	0.23
S-P 15 (big)	**oenothera biennis (L)**	2510	4.4	0.36
S-P 15 (small)	**oenothera biennis (L)**	3210	4.4	0.36
S-P 16	**calamagrostis epigeios (L) Roth**	70	8.0	0.54
S-P 17	**triticum (clurum vulgare)**	500	9.6	0.40
S-P 18 (big)	**calamagrostis epigeios (L) Roth**	8.4	13.3	0.37
S-P 18 (small)	**calamagrostis epigeios (L) Roth**	8.2	13.8	0.25
Avarage value			**8.8**	**0.37**
Standard deviation			**4.3**	**0.11**

The values of parameter evaluations describing a spread in values of vegetation contamination with ^{137}Cs on experimental sites are illustrated below in Figures (**Fig. 2.21**). The errors of their determination for level of significance 0.05 have been taken into account.

.

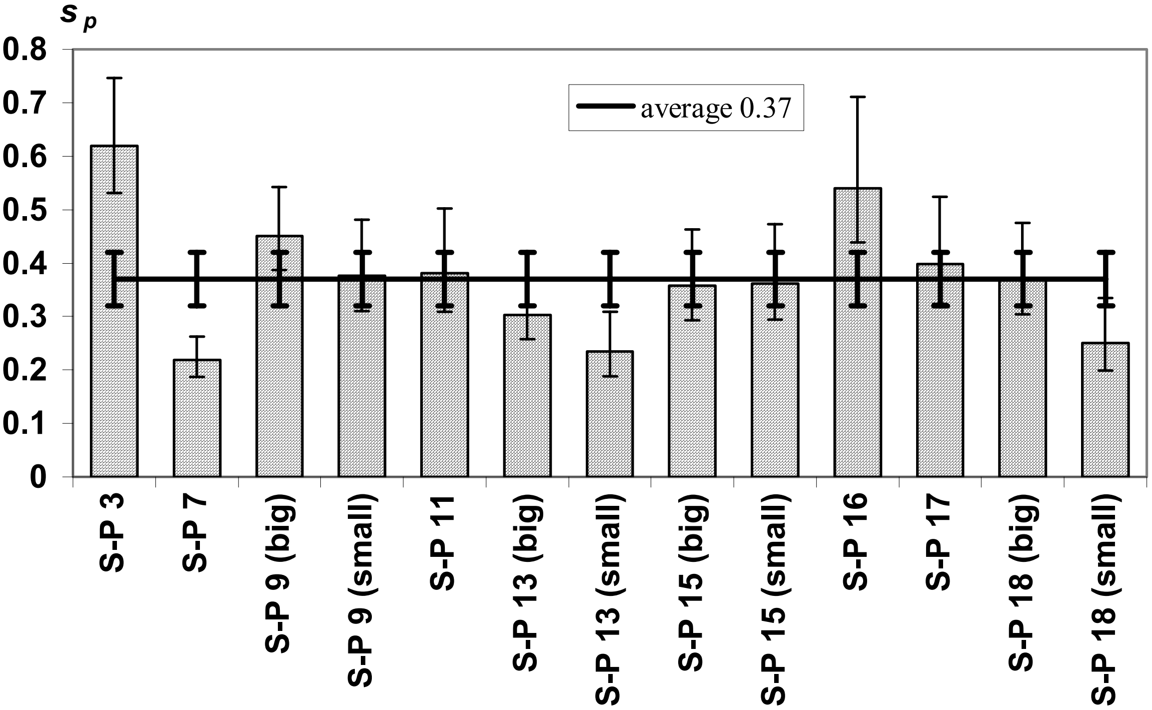


Fig. 2.21.- The values of standard deviation estimations of log of ^{137}Cs specific contents in the plants samples collected from various experimental sites.

The conducted analysis of variance of these outcomes has not shown any dependence of evaluations of parameter s_p on the considered factors (density contamination, type of fallout, type of vegetation and other performances of the site, including its sizes in set limits), and the observable differences have random character.

The analysis of P.F.Bondar's datas [69] for transfer factors of ^{137}Cs in agricultural plants has shown (see chapter 3) that the standard deviation of log of the specific ^{137}Cs content in plant samples does not depend on a type of agricultural plants and their productive parts and is estimated by value 0.38 (±0.14).

2.4 Conclusions under the chapter 2.

As the result of conducted researches the following has been established:

- the soil density contamination with ^{137}Cs and the specific ^{137}Cs content in plants on non-gradient with contamination sites located on the Chernobyl traces and on the global radioactive fallout is satisfactorily described with the logarithmically normal law of probability distributions;
- the standard deviation of log of the soil density contamination with ^{137}Cs of non-gradient with contamination sites for sampling squares > 0.005 m^2 does not depend on density contamination, type of fallout, features of landscape and at first approximation with errors of measurement of the radionuclide content in sub-samples of soil samples ≤ 10 % at the level $\pm 2\sigma$ is evaluated with value **0.30±0.09**;

- the standard deviation of log of the specific ^{137}Cs content in plant samples does not depend on the density contamination, type of fallout, type of vegetation. At first approximation with sampling squares of plants ≤ 1 m^2 and ratio error of measurement of the ^{137}Cs content in sub-samples of vegetative samples ≤ 10 % at the level $\pm\sigma$ is estimated with value 0.37±0.11, on the Chernobyl traces and on global radioactive fallout.

It is necessary to note, that the evaluation of standard deviation of log of the soil density contamination with ^{137}Cs of non-gradient with contamination sites $s_{5,so}$=0.30±0.09 was obtained by the results of soil sampling in 1999. However during the time according to dissolution of fuel particles and decrease of a volumetric heterogeneity of sample in the time it can only decrease. Therefore we can use it as conservative evaluation, in any following year after the accident on the Ch NPP.

3. Statistical characteristics of the transfer factors of ^{137}Cs in a "soil - plant" chain

In the given section the statistical characteristics of the transfer factors of ^{137}Cs in the "soil - plant" chain are considered within the limits of samples, fields, meadows, hay-makings and other arable lands (below they are named simply "samples") which are homogeneous on agrochemical performances of soil. Under their determination the transfer factor is a ratio of the specific radionuclide content in plants to the density of radionuclide contamination in soil (the sample) occupied by these plants. The ^{137}Cs contents in the plants is proportional to the density of the soil contamination, from here the demand of homogeneity of contamination of the ground areas, arable lands, fields, etc. do not superimposed on the obtained below conclusions

The radionuclide transition from a rootdwelled layer of soil into plants is described by the equation

$$C_p = Tf_{so,p} \cdot C_{so} \qquad (3.1)$$

where $Tf_{so,p}$ is the transfer factor in the chain "soil-plant".

Proceeding from the determination of the transfer factor and the results obtained in the sections 2. and 2.3, the transfer factor in the chain "soil - plant" is also a random value, as the quotient of two random values which are the levels of the contamination of soil and plants. On the basis of the conclusions, known in mathematical statistics [30–32], the distribution of probabilities of the transfer factor is described by a logarithmically normal law, as the composition of two logarithmically normal distributions and thus looks like

$$f(Tf_{so,p}) = \frac{1}{\sqrt{2\pi} \cdot Tf_{so,p} \cdot s_k} e^{-\frac{1}{2}\left(\frac{ln(Tf_{so,p})-k}{s_k}\right)^2} \qquad (3.2)$$

where k is the average value of the log of the transfer factor into plants;

s_k is the average quadratic declination of the log of the transfer factor.

Under realisation of the above described field researches on experimental sites the conjugate "soil - plant" samples were being taken. A connected pair of the plant samples and the soil samples selected immediately in the place where the plants grow is interpreted as conjugate "soil - plant" sample The typical spatially - statistical structure of the transfer factors of ^{137}Cs, constructed on the results of these samples measuring, is shown in the figures (Fig. 3.1–Fig. 3.3). Approximation of the probability distribution of possible values of the transfer factors by the logarithmically normal law gives good results. It confirms once again the correctness of earlier selected hypothesis about the logarithmically normal law of soil density contamination of soil and contamination of vegetation with ^{137}Cs on the non-gradient with radionuclides contamination samples.

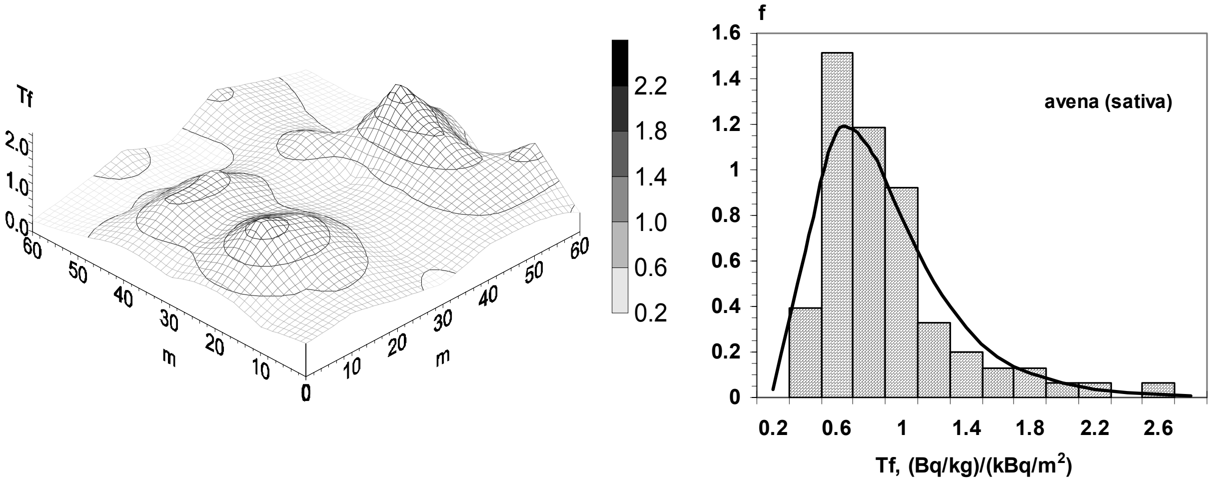


Fig. 3.1.- The spatially-statistical structure of transfer factor of ^{137}Cs in chain "soil - avena (sativa)" on the experimental site **S-P 13**.

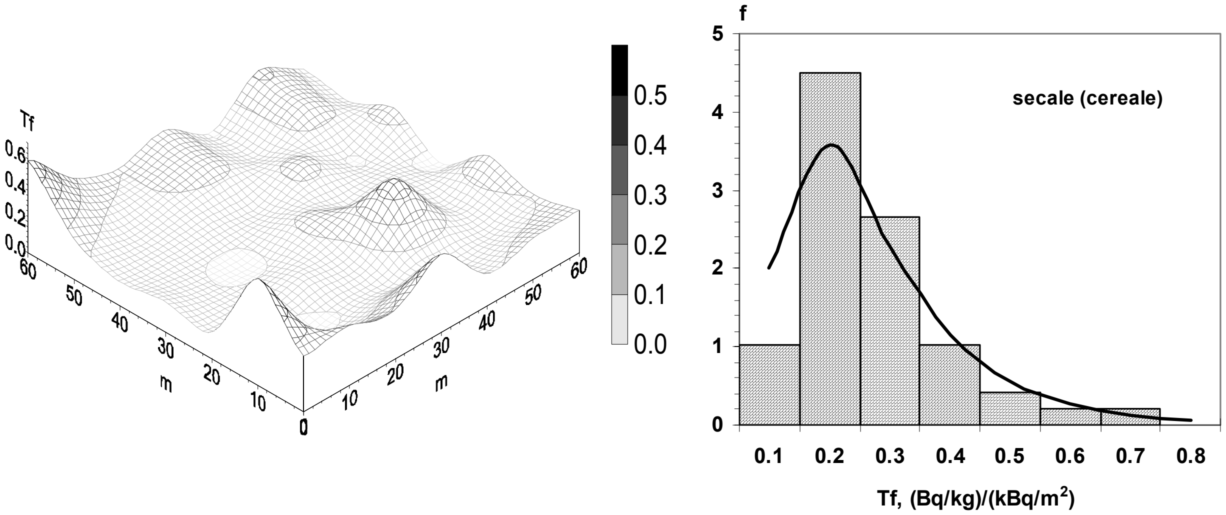


Fig. 3.2.- The spatially-statistical structure of transfer factor of ^{137}Cs in chain "soil - secale (cereale)" on the experimental site **S-P 9.**

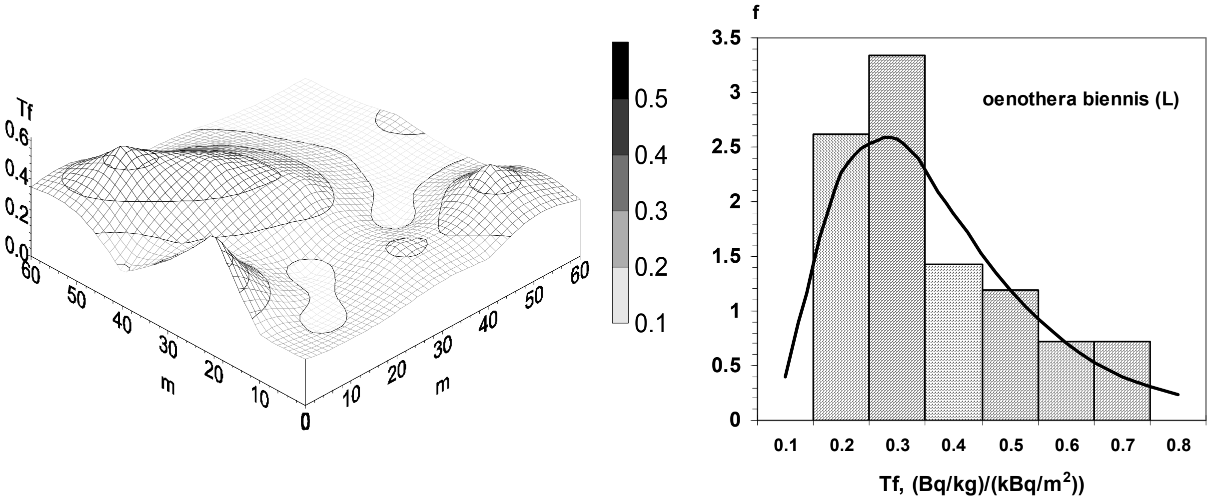


Fig. 3.3.- The spatially-statistical structure of transfer factor of ^{137}Cs in chain "soil - oenothera biennis (L)" on the experimental site **S-P 15**. Oenothera biennis (L) did not grow in the upper corner of the site.

Earlier the similar conclusions about the lognormality of the distribution of the transfer factor of ^{137}Cs in real industrial conditions were made in the work [63]. In this work the distributions of the transfer factors of ^{137}Cs into grain, fodder and vegetable agricultural crops of Ukrainian Polissya on the main types of soils in the regions, polluted with radionuclides, within the Kyiv, Zhytomyr, Rivne and Chernigiv areas have been analysed. The conducted researches have also shown, that the transfer factors normalised on agrochemical parameters of fields are random values and their density of distribution is well approximating by the logarithmically normal law.

3.1 Variability of the transfer factor of ^{137}Cs into plants and influence of various factors on it

Proceeding from the determination of the transfer factor into plants, the dispersion of its log s_k^2 is caused by two factors: by the dispersion of the log of specific content of ^{137}Cs in plants samples and dispersion of the log of the soil density contamination within the experimental sites. The conducted above analysis of valuations of dispersion of the log of the specific content of ^{137}Cs in plant samples and the log of soil density contamination on the experimental sites (which are homogeneous on agrochemical performances of soil) has shown, that the enumerated values do not depend on density contamination, the type of fall-outs, the features of landscape and kind of plants. Therefore, this statement will also be true for the dispersion of the log of the transfer factor.

On the one hand, proceeding from the known statistical conclusions [30–32], the dispersion of the log of the transfer factor of s_k^2 can be calculated as

$$s_k^2 = s_p^2 + s_{so}^2 - 2 \cdot r_{so,p} \cdot s_p \cdot s_{so} \qquad (3.3)$$

where $r_{so,p}$ is the correlation coefficient between the log of soil density contamination with radionuclide and the log of its specific content in plants.

On the other hand, it could be get while immediately processing the results of measurements of the conjugate "soil - plant" samples.

On the non-gradient on contamination samples the dependence between the soil density contamination with radionuclide and its specific content in plants should not be observed, because in this case all the deviations have a random character. The correlation coefficient between the log of the soil density contamination with the radionuclide and the log of its content in plants $r_{so,p}$ should be close to zero. The results of sampling on experimental sites prove this (Fig. 3.4). In the mentioned figure concentrated dependencies of the kind (3.1) are shown by straight lines for each kind of plants (for the transfer factor $Tf_{so,p}$ which is average on a site). It is seen from the mentioned figure, that for experimental observation of the dependence between soil density contamination and plants contamination, under existing conditions of statistical scatters, the distinctions of density contamination within the limits of the experimental site should reach the order of magnitude. It is necessary to note, that the absence of the mentioned correlation in the given experimental data within the limits of one sample, indicates non-gradient of selected experimental sites.

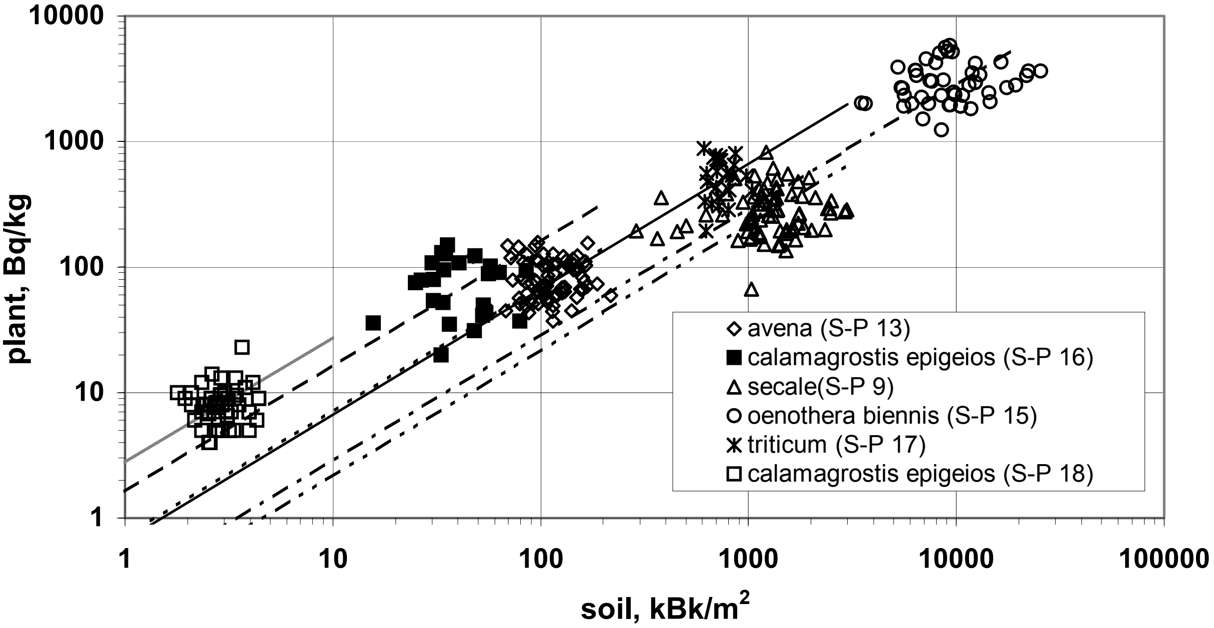


Fig. 3.4– The ratio of soil density contamination with ^{137}Cs to the specific contents ^{137}Cs in the plants on various experimental sites. The lines are appropriate dependence of the kind (3.1).

Proceeding from obtained above estimation the average value (for all the crops and soils) of the average quadratic deviation of the log of the transfer factor s_k calculated on the basis of the expression (3.3) equals

$$s_k = \sqrt{s_p^2 + s_{so}^2} \approx 0.48 \pm 0.10 .$$

On the other hand, we have the values of the average quadratic deviation of the log of the transfer factor of ^{137}Cs for some agricultural and wild plants, grown on experimental sites obtained immediately on the basis of the measurement of the conjugate "soil - plant" samples (Fig. 3.5). The average value of the average quadratic deviation of the log of the transfer factor of ^{137}Cs for all considered plants obtained on the basis of conjugated samples equals 0.49 (±0.06). This value with due regard for available errors of the measurement and statistical scatter coincides with obtained above estimated magnitude. This fact is additional confirmation of non-gradient of contamination of the experimental sites.

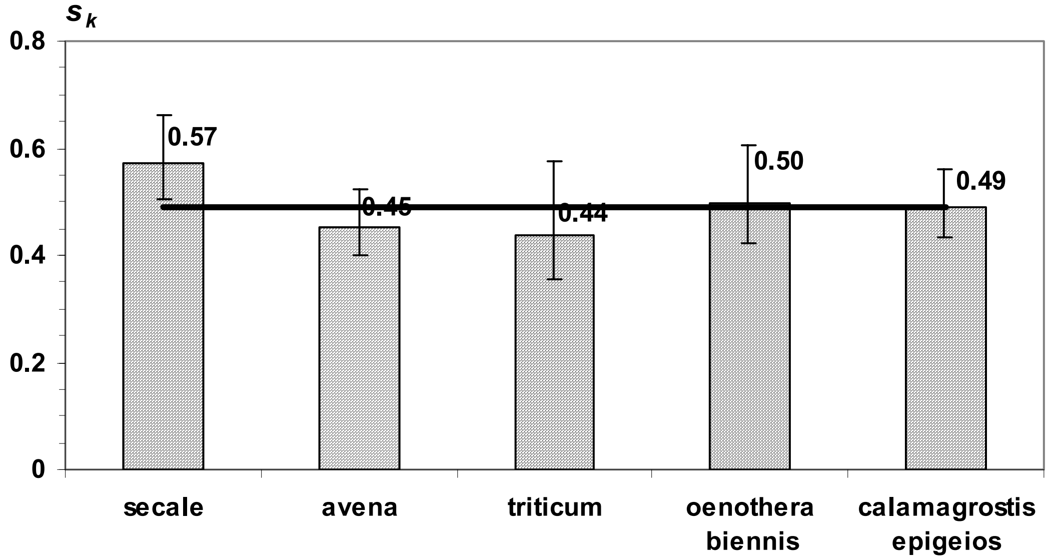


Fig. 3.5– The values of standard deviation of log of the transfer factor of ^{137}Cs into the various plants on the experimental sites.

P.F. Bondar [69,70,71] has made a great contribution into studying biological availability of isotopes of caesium and strontium and experimental determination of their transfer factors in agricultural plants for various types of soils. On the basis of long-term researches (1988-1992 years) he obtained the main regularities on biological availability of these isotopes, when artificially applying them in form of dissolution, as well as immediately on condensation trace of radioactive Chernobyl fallouts.

On the condensation trace of fall-outs the dynamics of the transfer factors of ^{137}Cs into plants is characterised by their gradual reduction. This is mainly caused by evacuation of ^{137}Cs from the rootdwelled layer of soil into subsoil horizons owing to vertical migration and fixing it by a hard phase of soil. Therefore the total variability of the transfer factor for concrete agricultural crop within few years consists of systematic component caused by its dynamics and a random component. The subject of our consideration is a random component. On the basis of dependencies describing the dynamics of the transfer factors of ^{137}Cs on turf-podsolic sandy and sandy loam arable soils [72], obtained under the results of long-term observations during the period of 1986-1994 years, the values of a systematic component of general dispersion of the log of the transfer factor of ^{137}Cs for various agricultural plants during conducted observations from 1989 to 1993 were evaluated. Its median value on all the considered crops is evaluated by magnitude 0.01. Under the statistical analysis of P.F.Bondar's data [69] to obtain the evaluation of dispersion in a definite year the magnitude of the systematic component was subtracted from the total dispersion of the log of the transfer factor of^{137}Cs for each culture. The obtained results are shown below in the figures (Fig. 3.6–Fig. 3.7 – variability of the log of the specific content of ^{137}Cs on each crop, Fig. 3.8 – the average results).

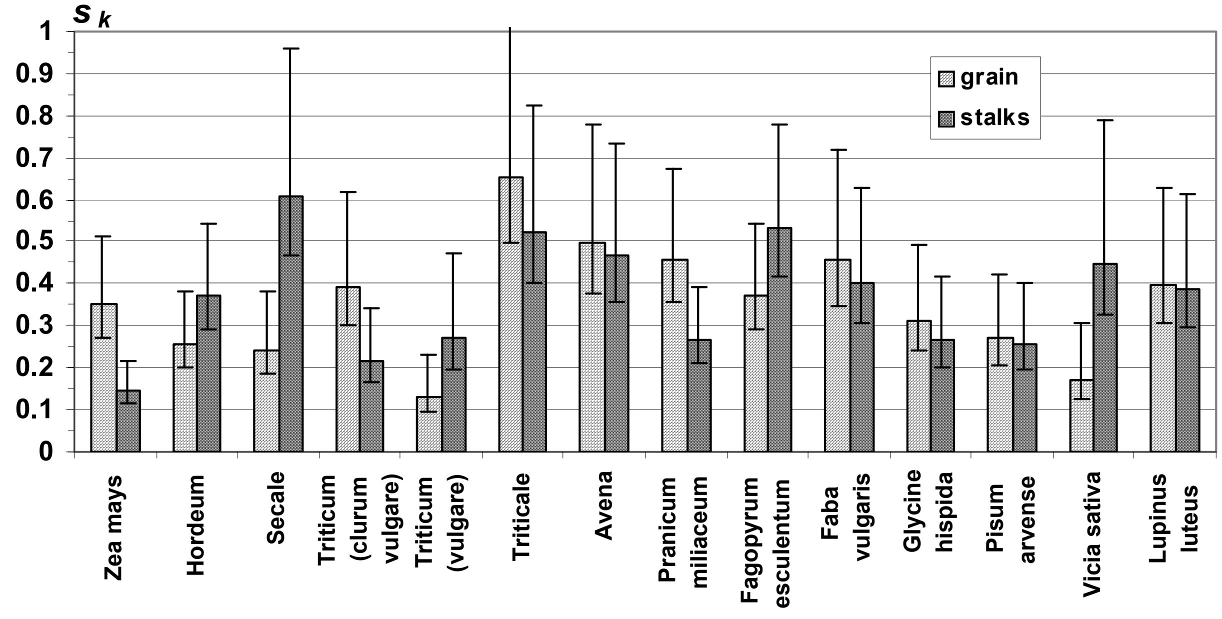


Fig. 3.6– The values of standard deviation of log of the transfer factor of ^{137}Cs into the various agricultural grain-crops in the definite year calculated on P.F. Bondar's data [69].

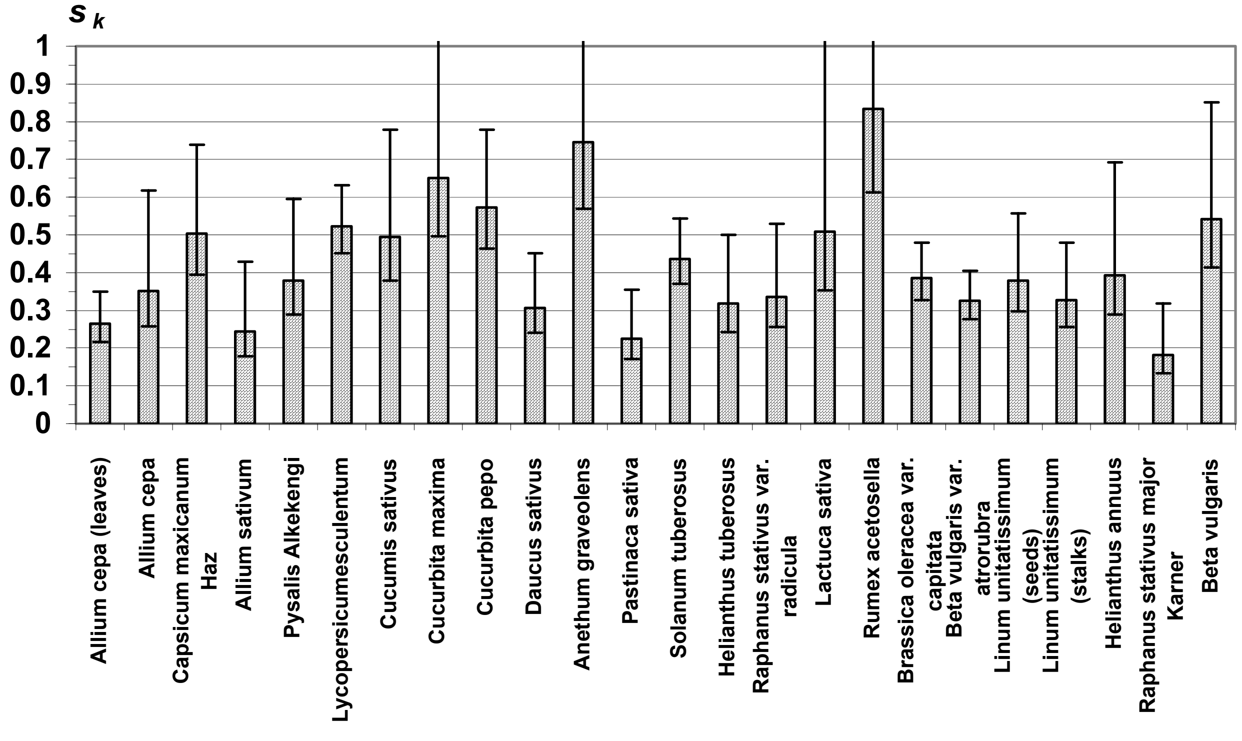


Fig. 3.7– The values of standard deviation of log of the transfer factor of ^{137}Cs into various vegetable and technical crops in the definite year calculated on P.F. Bondar's data [69].

As it is seen in the given results, an inter-specific distinction of average quadratic deviation of the log of the transfer factor of ^{137}Cs in a definite year has a random character. The equality of the variation of the transfer factor is quite obvious for a grain and stems of plants, as the contamination of the grain is proportional to contamination of the stems of plants. Thus, it is possible to consider as the established factor, that the average quadratic deviation of the log of the transfer factor of ^{137}Cs in a definite year does not depend on a kind of plants and their productive parts. The evaluation of this value obtained by us on P.F.Bondar's data [69] equals **0.38**(± 0.14).

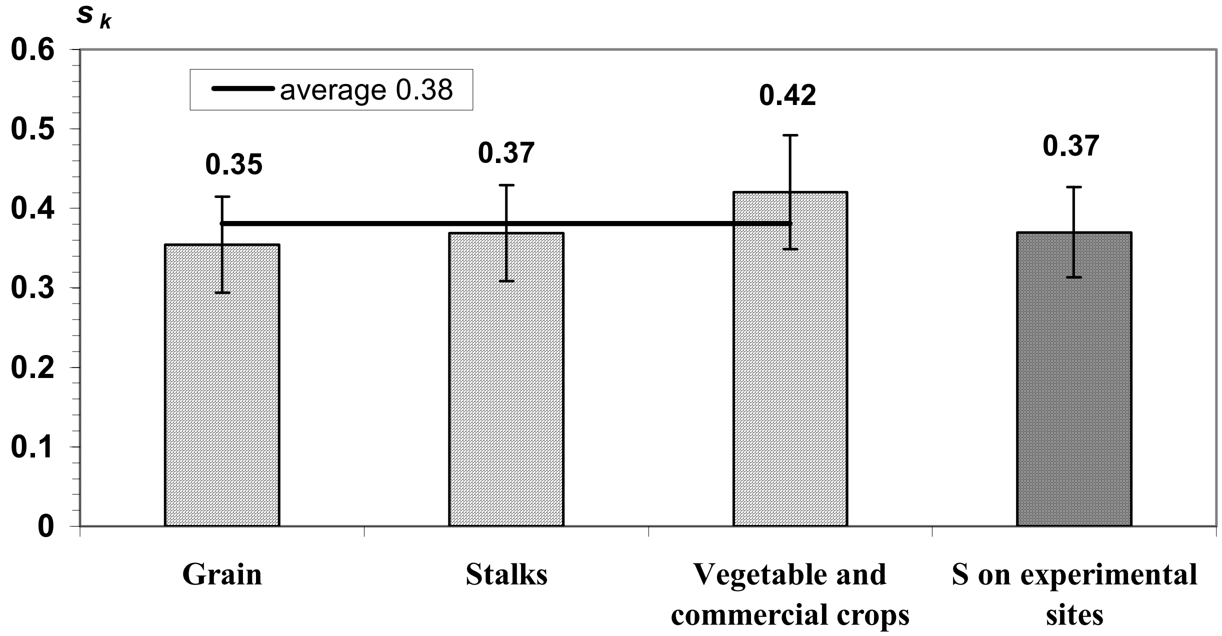


Fig. 3.8– Averaged volumes of standard deviation of log of the transfer factor of ^{137}Cs for various crop production groups in the definite year, calculated on P.F. Bondar's data [69].

It should be noted, that evaluated on P.F.Bondar's data [69] values of average quadratic deviation of the log of the transfer factor of ^{137}Cs into various plants, in the main, are the values of average quadratic deviation of the log of the specific content of ^{137}Cs in plants. It is connected with the fact that in the quoted work when calculating the transfer factor the average for the whole experimental field constant value of soil density contamination with ^{137}Cs was used. Therefore earlier evaluated variability of the ^{137}Cs content in plants on the homogeneous with contamination samples (the value s_p=0.37±0.11) practically coincides with the obtained evaluation of variability of the transfer factor (Fig. 3.8). If the conjugate samples of soil (5 injections by a sampler with a diameter 3.7cm located side by side) were taken on every experimental site, the average value of the average quadratic deviation of the log of the transfer factor of ^{137}Cs would be close to the value $s_k = \sqrt{s_p^2 + s_{so}^2} \approx \sqrt{0.38^2 + 0.31^2} = 0.48 \pm 0.12$. This value with due regard for available errors coincides with the value 0.49 (±0.06), which has been obtained on the experimental sites (Fig. 3.5).

Let us accept the average value equals 0.49(±0.06) in the first approximation for further accounts as the evaluation of the average quadratic deviation of the log of the transfer factor of ^{137}Cs on one field (sample) when accepting the above rule of selection of the conjugate "soil - plant" samples (the sampling square for plants ≤ $1m^2$ and > 0.005 m^2 for soil) irrespective of a kind of plants, type of soil and type of fall-outs.

In conclusion it is necessary to mention about one delusion, which can arise when analysing observed transfer factors and disregarding the statistical nature of estimations of radionuclides content in soil and plants received in the results of measurements. If formally construct the dependence of observing transfer factors from the same values of soil density contamination in the place of selection of the conjugate "soil - plant" samples, which were used for calculation of these transfer factors, it will look like the following (Fig. 3.9a). If the same observed transfer factors compare with the values of soil density contamination in the place of a selection of conjugate samples determined independently (on the results of a measurement of the another soil sample selected in the same place), then it will not be any dependence between considered magnitudes (Fig. 3.9b). This reflects a true position of things. The ratio, given on the figures, were obtained by the method of statistical modelling of soil and plants sampling on the sample which is non-gradient on radioactive contamination and has obtained above statistical characteristics ($s_{5,so}$=0.30±0.09; s_p=0.37±0.11).

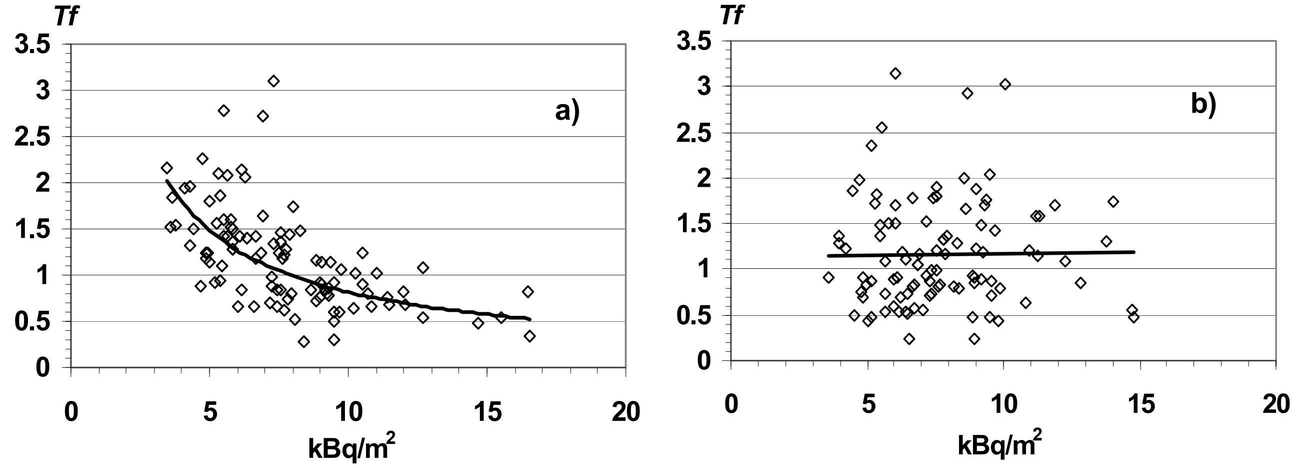


Fig. 3.9.- The ratio between observed transfer factors and soil density contamination: a) volumes of soil density contamination same to accounting Tf; b) independence volumes of soil density contamination into point of sampling of conjugate samples.

3.2 Conclusions on the chapter 3.

In the results of the conducted researches it is established that within the limits of the samples homogeneous on agrochemical performances of soil:

- the transfer factor of ^{137}Cs into plants is the random variable and it is described by the logarithmically normal law of the probability distribution;
- the average quadratic deviation of the log of the transfer factor of ^{137}Cs in plants does not depend on the density of contamination, the type of fall-outs, the kind of vegetation and its various parts; in the first approximation when se-

lecting the conjugate "soil - plant" samples, when squares of sampling for plants ≤ $1 m^2$ and > $0.005\ m^2$ for soil and relative error of measurement of the ^{137}Cs content in the samples of soil and plants does not exceed 10 % at a level $\pm\sigma$ is evaluated by the value 0.49 ± 0.06.

4. Statistical and temporary performances of milk contamination with ^{137}Cs in settlements

If talk about animal produce, milk is the most susceptible product to oscillations of the contamination level of cattle ration. A range of these oscillations (difference between minimum and maximum contamination levels) may reach as the value of this level or more depending on agroclimatic conditions during a pasturable period. Conditions of keeping cattle (stabling or pasturable) and the structure of cattle ration influences the milk contamination too. It is mentioned in the literature, that in a public sector the milk contamination with ^{137}Cs during the pasturable period is on the average in 1.4 times to milk contamination with ^{137}Cs in stabling period [72,73,74], however this figure may be changed depending on concrete conditions.

The analysis of numerous data on the milk contamination with ^{137}Cs both in a public sector, and in private small farms (PSF) accumulated during the post emergency period allows to consider the milk contamination in a settlement or in a farm during some period (for example, during a calendar year) as a magnitude that has stochastic nature. In a definite time the ^{137}Cs content in milk can be considered as a random variable with some law of probability distribution. Not going to detail on the nature of this chance, we note, that it is a consequence of a heterogeneity of contamination with ^{137}Cs of agricultural lands and natural heterogeneity of soil characteristics and it is conditioned by the specific features of plants and their portion in structure of cattle rations, individual features of animals, as well as by the influence of climatic factors etc. Inevitable errors of conducted measurements are also superimposed on these factors. Therefore, the first problem, that is the previous to determination of the necessary number of samples for an authentic evaluation of the milk contamination level in a definite settlement, is the analysis of statistical performances of milk contamination with ^{137}Cs in these settlements.

Eight settlements, in which the UIAR basic radiological points have been located, were selected for investigation the statistical performances of the specific ^{137}Cs content in cow's milk. Monthly volume of the milk samples, taken during 1997, is given in the table (Table 4.1).

Table 4.1. The number of a dairy herd and the monthly number of inspected cattle.

№	**Settlement**	**Coomb**	**Number of head in the herd (1997)**	**Monthly volume of selection**
M 1	**Polissya region, Ragivka v.**		166	9-18 (July-31)
M 2	Polisske region, Lugovyky v.		86	9-10
M 3	Tarashcha region, Kovshuvate v.	Klebaniha	135	9-13
		Budyatsky Yar	74	9-10
M 4	Ovruch region, Gladkovychy v.		447	10-18 (June-89)
M 5	Luginsk region, Lipniky v.		348	16-23

M 6	Narodichy region, Selets v.	Risy	105	13-19
		Brodok	71	14-15
M 7	Chernigiv region, Pacul' v.		399	9-18 (June-38)
M 8	Dubrovichy region, Milyachy v.		540	16-21 (July-32)

The characteristic statistical structure and dynamics of the milk contamination during the year for some settlements is shown in the figures (Fig. 4.1–Fig. 4.4). In the given figures the median values of milk contamination with ^{137}Cs (annual trend) and the upper values shown with confidential probability p=0.9 are shown. A quantity of herds, pastured on various pastures, and their number can exert a significant influence on the milk contamination with ^{137}Cs in the settlement at a definite moment of the pasturable period (Fig. 4.2).

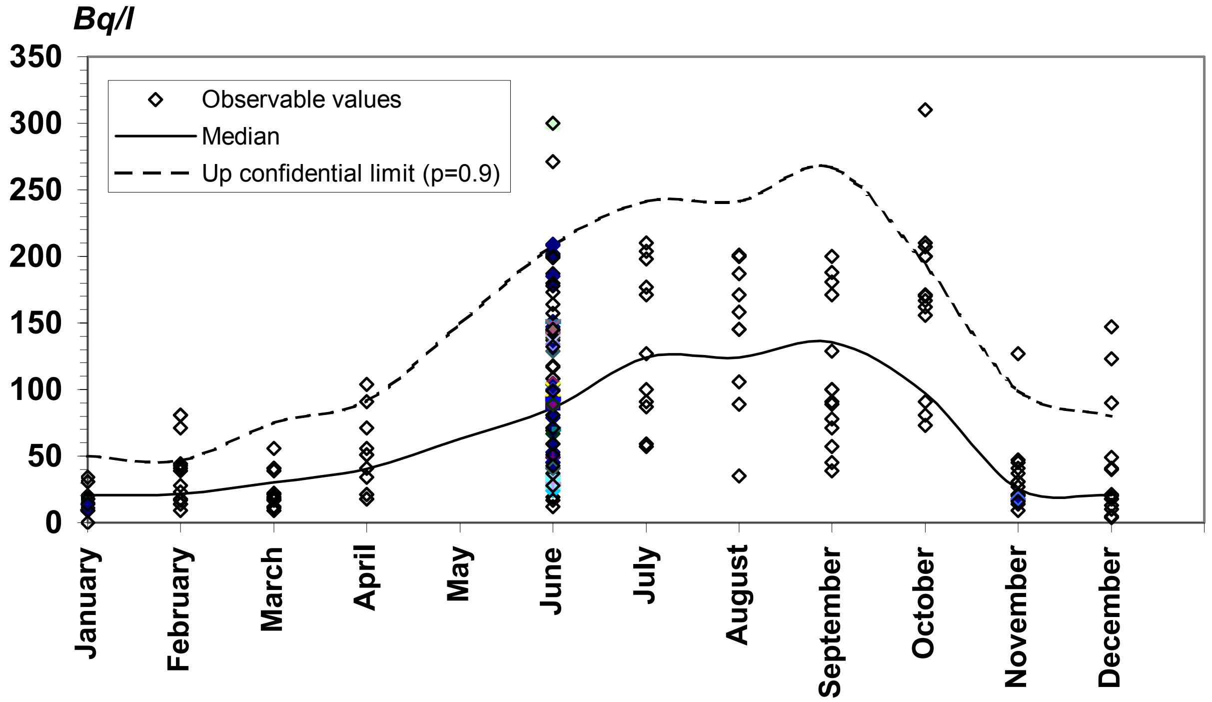


Fig. 4.1. - Annual dynamics of milk contamination with ^{137}Cs for the site **M4**.

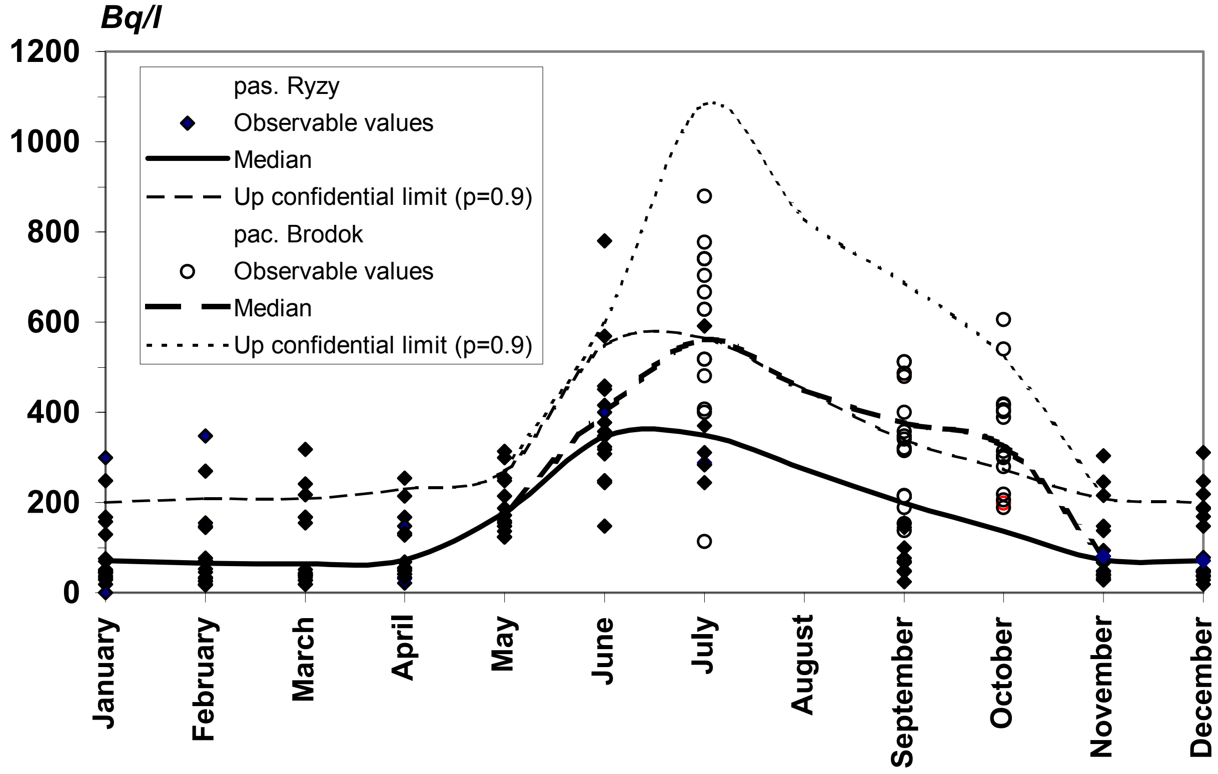


Fig. 4.2. - Annual dynamics of milk contamination with ^{137}Cs for the site **M6**.

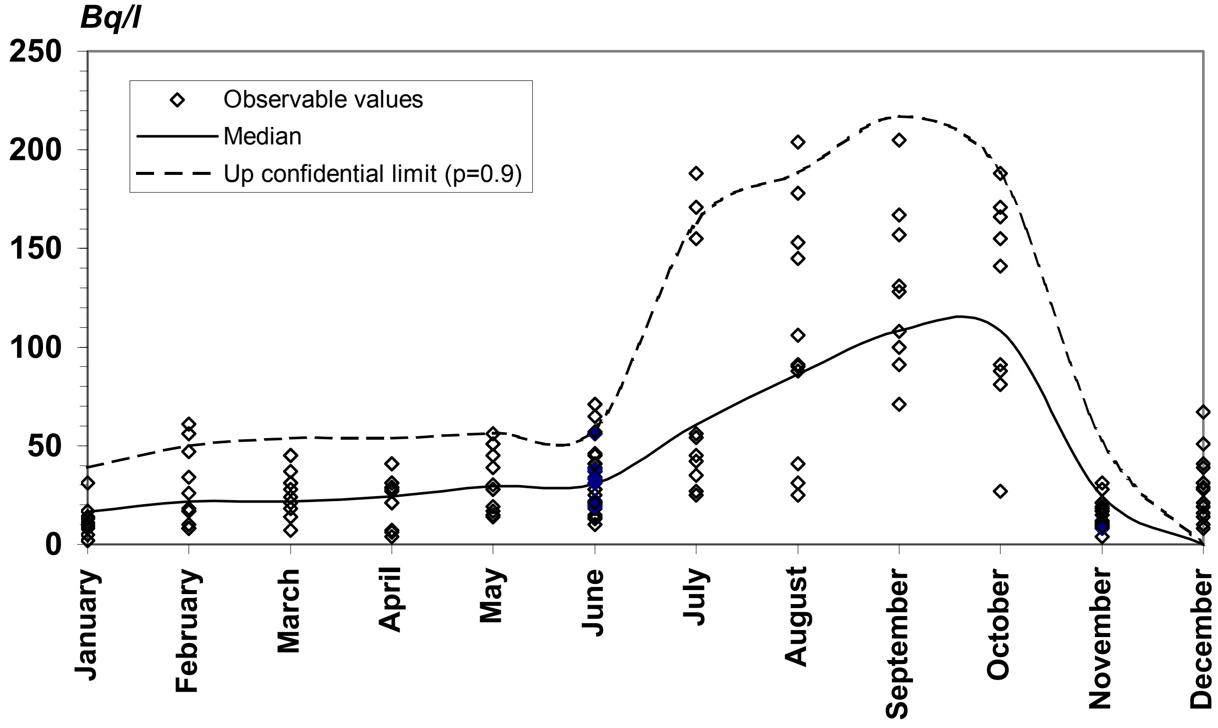


Fig. 4.3. - Annual dynamics of milk contamination with ^{137}Cs for the site **M7**.

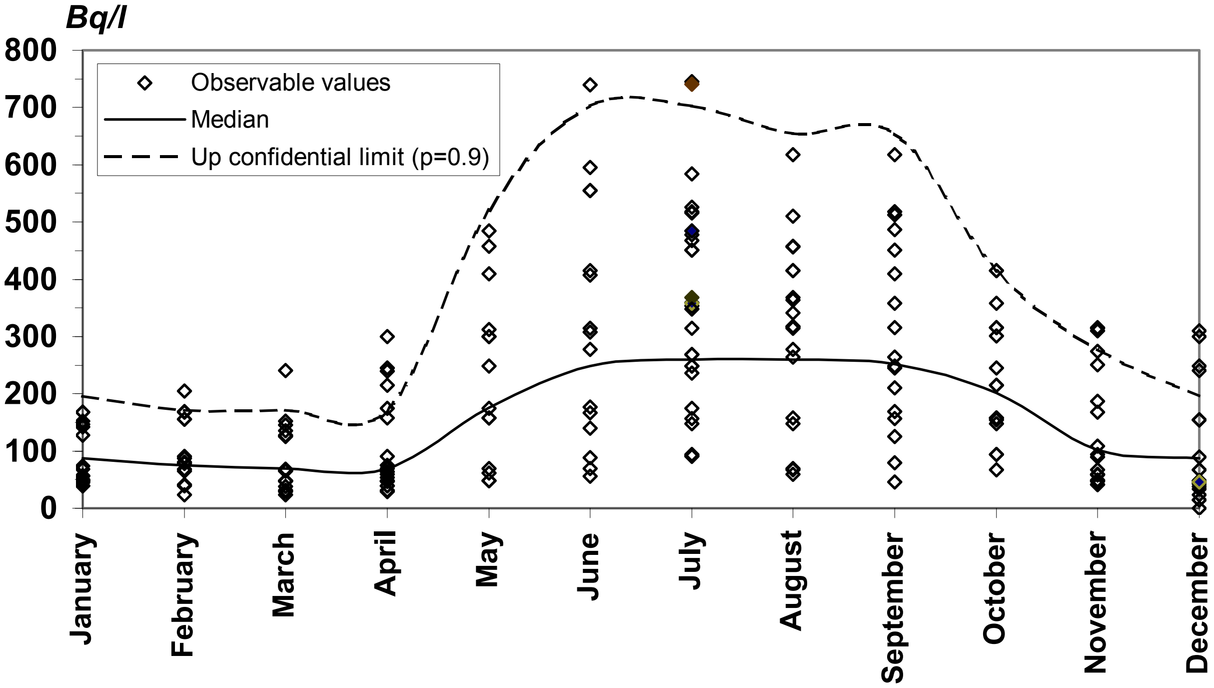


Fig. 4.4. - Annual dynamics of milk contamination with ^{137}Cs for site **M8**.

Statistical processing of obtained data and analysis of numerous results of ^{137}Cs concentration in milk measurement in the settlements located in different regions, show, that the specific activity of milk at a considered time moment has a significant scatter and right-hand asymmetry. The reasons causing the scatter of ^{137}Cs concentration in milk have been described in detail in the literature (for example in [75]). The processing of the statistical material obtained in numerous experiments with different groups of experimental cattle and observation of the animals, in natural conditions, have shown, that ^{137}Cs concentration in cows milk at time moment t $C_m(t)$, as especially positive magnitude, is well described by the logarithmically normal law of probability distribution [76],which in this case is like the following

$$f(C_m(t)) = \frac{1}{\sqrt{2\pi}\cdot C_m(t)\cdot s_m(t)}e^{-\frac{1}{2}\left(\frac{ln(C_m(t))-\mu_m(t)}{s_m(t)}\right)^2} \qquad (4.1)$$

where $\mu_m(t)$ is the average value of the log of ^{137}Cs concentration in milk in the t time moment; $s_m(t)$ is the standard deviation of the log of ^{137}Cs concentration in milk in the t time moment.

The conducted statistical analysis of the results of the milk samples measurement, taken in the settlements has shown, that the probability distribution for monthly average ^{137}Cs concentration in cow's milk $C_m(t)$ at definite t time moment, in the settlement (within one herd) both in stabling and pasturable period, does not contradict the logarithmically normal law.

This is shown as an example in figure (Fig. 4.5). This is shown as an example in figure (Fig. 4.5) for one settlement as in characteristic months of the stabling and pasturable keeping of cattle, and as a whole for these periods. For histogram construction for stabling and pasturable periods, the milk contamination with ^{137}Cs, selected during a month, has standardised on corresponding median.

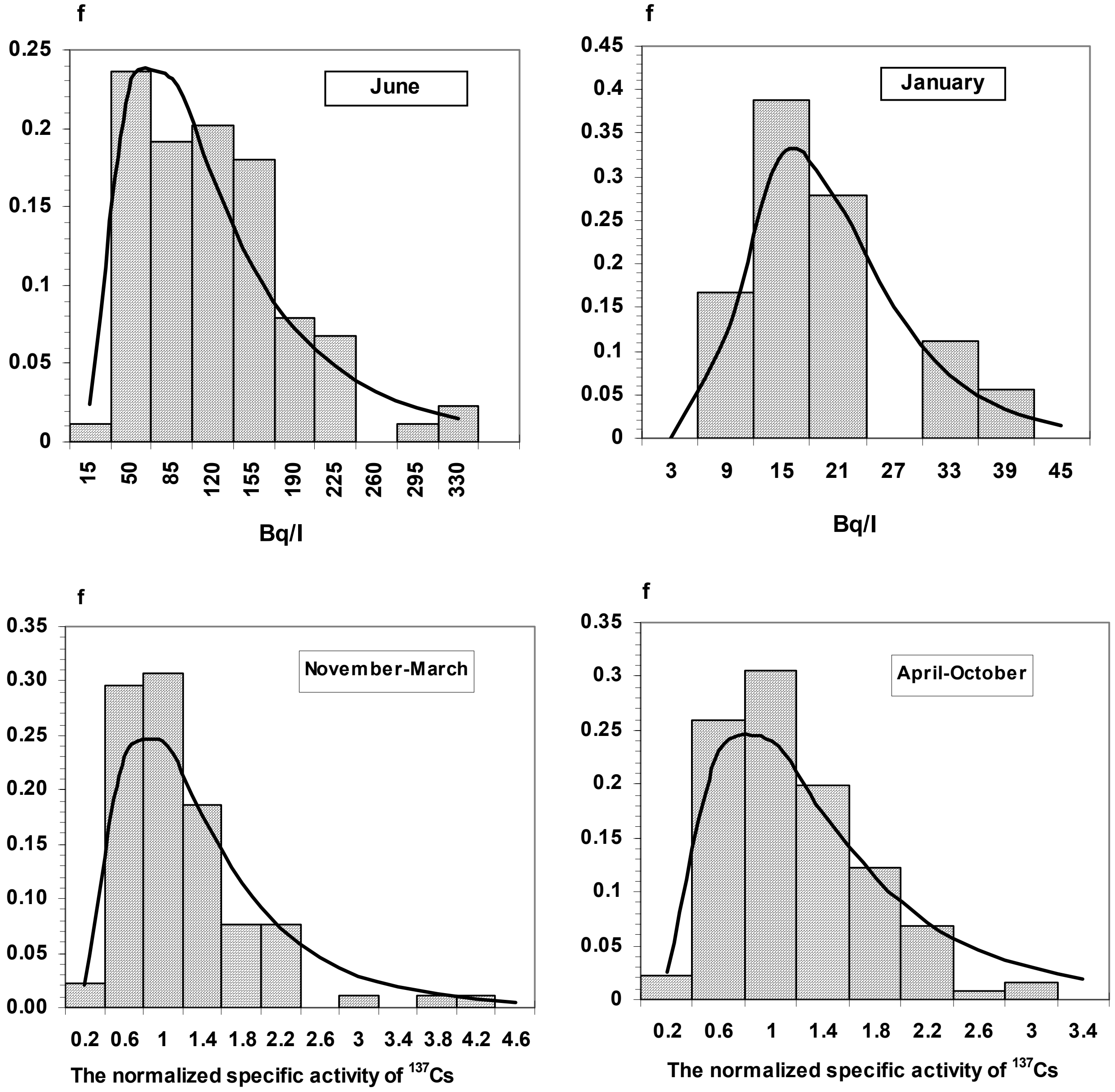


Fig. 4.5. – Histograms of milk contamination with ^{137}Cs for the site **M4** in typical months of stabling and grazing periods and their approximation by the logarithmically normal law.

The logarithmically normal law of probability distribution will be used in future when evaluating and analysing of statistical performances of milk contamination in the settlements.

4.1 Analysis of dynamics and statistical performances of milk contamination with ^{137}Cs in the settlements

Typical results of approximation of observed dynamics of milk contamination with ^{137}Cs in the settlements are shown in figures (Fig. 4.1–Fig. 4.4) by continuous function. The approximation is conducted as following. The parameters of $\mu_m(t)$ and $s_m(t)$ distribution were evaluated on the basis of obtained results for every month. A median of milk contamination with ^{137}Cs was calculated on $\overline{C}_m(t)=exp(\mu_m(t))$ formula. The upper bound of observed values with q=0.1 level of meaningfulness (there is no more than 10 % of observed values above it) was calculated on $C_m^{up}(t)=exp(\mu_m(t)+1.28\cdot s_m(t))$ formula. The obtained values were smoothed out by the method of the sliding median /66/ on three points when constructing a diagram.

The statistical analysis of the results of conducted measurements in the settlements of Ragivka and Lugovyky in the Polisske region located along the Poliss'ke-Kyiv highway has shown statistically insignificant difference in the specific content of ^{137}Cs in cow's milk. In this connection, when considering and analysing, the results of measurements of the specific content of ^{137}Cs in cow's milk in these settlements were united, and this increases the volume of statistical data and therefore the truth of received estimations. For the same reason the results of measurements of the specific ^{137}Cs content in cow's milk in the settlement of Kovshevate of the Tarashcha region obtained for different pastures (coomb) were united. The results of measurements of the specific ^{137}Cs contents in cow's milk in the settlement of Selets of the Narodichy region obtained for different pastures (coomb), have been analysed separately because of statistically important difference (Fig. 4.2) between them.

As it is seen from the given figures, the seasonal specific ^{137}Cs content in cow's milk in the investigated settlements, conforms to the regularities mentioned in [74]. However the dynamics of the specific ^{137}Cs content in milk for every settlement has its own characteristic features.

The median values of the specific ^{137}Cs content in milk were normalised on the average specific ^{137}Cs content in milk, which is characteristic for the stabling period (November - March) to compare and analyse these features for every settlements more conveniently. The obtained results are given in figure (Fig. 4.6), where the multiplicity of increase in milk contamination with ^{137}Cs in the pasturable period according to stabling period is given on ordinate axis. It is seen from this figure, that the median of the specific content of^{137}Cs in milk can increase from 2 up to 8 times in the investigated settlements during the pasturable period. The maximum specific^{137}Cs content in milk is during the spring and summer period in some settlements, but in the other settlements - during the autumn and summer period. Two groups of the settlements were selected. The first one is a classical case [74], and it is characterised by more or less uniform, increase rather low one (on the average in 2-3 times) of the specific^{137}Cs content in milk during all pasturable period. In the second group the increase of the specific ^{137}Cs content in milk during the pasturable period has a strongly expressed peak character, with significant increase at the peak moment (in 6-8 times).

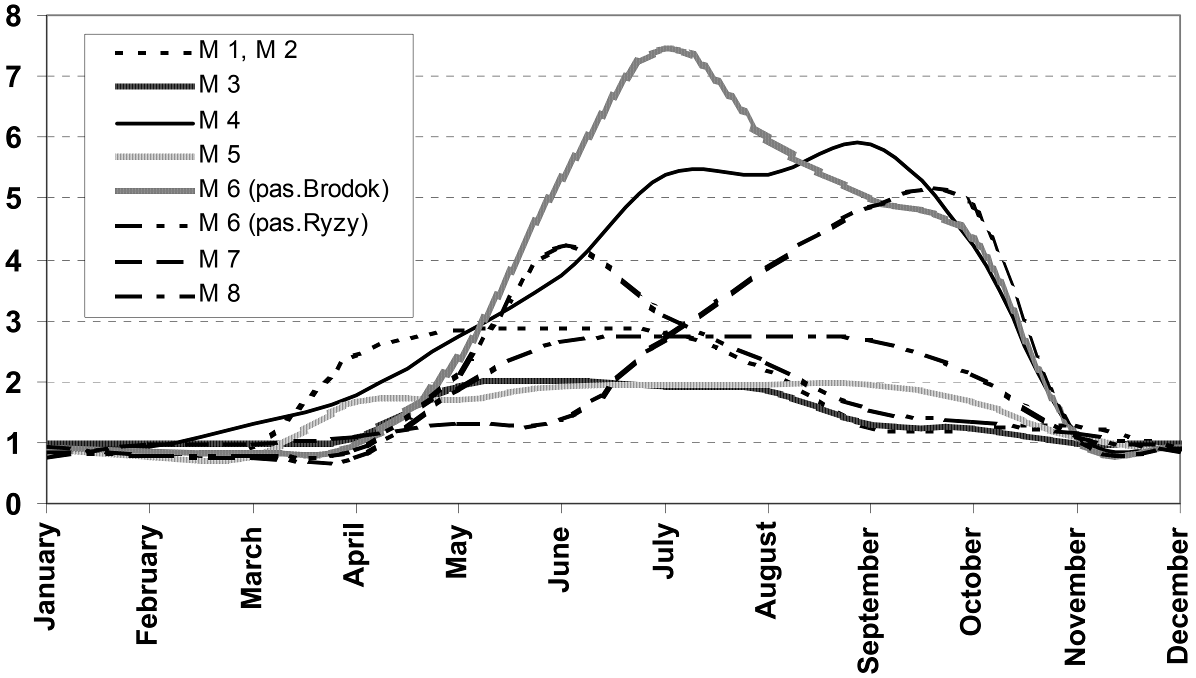


Fig. 4.6.- Multiplicity of milk contamination with ^{137}Cs increase in grazing period comparing to stabling period in different settlements.

The variability of observing values during the year is another important statistical performance of the ^{137}Cs content in milk. The s_m value which is the standard deviation of the log of the ^{137}Cs concentration in milk is considered as the performance of variability according the selected law of probability distribution (4.1) The choice of s_m value for study milk contamination variability with radionuclides and its performances in the settlements have been made on the same reasons, as for study variability of soil and plants density contamination (see sections 2.2.1 and 2.3.1). The seasonal modifications of this performance are shown below in figure (Fig. 4.7). The proper trustworthy intervals under the trustworthy probability p=0.9 for calculated s_m values are given there, too.

It is seen in the given figure, that the variability of the specific ^{137}Cs content in cow's milk has a seasonal character too. One of the reasons that explain this fact is: the ration of cattle, belonging to different owners, has mainly the same ^{137}Cs contamination during the pasturable period (the animals are pastured together). During the stabling period the contamination of cattle's ration is more individual (owners lay in hay for the cattle on different lands and combine it with individual feeding).

The researches and analysis of cow's milk contamination with ^{137}Cs [**Ошибка! Закладка не определена.**,72] conducted earlier, in 1990-1992 in UIAR have shown, that in the spring-summer period the standard deviation of the log of the specific content of ^{137}Cs in milk, for the settlements of the Kozeletsk region in the Chernigiv area, is within the limits of $0.15 < s_m < 0.30$, and it is within the limits of $0.16 < s_m < 0.40$ for the settlements of the Kamen'-Kashyrsky, Lubeshovsky and Manevichsky regions in the Volyn area.

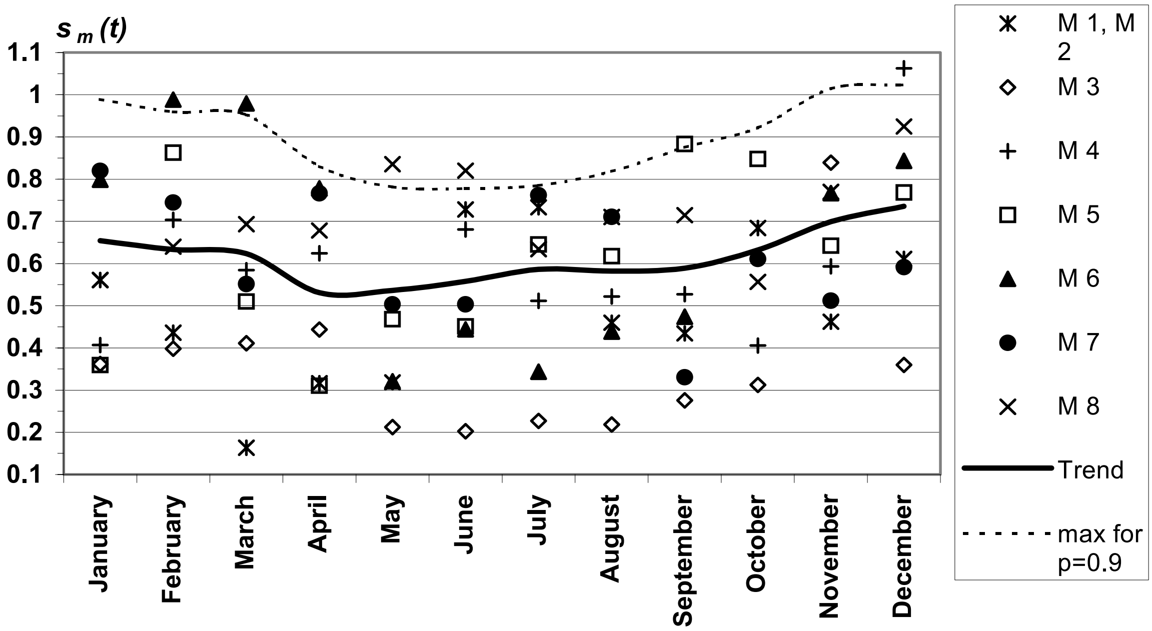


Fig. 4.7. - Annual dynamics of standard deviation of the log of milk contamination with ^{137}Cs in settlements.

As it is seen from the comparison, the values of standard deviation of the log of the ^{137}Cs concentration in cow's milk in settlements, obtained in the given work, are higher, than the values in quoted literature sources and they equal on the average: 0.67 for the stabling period (November - March); 0.56 for the pasturable period (April - October). The upper limits of the average values accordingly equal to 0.9 and 0.8.

4.2 Stochastic model of the milk contamination with ^{137}Cs in the settlements

Annual dynamics of the contamination with ^{137}Cs of cow's milk in definite region, settlement and cattle's herd, pastured on the definite pastures (in the coomb) has its specific features. It is expressed as in absolute and relative (in relation to milk contamination during the stabling period) value of peak of milk contamination with ^{137}Cs and at the moment of this peak approach. The highest milk contamination with ^{137}Cs takes place for one group of animals in the spring-summer period (April - July), for the others - in the autumn-summer period (July - October), and for the third group it take place practically during the all pasturable period (May - September). All this indicates that it is necessary to group animals on the lands, where they are pastured to get authentic evaluations of the cow's milk contamination level with ^{137}Cs in the definite settlement during the pasturable period. This problem is facilitated when the cattle are being pastured as one or two herds, every one on its pastures or coomb. We consider each of the herds as elementary group of cattle, and a stochastic model of the specific content of ^{137}Cs in milk is proposed for it below. It is necessary to note, that cattle binding to the pastures and coomb (forming elementary groups of animals) is necessary only during the pasturable period (see Fig. 4.6). During the stabling period it is possible to consider all cattle in the given settlement as the elementary group of animals.

When creating a network of UIAR basic radiological points, the basic farms were selected to cover all possible regional features causing contamination of agricultural production (including milk) with radionuclides on the territory, polluted in the result of the accident on Ch NPP. Thus, it is possible to consider, at the first approximation the variability of the ^{137}Cs content in milk in investigated PSF of the settlements on the whole is typical for the territory undergone radioactive contamination in the result of the accident on the Ch NPP. The statistical analysis of a milk variability parameter (s_m - Fig. 4.7) has not revealed the characteristic peculiarities connected with settlements' disposition, and its oscillation from one settlement to another one has a casual character. Therefore we consider the s_m parameter as a random variable, which is subjected at the first approximation to the normal law of probability distribution. The performances of this random variable are indicated below in table 4.2, and obtained standard deviation does not depend on the period of keeping cattle.

Table 4.2.- Statistical performances of standard deviation of the log of ^{137}Cs concentration in milk of PSF

Characteristics	Logarithm of ^{137}Cs concentration in milk	
	Stabling period	Pasturable period
Average value	0.67	0.56
Standard deviation	0.19	0.19
Coefficient of variation	0.28	0.34

The statistical data analysis for all considered settlements was conducted to reveal correlative connections between the results of measurements conducted during adjacent months. It has shown the absence of correlative connection between the results within the stabling period and within the pasturable one (a module of a correlation coefficient r~0.1) and the presence of the connection on the boundaries of the periods (the module of the correlation coefficient r~0.6). Therefore we consider at the first approximation, that the ^{137}Cs concentration in milk of elementary cattle's group for every time moment t is a random variable with the logarithmically normal law of a probability distribution at every time moment during the year (4.1). This distribution for every period has its spread parameter (s_m is the standard deviation of the log of ^{137}Cs concentration in milk). Then is possible to use the following stochastic model for statistical description of ^{137}Cs concentration in milk.

$$f(C_m(t))=\begin{cases}\dfrac{1}{\sqrt{2\pi}C_m(t)\cdot s_m^{st}}e^{-\frac{1}{2}\left(\frac{ln(C_m(t))-\mu_m^{st}(t)}{s_{м}^{c}}\right)^2} & (t\supset november\text{ - }march\text{ }) \\ \dfrac{1}{\sqrt{2\pi}C_m(t)\cdot s_m^{pas}}e^{-\frac{1}{2}\left(\frac{ln(C_m(t))-\mu_m^{pas}(t)}{s_{м}^{n}}\right)^2} & (t\supset april-октябрь\text{ }) \end{cases} \quad (4.2)$$

where $\mu_m^{st}(t)\left\{\mu_m^{pas}(t)\right\}$ is the average value of the log of ^{137}Cs concentration in milk during the stabling (pasturable) period; $s_m^{st}(s_m^{pas})$ is the standard deviation of the log of ^{137}Cs concentration in milk during the stabling (pasturable) period.

The offered stochastic model can be simplified if consider, that the dynamics of the milk contamination (a classical case) is absent within the pasturable and stabling periods and the difference in the average contamination levels exists between the periods only. However, such model of the description of ^{137}Cs concentration in milk for the pasturable period is rough enough (see Fig. 4.6). But if evaluate the average log of ^{137}Cs concentration in milk during the pasturable period by the results of the measurement of ^{137}Cs in milk in the most critical period (the period of the highest ^{137}Cs concentration in milk), the evaluations, obtained on the basis of simplified model, will be conservative (in store).

As it has already been mentioned, the critical period for every elementary group of cattle is individual and it is determined by both regional, and local conditions and furthermore it will have oscillations year after year. Every settlement which was subjected to radioactive contamination, has to be inspected monthly (at least during the pasturable period) within several years to obtain the most authentic valuations (determination of this period and its annual oscillations). In connection with a large labour-consuming character of this problem and probabilistic nature of the most critical period coming (the time of the highest ^{137}Cs concentration in milk), the following assumption has been made: the most critical period coming does not depend on the settlement but it is connected with weather conditions and has a casual character. It follows from this fact, that the highest ^{137}Cs concentration in milk (a median value) may be realised at any moment of the pasturable period depending on a definite year and weather conditions.

Thus, the evaluation of the parameters $\mu_m^{st}(t)\left\{\mu_m^{pas}(t)\right\}$ for every elementary group of cattle has to be executed annually during the pasturable and stabling period.

The parameters describing a scatter of ^{137}Cs concentration in milk $s_m^{st}(s_m^{pas})$, has to be defined annually more exactly in data totality for every period. At the first approximation to obtain the conservative valuations connected with milk contamination with ^{137}Cs in the settlements of all territory subjected to radioactive contamination in the result of the accident on Ch NPP , they should be accepted equal to $s_m^{st}=0.9; s_m^{pas}=0.8$.

4.3 Conclusions on chapter 4.

In the results of conducted researches:

- It has been established, that milk contamination with ^{137}Cs in the settlements at specific time is described satisfactorily by the logarithmically normal law of probability distribution;
- The statistical parameters of cow's milk contamination of PSF with ^{137}Cs during the pasturable and stabling periods have been evaluated; it was obtained, that the relative variability of ^{137}Cs content in cow's milk in the settlements does not depend on regional features and is determined by the conditions of the keeping cattle only (the stabling period and the pasturable period);
- The standard deviation of the log of the specific content of ^{137}Cs in cow's milk does not depend on the location of the settlements; it equals 0.67±0.19 during the stabling period and 0.56±0.19 during the pasturable period for homogeneous on conditions of pasture (keeping) cattle herbs if measurement errors do not exceed 15 % at a level $\pm 2\sigma$;

The stochastic model for the description of cow's milk contamination with ^{137}Cs during a year in the settlements has been proposed.

5. Radioactivity measurement in heterogeneously contaminated samples

As it has already been noted, the contamination of the environment with radioactive particles had become one of the results of the Chernobyl catastrophe. Th0se particles were either fine dispergated nuclear fuel of the destroyed block (fuel particles), or initially inert aerosol particles which during the accident become the condensation centres of highly mobile volatile radioisotopes of iodine, caesium, ruthenium oxides etc. (condensation particles). Despite a considerable amount of time past after the accident, the fuel particles are still the great factor of soil contamination, in particular in the boundaries within the 30-km zone of Ch NPP [2,3,4]. The presence of such particles in selected samples can essentially influence statistical reliability of the result of a single measurement. It is stipulated by the fact that preparation process of such sample for a measurement by a standard technique [77](soil drying up to an air - dry condition, grinding of clots and sifting through a sieve with an orifices 1 mm in a diameter) does not ensure homogeneity of activity distribution in all sample volume (homogenisation of the sample). An instrument measurement error of radionuclides activity in measured sub-samples (the part of the sample) will not characterise the error of radionuclides determination in the samples as a whole. And this error determines reliability of estimated parameters and performances: a contamination density of the territory, soil and vegetation specific activity, the radioactive aerosols concentration in the atmosphere and intensity of their fall-out etc [78, 79, 80]. It is the evidence of necessity to improve the methods of carrying out gamma - spectrometric, radiometric and radiochemical measurements of samples activity and correct evaluation of measurement error under condition of volumetric heterogeneity contamination of the selected samples.

To measure radioactivity in the samples of loose materials and liquid by γ-spectrometer, the measuring vessels of various geometry were used. Now the Marinelle flasks, which capacities of 0.5 and 1 liter and cylindrical flacks with a capacity of one litre and less are mostly, distributed. The instrument error of the measurement and the error of determination of radionuclides content in the sample depend on either that flask (geometry of the measured sample) use. In the present section the errors of determination of radionuclides content in the sample when using three geometry will be analysed: the Marinelle flask 1000 cm^3; the cylindrical measuring flask 1000 cm^3 (diameter: 9.1 cm, height: 15.4 cm), widely distributed in West European countries; the cylindrical measuring vessel of 100 cm^3 (diameter: 6.8 cm, height: 3 cm) which is the main geometry used in UIAR when realising the mass analyses.

In the given work the attempt to evaluate an error of determination of the radionuclides content in a single soil sample stipulated by a volumetric heterogeneity contamination of measured sample, to determine its contribution to a common error of evaluation of the median of the density soil contamination on non-gradient with contamination sites has been made.

5.1 Probability distribution of measured activity in a single sample

The method of dividing sample into some parts (further "method of division") is generally applied as the main method reveal fuel particles in soil sample (the presence of volumetric heterogeneity of sample contamination). The method of sequential intermixing of a measured sample (further " method of intermixing ") is also used under γ-spectrometry [81,82].In the quoted works when accounting the probability of detection of fuel particles by in sample the " method of division" and the " method of intermixing " of the measured sub-sample, a logarithmically normal probability distribution is used for approximation of received values of activity.

The conducted theoretical researches on modelling of the ” method of intermixing “ of a measured sample containing fuel particles, have shown, that the distribution of registered activity has obvious right-hand asymmetry [83].It was supposed when modelling, that after every measurement the sample material is emptied out of the measuring vessel, intermixed in conditions preventing its loss and put back into flask for the following measurement. The probability distribution of registered activity was obtained by the theoretical calculations on the basis of Poisson distribution. The kind of theoretical distribution density of probability of measurements results of uniformly polluted sample and the sample containing a "hot" particle, which has the same activity, as uniformly polluted sample is shown. in figure (Fig. 5.1), borrowed from [83], when calculating, the cylinder 1000 cm^3 volume (diameter: 9.1 cm, height: 15.4cm) was taken as a measuring vessel.

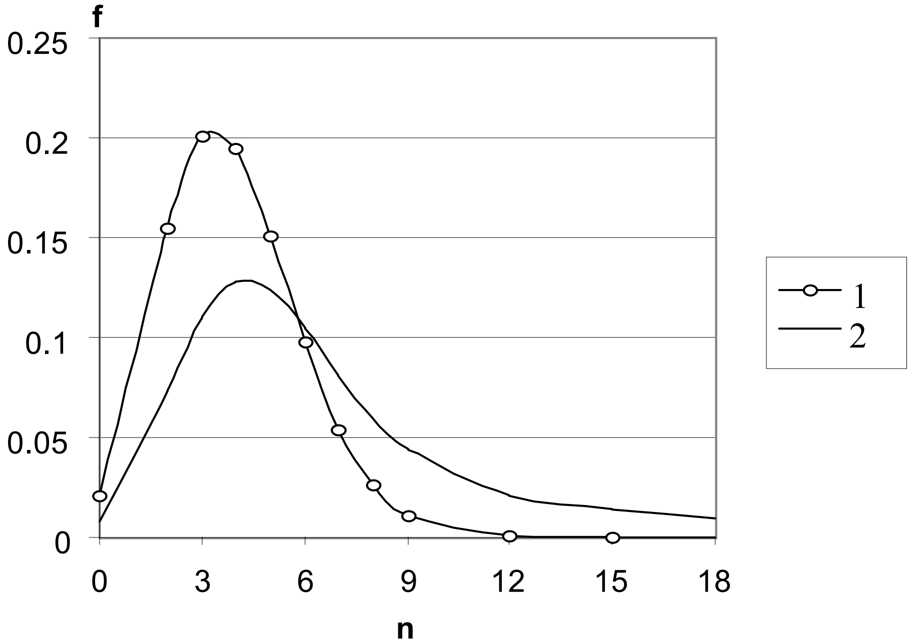


Fig. 5.1 Typical kind of theoretical differential distributions of outcomes of γ-spectrometric measurements when mixing the measured sub-samples: **(1 - is uniformly contaminated measured sub-sample of soil; 2 – the same measured sub-sample, but containing hot particle with the same activity)**.

The registered number of gamma quantum, which is proportional to common or specific activity of the measured sample, is taken as the measurement result in the figure. As it is seen in the indicated figure for different calculated cases, the distributions have obvious right-hand asymmetry and are close to logarithmically normal distribution.

The experimental researches on study of a volumetric heterogeneity of radioactive contamination of soil samples containing fuel particles were conducted in UIAR. The real soil samples selected on the free fuel component of radioactive contamination territories were used in the experiments. These soil samples had specific activity on ^{137}Cs ≈ 50 Bq/kg, ≈ 1700 and ≈ 4900 Bq/kg. After standard sample preparation, one measured sub-sample 1000 cm^3 volume (O_1, O_2 and O_3) was taken from every soil sample and three series of measurements were conducted: the first one was control and two series were with artificially applied particles. Before the second series of the measurement one fuel particle was added in every sub-sample: 152 ±11 Bq ^{137}Cs $\rightarrow O_1$; 1310±39 Bq ^{137}Cs $\rightarrow O_2$; 1060±21 Bq ^{137}Cs $\rightarrow O_3$, before the third series of the measurements one fuel particle was added: 151±9 Bq ^{137}Cs $\rightarrow O_1$; 410±16 Bq ^{137}Cs $\rightarrow O_2$; 538±22 Bq ^{137}Cs $\rightarrow O_3$. Every series of measurements of ^{137}Cs (25 times), both with the artificially applied particles and without them, was conducted a g-spectrometer in two geometries: in the cylindrical measuring flask 1000 cm^3 and in the Marinelle flask 1000 cm^3. The soil was emptied out after every measurement, intermixed in conditions preventing loss, and was put into the flask for the following measurement.

The check on a Kolmogorov’s goodness-of-fit test under 0.05 value level has shown, that the results of the measurement of sub-sample activity of every soil in the Marinelle flask and in the cylindrical flask do not contradict the logarithmically normal law of a probability distribution, both without fuel particles, and with artificially applied fuel particles. The satisfactory approximation by the logarithmically normal law is given in figures (Fig. 5.2–Fig. 5.4). The histograms and appropriate to them theoretical logarithmically normal probability distributions, of specific activity values of soil sub-sample for different variants of experiment, are given in the indicated figures as an example.

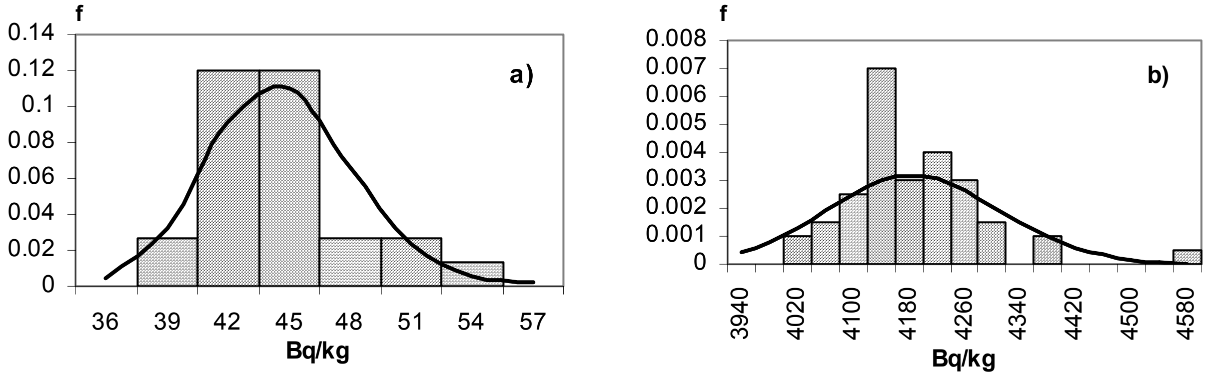


Fig. 5.2. Distribution of measurement outcomes ^{137}Cs in the sub-sample of soil sample (the Marinelle flask, 1000 cm^3) without the fuel particles: **a)** 400 km from Ch NPP along the south trace; **b)** 50 km from Ch NPP along the south-west trace.

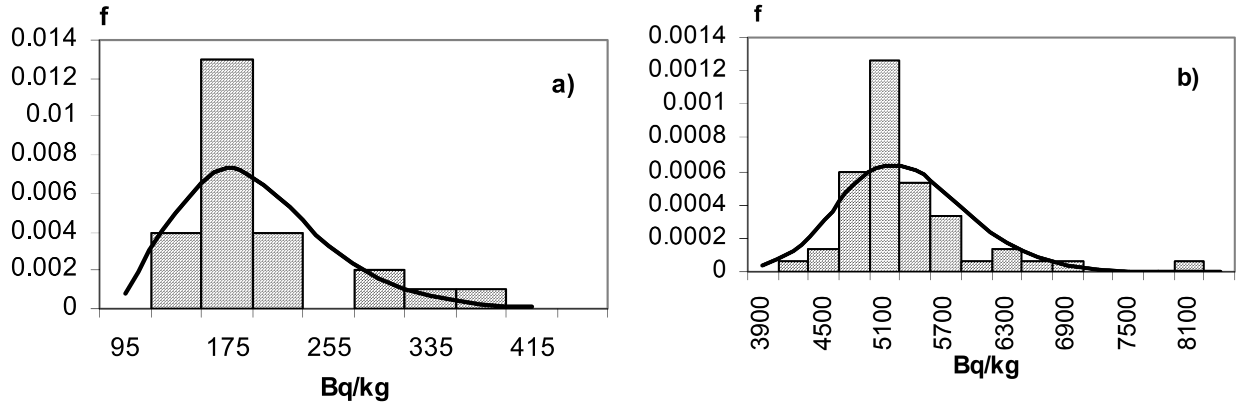


Fig. 5.3. Distribution of measurement outcomes ^{137}Cs in the sub-sample of soil sample (the Marinelle flask, 1000 cm^3) with one fuel particle: **a)** 400 km from ChNPP along the south trace; **b)** 50 km from ChNPP along the south-west trace.

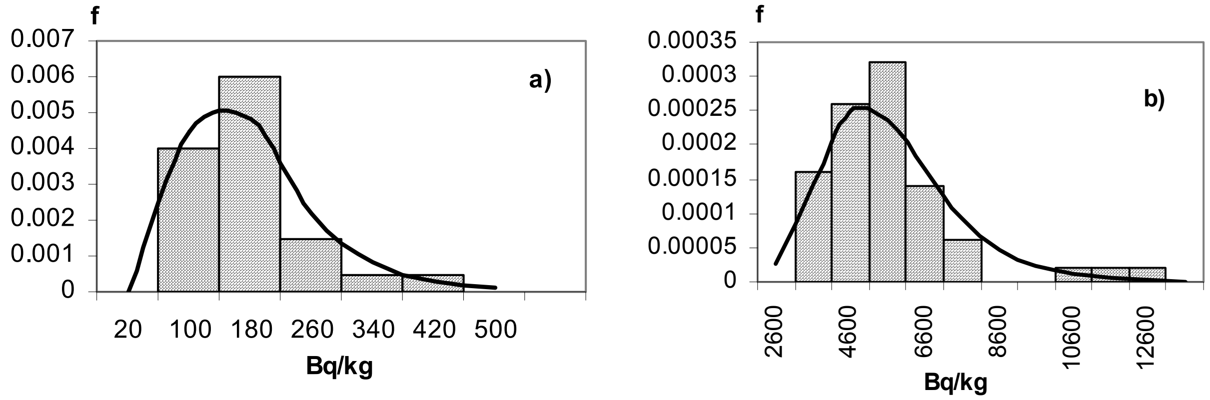


Fig. 5.4. Distribution of measurement outcomes ^{137}Cs in the sub-sample of soil (the cylindrical flask, 1000 cm^3) with two fuel particles: **a)** 400 km from ChNPP along the south trace; **b)** 50 km from ChNPP along the south-west trace.

As the errors of the soil samples measurement of middle (≈ 1700 Bq/kg) and high (≈ 4900 Bq/kg) activity were close and the histograms of distribution of measured activity of single sample for them have appeared the similar, the results for the soil with high activity only are indicated in figures.

The research workers of UIAR selected more than 1300 soil samples from April till October, 1997 when realising the mapping of the 30-km zone of Ch NPP [52]. To measure the ^{137}Cs content in them, four parallel sub-samples were selected from every sample: three sub-samples for the measurement in the cylindrical flask 100 cm^3 volume (diameter: 6.8 cm, height: 3 cm) and one sub-sample for the measurement in the Marinelle flask of 1000 cm^3. The measurements of the sub-sample specific activity were conducted by the gamma - spectrometer with the semiconducting detector (GEM-30185) and passive protection. The results of the measurements were considered as satisfactory, if the relative spread between the values of^{137}Cs specific activity in the parallel sub-samples did not exceed 15 %. Otherwise they were again mixed, carefully homogenised, and again measured under the same scheme. Such procedure is a combination of the “method of division" of sample with the “

method of intermixing” (below "a generalised method"). It coincides with the "method of division" of samples when there is one iteration.

The intermixed and measured more than once samples were selected from this data bank for an evaluation of type of distribution of possible values of specific activity of the samples to study the statistical performances of measured values of specific activity of single sample. The selection of soil samples (n ≈ 200) with increased volumetric heterogeneity of contamination was received As a result. The statistical analysis of the results of the measurements of the soil sample activity, taken from single soil samples, from this selection has shown, that these results of measurements both in the cylindrical flask 100 cm^3, and in the Marinelle flask 1000 cm^3, are also satisfactorily described (do not contradict) by the logarithmically normal laws of the probability distribution. The characteristic results of such analysis for separated points located on different traces of radioactive Chernobyl fallouts, when measuring the ^{137}Cs specific activity in the cylindrical flask 100 cm^3, are shown below in Figure (Fig. 5.6). The continuous curves show the approximation of empirical distributions by the logarithmically normal law of a probability distribution.

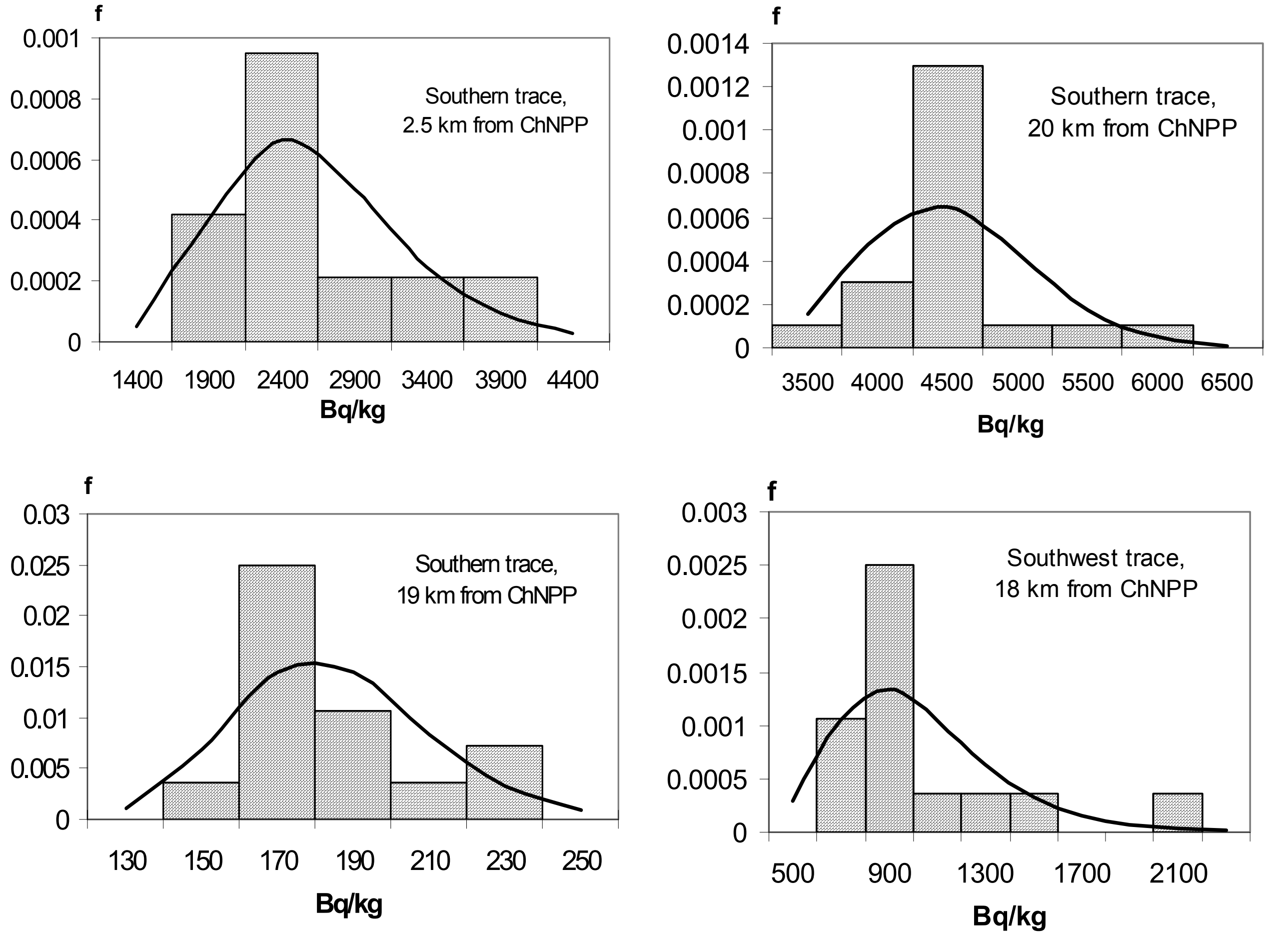


Fig. 5.5– Distributions of measurement outcomes of specific activity ^{137}Cs in single soil samples in some points of the 30-km Zone.

To analyse, what class the probability distributions of measured values of the ^{137}Cs specific activity in single samples for concrete traces of fallouts belong, four sets, describing a western trace, a northern trace, a southern trace and a south-west condensation trace, were formed, out of points of either trace of radioactive fallouts. For every sample ensemble the values of the ^{137}Cs specific activity for the same sample, obtained under measurement of cylindrical 100 cm^3 sub-samples, were normalised on its empirical median and were incorporated in uniform selections, characterising either trace. The statistical analysis of the selections, obtained by such way, has shown, that their probability distribution is also satisfactorily described (does not contradict) by the logarithmically normal law (Fig. 5.6, continuous curves).

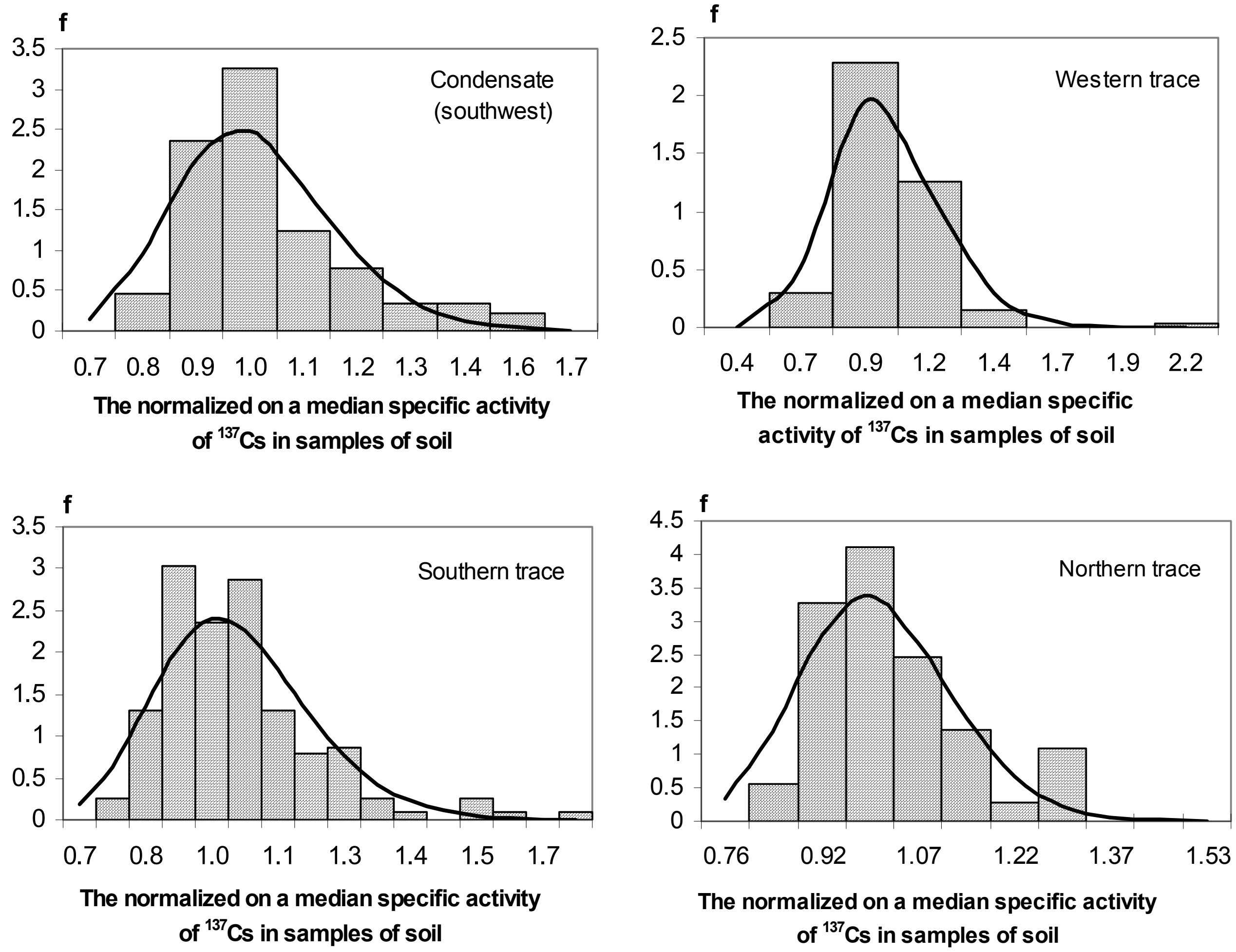


Fig. 5.6– Distribution of the volumes of the specific activity ^{137}Cs normalised on median in the sub-samples of soil samples selected on the different traces of Chernobyl radioactive fallouts.

The probability distribution of measured values of the ^{137}Cs specific activity in single soil samples selected on global fallouts was similarly analysed. Five samples from every site were taken from the soil samples aggregate selected on the **S-P 18** and **S 19** plots (uniformly on width and at random on plot's length). It was obtained 6-7 cylindrical sub-samples 100 cm^3 volume (diameter: 6.8 cm, height: 3 cm) out of all soil of every sample, where the ^{137}Cs activity was measured. The obtained series of the^{137}Cs specific activity values for every sample was normalised on its empirical median. These results were incorporated in uniform selection. Such selection characterises a volumetric heterogeneity of ^{137}Cs contamination of single soil sample selected on global fallouts and prepared for a measurement by a standard technique /i/. The statistical analysis of this selection has shown, that the probability distribution of results of the measurement of ^{137}Cs activity in the indicated geometry is also satisfactorily described (does not contradict) by the logarithmically normal law (Fig. 5.7).

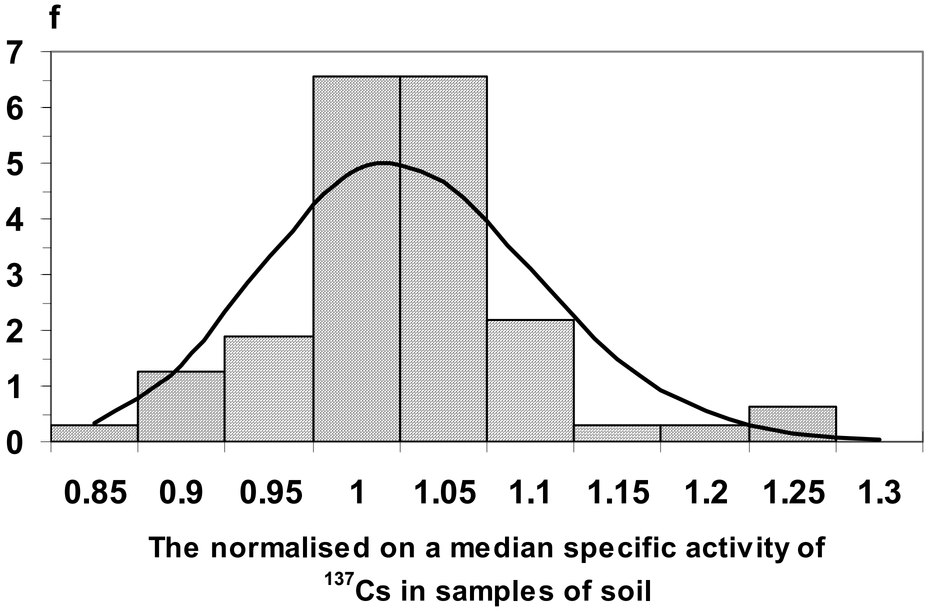


Fig. 5.7– Distribution of the values of specific activity ^{137}Cs and ^{90}Sr normalised on median in sub-samples of soil samples selected on the sites S-P 18 and S 19.

Thus, the results of the conducted analysis show, that the logarithmically normal probability distribution can be taken as a theoretical model of the law of the probability distribution of the results of γ-spectrometric measurements of soil samples with different geometry, which has a volumetric heterogeneity. This distribution will be fair as for the method of sample division into different parts, as well as for the method of sequential intermixing of the measured sample, and for their combination. However, the parameters of the distribution law, in general, will be their own for each case. Such distribution is characteristically for single soil samples selected as on Chernobyl traces of radioactive fallouts, and on background sites samples (on global fallouts).

While in UIAR the researches on kinetics of Chernobyl fuel particles dissolution and radionuclides leaching from them in different soils were conducted [18] 115 soil samples were selected on main (western, northern and southern) traces of radioactive Chernobyl fallouts and at a distance of up to 50 km from Ch NPP. Three subsamples 100 cm^3 volume were taken out of every sample, in which the ^{137}Cs and ^{90}Sr activity was measured (the method of sample division into different parts was applied). The ^{137}Cs content in the sub-samples was determined by γ-spectrometry, and ^{90}Sr content by radiochemistry. To determine the type of distribution of possible values of specific activity in soil sample the obtained results of the measurements of the ^{137}Cs and ^{90}Sr content in sub-samples of every sample were normalised on its median value and were integrated into one selection. The statistical analysis of integrated selection (for ^{137}Cs and ^{90}Sr) has shown, that they also are satisfactorily described by the logarithmically normal law of a probability distribution (**Fig. 5.8**).

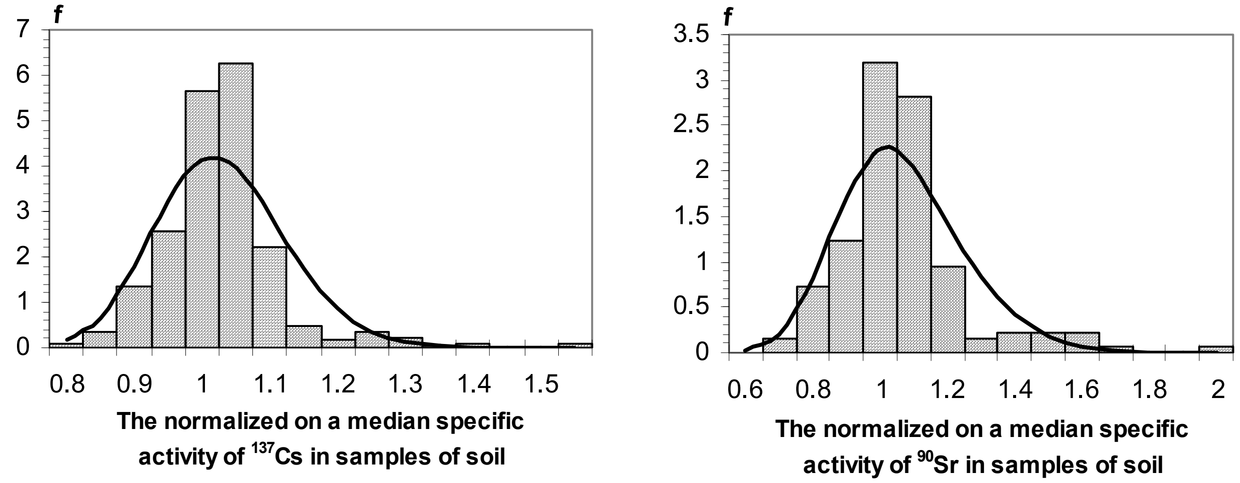


Fig. 5.8– Distribution of the values of specific activity ^{137}Cs and ^{90}Sr normalised on median in sub-samples of soil samples obtained by the results of [18].

As it is seen in the indicated figures, the distribution of the measured specific activity ^{137}Cs and ^{90}Sr of single soil samples selected on different traces of radioactive fallouts within the 30-km zone, and also in places with different contamination levels, are satisfactorily described by the logarithmically normal law.

Thus, for single soil sample prepared for a measurement by a standard technique /i /, the result of the measurement of samples activity taken out of them, characterises the content of either radionuclide in the sample only with the accuracy to some probability distribution, that is mainly stipulated by a heterogeneity of sample contamination, instead of instrument error of measurement. This distribution is measure of obtained information (measure of knowledge) about the specific radionuclide content in the sample and is satisfactorily described by the logarithmically normal law of probability distribution. This statement is fair under γ-spectrometry, as for the "method of division" of samples, as well as for the “ method of intermixing”, and for their combination. However, the parameters of the distribution law for each case can differ. It is fair as for soil samples containing fuel particles, and for samples, polluted with the soluble radionuclide forms (including the global fallouts).

5.2 Distribution parameters of measured activity in the single soil sample

It is impossible to evaluate the exactness (reliability) of measurement results without knowledge of numerical performances of variability of possible values of sample specific activity. As the type of a probability distribution law of specific activity of heterogeneously polluted soil sample, when measuring samples taken out of it, has been determined, the study of statistical performances of specific activity of such sample comes to evaluation and analysis of the parameters of the logarithmically normal distribution law.

The dispersion of the log of specific soil sample activity when using different methods of the measurement is the subject under consideration in the given section. In general the magnitude of this dispersion is determined by fuel particles (their sizes and amount) presence in the sample, as well as by geometry (when γ-spectrometry) and a measured sub-sample volume, instrument error of the measurement.

The results of γ-spectrometric and radiochemical measurements of sub-samples soils, taken out of samples conducted when constructing the map of ^{90}Sr contamination within the 30 km zone [52], the research results of kinetics of dissolution of Chernobyl fuel particles and radionuclides leaching from them in natural conditions [18] and the results obtained on background samples have been used as a statistical material.

General dispersion of a log of evaluation of specific activity of sample containing fuel particles, $s^2_{sum.sa.}$, stipulated by all random factors for given geometry of the measured sub-sample, let's present as

$$s^2_{sum.sa.} = s^2_{n.sa.} + s^2_{inst}, \qquad \textbf{(5.1)}$$

where $s^2_{n.sa.}$ is dispersion stipulated by heterogeneity of polluted single sample;

$s^2_{inst} \cong \left(\frac{\delta_{inst}}{z \cdot 100}\right)^2$ is dispersion stipulated by a relative instrument error of a measurement of specific activity of δ_{inst} sub-sample, taken on ± z·σ level.

When realising measurements in UIAR z=2.

The $s_{n.sa}$ magnitude depends only on non-uniformity of activity distribution in soil sample (mainly on the presence of fuel particles) and consequently it is a parameter of a volumetric heterogeneity of sample contamination with either radionuclide. In Figure (Fig. 5.9) given below, the values of this parameter obtained in described above experiments on artificial contamination of soil samples by fuel particles when γ-spectrometric measuring their activity in the Marinelle flask 1000 cm^3 (the "method of intermixing") are shown. An instrument error of the measurement of sub-sample was 4-10 % without particles and 2-4 % with particles at two sigma level. The conducted experiments prove, that the actual relative error of sample activity determination under single measurement of the sample taken out of it can exceed 7 and more times instrument ratio error which is usually accepted as error of activity determination in sample.

It is necessary to stress that when fuel particles (condensation Chernobyl and global fallouts) are absent in soil sample, the parameter of a volumetric heterogeneity of contamination of such sample with ^{137}Cs when preparing it for the measurement by standard technique [84]does not equal zero. As it is seen in the diagram (Fig. 5.9) when measuring^{137}Cs activity in the Marinelle flask 1000 cm^3 (the "method of intermixing"), this parameter for a stipulated kind of contamination equals

$s_{n.sa.} = 0.07^{+0.033}_{-0.019}$. When measuring ^{137}Cs activity in the cylindrical flask 100 cm^3 (diameter: 6.8 cm, height: 3 cm) (the "method of division") the parameter of a volumetric heterogeneity of contamination of samples selected on **S-P 18** and **S 19** sites (global fallouts) equals $s_{n.sa.} = 0.07 \pm 0.02$.

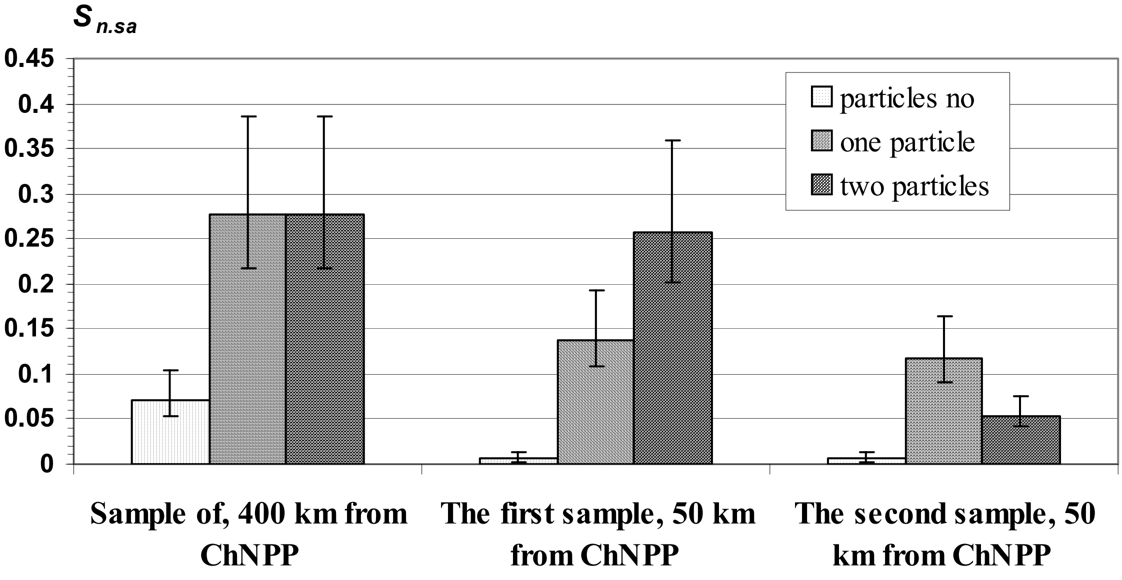


Fig. 5.9 . Value of spatial non-uniformity index of soil sub-samples contamination with ^{137}Cs when artificial insertion of fuel particles in Marinelle flask 1000 cm^3.

The values of parameter $s_{n.sa}$ when measuring ^{137}Cs specific activity for different traces of radioactive fallouts, as according to all results from UIAR data bank (the full selection), and for the selection with increased volumetric heterogeneity of contamination, taken at the moment of measurement in 1997 are shown below in Figure (Fig. 5.10). It is seen in them, that for fuel fallout traces, the $s^{Cs}_{n.sa.}$ magnitude, as a whole is a little bit more, than for the other parts within the 30-km zone. The slight difference of this parameter for different parts of the 30-km zone proves that the probability of “hot” particles presence in the selected soil sample l is essential for any place in the zone. It is also confirmed by the fact that the average value of this parameter for soil samples selected in the settlement of Bober and its neighbourhoods (the condensation trace) when measuring in the cylindrical flask 100 cm^3 equals 0.03, that is 4 times and more as little than for less polluted south-west direction within the 30-km zone ($s^{Cs}_{n.sa.} = 0.133$). The results when selecting samples with increased volumetric heterogeneity are conservative (a little bit overstated) evaluations for appropriate parameters.

In that case if the measurements of the ^{137}Cs content are carried out in the Marinelle flask 1000 cm^3, the $s^{Cs}_{n.sa.}$ value should be a little bit less (the large mass is measured, and it locates more uniformly around the detector). It is confirmed experimentally by conducted measurements. Conservative evaluations of $s^{Cs}_{n.sa.}$ (for sample aggregates, in which the ^{137}Cs content was measured repeatedly in both geometry) for the Marinelle flask 1000 cm^3 on the averages 1.3 times as little, than when measuring in the cylindrical flask 100 cm^3. This ratio can be used to get the first approximation of average evaluations of $s_{n.sa.}^{Cs}$ on full selection for the Marinelle flask 1000 cm^3.

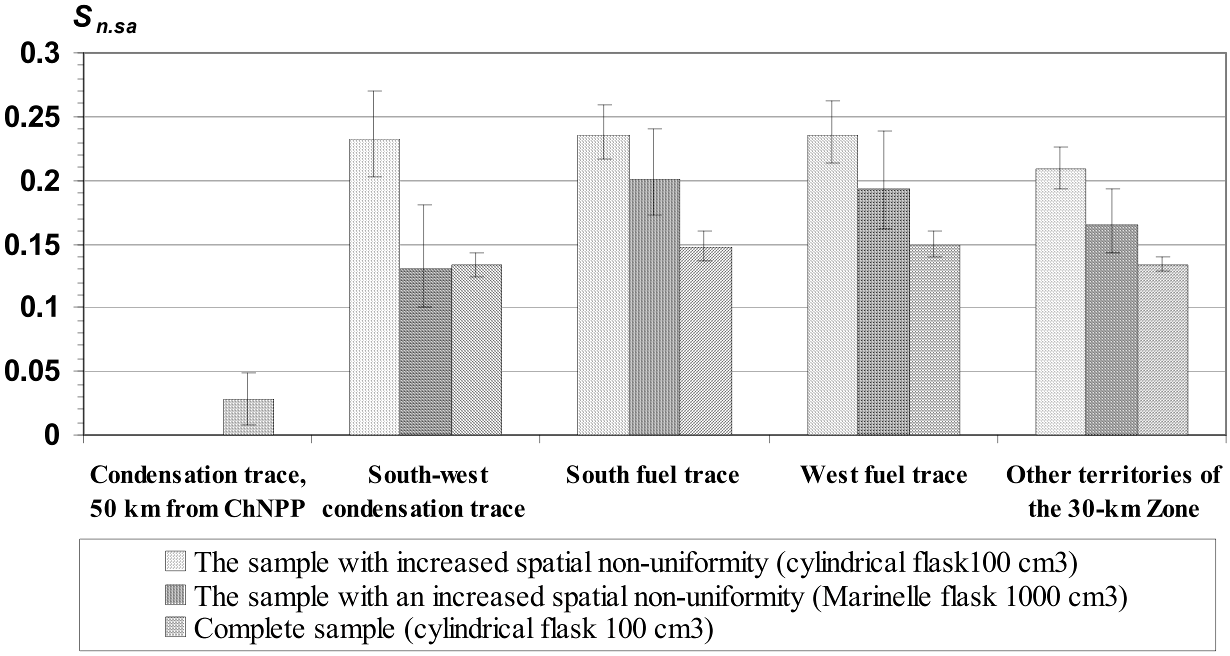


Fig. 5.10. Value of spatial non-uniformity index of soil samples contamination with ^{137}Cs selected in the 30-km Zone in 1997 when γ-spectrometric measurements of their activity in different geometry flasks.

Thus, the average quadratic deviation of the log of specific activity of single sample on ^{137}Cs when measuring the cylindrical sub-sample 100 cm^3 volume, stipulated by its volumetric heterogeneity for the 30 km zone as a whole in **1997** as the first approximation was equal **0.14±0.01** average evaluation), and **0.23±0.03** (conservative evaluation). The appropriate evaluations when measuring the Marinelle flask 1000 cm^3 volume are equal **0.11±0.02** (average evaluation), and **0.17±0.03** (conservative evaluation). Here and below the errors of $s_{n.sa.}$ evaluations are taken at two-sigma level here and below.

The experimental data obtained when studying kinetics of Chernobyl fuel particles dissolution in soils within the 30-km zone in the works [18], allow to evaluate $s_{n.sa.}$ magnitude for ^{137}Cs, ^{90}Sr and their ratio on the results of the measurement of these radionuclides in the same soil samples containing fuel particles. The portion of ^{90}Sr activity got at the moment of measurement on fuel particles *ΔFP* is appreciated in quoted works too. It is shown there, that this magnitude can be tracer of the portion of fuel component of radioactive fallouts in the nearest zone of the catastrophe in Ch NPP (up to 60 km). Therefore we will briefly call the *ΔFP* magnitude as the portion of fuel component in radioactive fallouts. Its influence on $s_{n.sa.}$ value which is the volumetric non-uniformity index of soil sample contamination with the radionuclides (^{137}Cs, ^{90}Sr and TUE) has been analysed below.

As it has already been noted, the study of kinetics of Chernobyl fuel particles dissolution in soils was conducted during 1995-1997. To receive correct evaluations for $s_{n.sa.}$ and to compare them with the evaluations of mentioned above (Fig. 5.10), the portion of the fuel component of radioactive fallouts in every sample was given (recounted) for the middle of 1997 on the basis of obtained dependencies on fuel particles dissolution in natural conditions [18]. Because of small number of measured sub-sample (n=3), the evaluations of $s_{n.sa.}$ for every soil sample were obtained on the base of the range of measured activity of its sub-sample 100 cm^3 volume, according to the well known conclusions of mathematical statistics [30–32]. As the average error of determination of the portion of insoluble fuel particles activity in every sample was ~10 %, the results of the measurements were grouped together (Fig. 5.11).

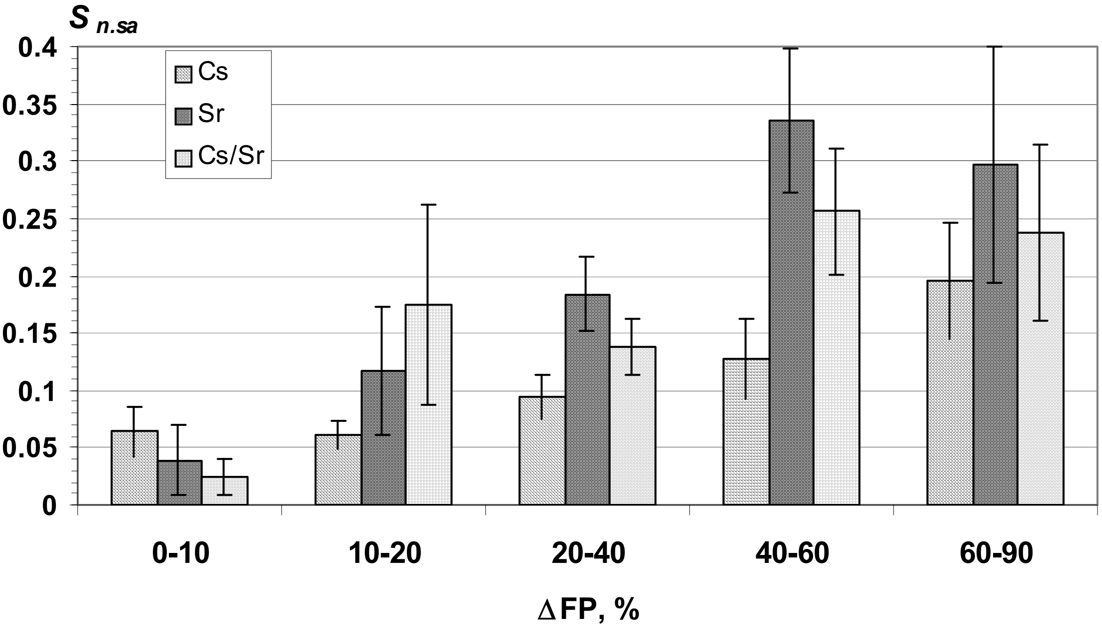


Fig. 5.11. The influence of the fuel component part in radioactive fallouts on spatial non-uniformity of contamination of soil sample when determining specific activity and ratio ^{137}Cs to ^{90}Sr in 1997.

It is seen in the indicated figure, that when the portion a fuel component in ==FP radioactive fallouts increases, the $s_{n.sa.}$ magnitude grows both for ^{137}Cs and ^{90}Sr, and for their ratio, but the greatest growth is watched for ^{90}Sr. The appropriate average values of these parameters are equal: $s_{n.sa.}^{Cs} = 0.11 \pm 0.03$, $s_{n.sa.}^{Sr} = 0.20 \pm 0.05$, $s_{n.sa.}^{Cs/Sr} = 0.17 \pm 0.05$. It is quite natural, if some fuel particles, which mainly determine the sample activity, have got into sample, and then the volumetric heterogeneity will be very high (see the results on artificial entering fuel particles into sample). The $s_{n.sa.}$ magnitude for ^{137}Cs with the 0-10 % portion of the fuel component in radioactive fallouts is indicated in figure (Fig. 5.11) with due regard for corresponding value obtained on a condensation trace in the settlement of

Bober and nearby (см. Fig. 5.10). The difference between the $s_{n.sa.}^{Cs}$ and $s_{n.sa.}^{Sr}$ magnitudes begins with *ΔFP* more than 10 %, with *ΔFP* less than 10 % $s_{n.sa.}^{Sr} \cong s_{n.sa.}^{Cs}$. More smoothed influence of the fuel component portion in radioactive fallings on the $s_{n.sa.}^{Cs}$ magnitude is stipulated by the presence of a big part of ^{137}Cs activity in the structure of a condensation (soluble) component of soil contamination with radionuclides. ^{137}Cs dissolved in soil moisture is mainly fixed by soil, and ^{90}Sr migrates in more deep layers especially in sandy soils Therefore volumetric heterogeneity of soil sample contamination with ^{137}Cs of the higher layer will be as a whole less noticeable, than on ^{90}Sr, till complete dissolution of fuel particles. In time and with the decrease of the fuel component portion the $s_{n.sa.}$ magnitude for ^{137}Cs and ^{90}Sr and their ratio will decrease too and will stay within the limits of 0.05-0.09 (evaluations for the condensation and global fallouts).

The general dispersion of the log of possible values of the specific activity of ^{137}Cs, ^{90}Sr and their ratio in soil sample is calculated according to expression (5.1). when $s_{inst\ Cs/Sr}^{2} = s_{inst\ Cs}^{2} + s_{inst\ Sr}^{2}$. When $\delta_{inst} \leq 10\%$ at ±2σ level the relative error of determination of the indicated magnitudes in sample, is completely determined by a $s_{n.sa.}^{2}$ dispersion, neglect of which can cause serious errors when interpreting the results of the measurement

The fact, that $s_{n.sa.}^{Cs/Sr}$ magnitude is between the $s_{n.sa.}^{Cs}$ and $s_{n.sa.}^{Sr}$ magnitudes is explained only by statistical properties of considered valuations. For difference of correlated random values (when ln (Cs) and ln (Sr) are in the same soil sample), this ratio is fair

$$s_{sum.sa.}^{Cs/Sr} = \sqrt{\left(s_{sum.sa.}^{Cs}\right)^{2} + \left(s_{sum.sa.}^{Sr}\right)^{2} - 2 \cdot r_{ln(Cs),ln(Sr)} \cdot s_{sum.sa.}^{Cs} \cdot s_{sum.sa.}^{Sr}}, \qquad (5.2)$$

where $r_{ln(Cs),ln(Sr)}$ the correlation coefficient equals 0.94 in this case.

As it was already noted, ^{90}Sr is the indicator of the fuel component presence, including TUE in radioactive Chernobyl fallouts within the nearest zone of the catastrophe in Ch NPP. As the leaching of all radionuclides of a fuel matrix (in particular $^{239+240}$Pu) is parallel to ^{90}Sr, then for parameters of a volumetric heterogeneity of soil sample on these radionuclides the similar picture (Fig. 5.11) will be observed, probably with some single-error correction on their migration ability. Therefore at the first approximation it is possible to consider, that the volumetric heterogeneity parameters of soil sample on ^{90}Sr and $^{239+240}$Pu, selected in the mentioned zone, will be similar, $s_{n.sa.}^{Sr} \cong s_{n.sa.}^{Pu}$.

Thus, the volumetric heterogeneity parameter of soil sample contamination $s_{n.sa.}^{Cs}$ (when γ-spectrometry is used) for "a generalised method" and measurement of the cylindrical 100 cm^{3} sample, as a whole for the 30-km zone for 1997 (Fig. 5.10) at the first approximation is evaluated by 0.14±0.01 magnitude (the average evaluation), 0.23±0.03 (the conservative evaluation). The corresponding evaluations when measuring the Marinelle flask 1000 cm^3 volume are equal 0.11±0.02 (the average evaluation), 0.17±0.03 (the conservative evaluation). As in the analysed samples the activity portion, stipulated by fuel particles was not determined, these evaluations correspond to the average content of the fuel component in soil samples within the 30- km zone. For samples, in which the fuel component portion (" a method of division " of the samples, 100 cm^3 sub-sample, Fig. 5.11) was measured, corresponding average values of the a volumetric heterogeneity parameter of soil sample for 1997 are equal: $s_{n.sa.}^{Cs} = 0.11 \pm 0.03$, $s_{n.sa.}^{Sr} = 0.20 \pm 0.05$, $s_{n.sa.}^{Cs/Sr} = 0.17 \pm 0.05$. The $s_{n.sa.}^{Cs}$ value obtained for this selection is close to the evaluation above indicated 0.14±0.01, however it is lower a little. It can be explained by two reasons. The first reason explains that the different statistical methods of the evaluation of dispersion of the ^{137}Cs specific content in the soil sample (the direct method and the method based on a range of measured activity of the sub-samples) were used). The second reason is the higher evaluation has been obtained on the results of the measurement of the samples, selected uniformly within the 30-km zone, and the lower evaluation has been obtained on the results of the measurement of the samples, selected on different soil types in the 30-km zone. The sample selection places were located irregularly within the zone. The proximity of obtained valuations allows to assert: the first - obtained average evaluations of the $s_{n.sa.}^{Sr}$ and $s_{n.sa.}^{Cs/Sr}$ magnitudes (Fig. 5.11) are at the first approximation fair for all territory within the 30-km zone; the second -the $s_{n.sa.}^{Cs}$, $s_{n.sa.}^{Sr}$ and $s_{n.sa.}^{Cs/Sr}$ magnitudes at the first approximation identical, as when using "the method of division ", and " the generalized method " of determination of the soil sample contamination Hereinafter we will use more conservative 0.14±0.01 evaluation for magnitude $s_{n.sa.}^{Cs}$ (100 cm^3 sub-samples). The $s_{n.sa.}^{Cs}$ parameters of the heterogeneity of sample contamination (for both geometries) and $s_{n.sa.}^{Sr}$ at the first approximation (the conservative evaluation) is possible to evaluate by average 0.07±0.02 magnitude for condensation traces outside of the 30-km zone (the fuel component portion is less than 10 %) and global radioactive fallouts.

The experimental data (Fig. 5.11) allow to obtain the modelling dependencies of the selected parameter of the sample volumetric heterogeneity $s_{n.sa.}$ from the fuel component portion in radioactive fallings *ΔFP*. As the $s_{n.sa.}$ magnitude is limited above and below, dependence of a kind in the slope of “S” was taken as a model

$$s_{n.sa.}(\Delta FP) = \frac{c}{1 + a \cdot exp(-b \cdot \Delta FP)} \quad . \qquad (5.3)$$

The obtained values of model parameters when measuring ^{137}Cs and ^{90}Sr in 100 cm^3 sub-samples are listed in table (Table 5.1), and the approximation results are shown in Figure (Fig. 5.12). When ^{137}Cs measuring in the Marinelle flasks 1000 cm^3 corresponding values of the parameters should be reduced 1.3 times as many

Table 5.1 Parameters of the model of $s_{n.sa.}$ dependence from *ΔFP*

$s_{n.sa.}$	***a***	***b***	***c***
for ^{137}Cs	4±0.5	0.03±0.01	0.25±0.05
for ^{90}Sr	4±0.5	0.067±0.028	0.32±0.05
for ^{137}Cs/^{90}Sr	4±0.5	0.081±0.045	0.25±0.05

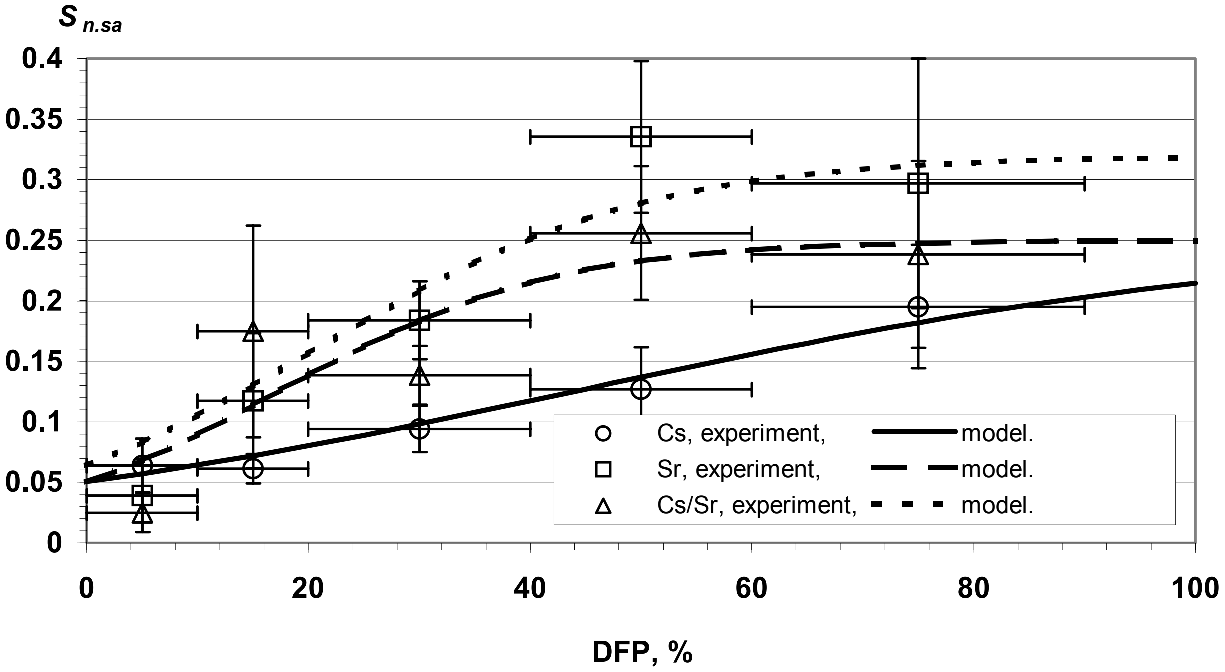


Fig. 5.12. Approximation of dependence of the spatial non-uniformity index of sample on its fuel component part by the selected model.

The fuel component portion in the soil radioactive contamination decreases every year and depends on genesis (a degree of fuel particles oxidation at the moment of catastrophe), soil acidity and the time past after catastrophe. Therefore the values of the parameters of the sample volumetric heterogeneity on different radionuclides will decrease

It is possible to predict the value of the parameter of the volumetric heterogeneity of the single soil sample selected within the 30-km zone of Ch NPP, when measuring ^{137}Cs and ^{90}Sr, the ratio ^{137}Cs/^{90}Sr and $^{239+240}$Pu for any specific time after 1997 when using the obtained previously maps of constant fuel particles transformation k in the soils within the 30-km zone of Ch NPP and dependence on time $\Delta FP=\Delta FP_0 \cdot EXP(-k\ t)$ [**18**], on the basis of the model (5.3) and obtained above $s_{n.sa.}$ values for 1997.

The results of the conducted statistical analysis of numerous measurements of ^{137}Cs and ^{90}Sr activity in soil samples containing radionuclides both in the structure of fuel particles and in the soluble forms, allow to make the following conclusions:

- The considered samples have the volumetric contamination heterogeneity, which is not eliminated in the process of sample preparation (homogenisation);
- On the basis of the experimental data the average evaluations were obtained within the 30 km zone of Ch NPP for 1997 of the parameter of the volumetric heterogeneity of single soil sample when γ-spectrometric measuring of ^{137}Cs in two geometry and when radiochemical measuring ^{90}Sr and $^{239+240}$Pu:
  - **0.14±0.01** (the average evaluation), **0.23±0.03** (the conservative evaluation) when measuring ^{137}Cs in the cylindrical 100 cm^3 samples and **0.11±0.02** (the average evaluation), **0.17±0.03** (the conservative evaluation) when measuring in the Marinelle flask 1000 cm^3;
  - **0.20±0.05** (the average evaluation), when measuring ^{90}Sr and $^{239+240}$Pu in the sub-samples 100 cm^3 volume;
  - **0.17±0.05** (the average evaluation), when measuring the ratio Cs/Sr in the sub-samples 100 cm^3 volume.
- The dependencies of the parameter of the volumetric heterogeneity of soil sample when measuring ^{137}Cs, ^{90}Sr and their ratio from the fuel component portion in the samples of 100 cm^3 volume, which allow to predict a value of the considered parameter for the soil samples within the 30-km zone of Ch NPP when measuring ^{137}Cs and ^{90}Sr ($^{239+240}$Pu) for any specific time after 1997 were obtained;
- The parameter of the contamination heterogeneity of sample $s_{n.sa.}^{Cs}$ (for both geometry) and $s_{n.sa.}^{Sr}$ when measuring the sub-samples of 100 cm^3 volume, at the first approximation (the conservative evaluation) is equal **0.07±0.02** for condensation traces of radioactive fallouts, both within the 30-km zone, and outside of it (the fuel component portion is less than 10 %).

5.3 Optimisation of the number of measured sub-samples when evaluating the median of specific activity of soil sample containing hot particles

If there are fuel particles in soil sample (generally "hot" particles), than, as it shown in the previous section, the standard process of sample preparation to the measurement does not ensure homogeneity of activity distribution in all volume of sample (homogenisation). In that case, when from the sample, prepared for the measurement, we take only one sub-sample to determine specific activity of either radionuclide in it, the results of the measurement of this sub-sample, with due regard for instrument error, can essentially differ from a true value of specific activity of all sample. In this case it is necessary to measure some sub-samples for a required (desirable) exactness of determination of the radionuclide specific activity.

Knowing the total dispersion of possible values of the specific activity of soil sample, with the volumetric heterogeneity (5.1), evaluated in the previous section for different conditions, it is possible to determine minimum necessary number of the measured sub-samples required for an evaluation of the median of radionuclide specific activity in the soil sample with a specific relative error δ when the confidence probability p and relative instrument error of the measurement δ_{inst} taken at ±2σ level.

$$n \geq \left\{ \frac{U_p \cdot s_{n.sa}}{ln(1+\delta)} \right\}^2 + \left\{ \frac{0.005 \cdot U_p \cdot \delta_{inst}}{ln(1+\delta)} \right\}^2 = n_{n.sa} + n_{inst}, \qquad (5.4)$$

where $n_{n.sa}$ is the number of the measured sub-samples, stipulated by the volumetric heterogeneity of sample (fuel particles presence in the sample); n_{inst} is the number of the measured sub-samples, stipulated by an error of the measurement δ_{inst}, taken at ±2σ level (such level, as a rule, is accepted in existing techniques of the measurement).

The conducted above analysis of the volumetric heterogeneity of radioactive contamination of soil samples selected within the 30- km zone of Ch NPP and containing fuel particles and obtained homogenisation process, has shown, that the parameter of this heterogeneity $s_{n.sa.}$ on ^{90}Sr is higher, than on ^{137}Cs. It is stipulated the fact that at the moment of selection and measurements of samples the portion of ^{90}Sr activity, concentrated in fuel particles, was more, than the corresponding portion of ^{137}Cs (the essential part of ^{137}Cs has fallen dropped out as condensation (soluble) component). In connection with fuel particles dissolution, the portion of the fuel component in radioactive soil contamination during every year decreases and the difference between the parameters of the volumetric heterogeneity on different radionuclides decreases. The values of this parameter for soil samples prepared on a standard technique [77] and for use when measuring the sub-samples 100 cm^3 volume for all radionuclides of the Chernobyl fallouts approximates to the $s_{n.sa.}$= 0.07±0.02 magnitude.

In the accordance with evaluations, conducted in the previous section, during 2003 the parameter of the volumetric contamination heterogeneity of the soil samples taken in majority of places within the 30-km zone when measuring the sub-samples in 100 cm3 volume, is evaluated on ^{137}Cs by the magnitude close to $s_{n.sa.}^{Cs}$=0.1, and on ^{90}Sr by the magnitude close to $s_{n.sa.}^{Sr}$=0.15. Let's accept these magnitudes as base values of the parameters of the volumetric contamination heterogeneity of single soil sample on ^{137}Cs and, accordingly, on ^{90}Sr. The value of δ_{inst} =0.1 at ±2σ level is regarded as the base value of a relative instrument error of the measurement of specific activity of the soil sub-sample. The nomogram is given below in Figure (Fig. 5.13) приведена номограмма, которая позволяет быстро определить минимально which allows to determine fast the minimum necessary number of the measured soils sub-samples (100 cm^3 volume) for the evaluation of the median of sample specific activity under base values of the parameters of the volumetric heterogeneity with the specific error and confidence probability p=0.95. On the nomogram the number of the measured sub-samples, calculated on the formula (5.4), is rounded off to the whole value.

It is seen from the indicated nomogram: if now (2003) from soil sample (with fuel particles) only one sub-sample 100 cm^3 volume is taken for measuring (as it is usually done) and instrument error of the measurement of a radionuclide in a sample < 10 % at the $\pm 2\sigma$ level , under of confidence probability p=0.95 the total relative error of the determination of the ^{137}Cs content in the sample can be less than 17 %, and ^{90}Sr will be 24 % (on the average within the 30- km zone of Ch NPP).

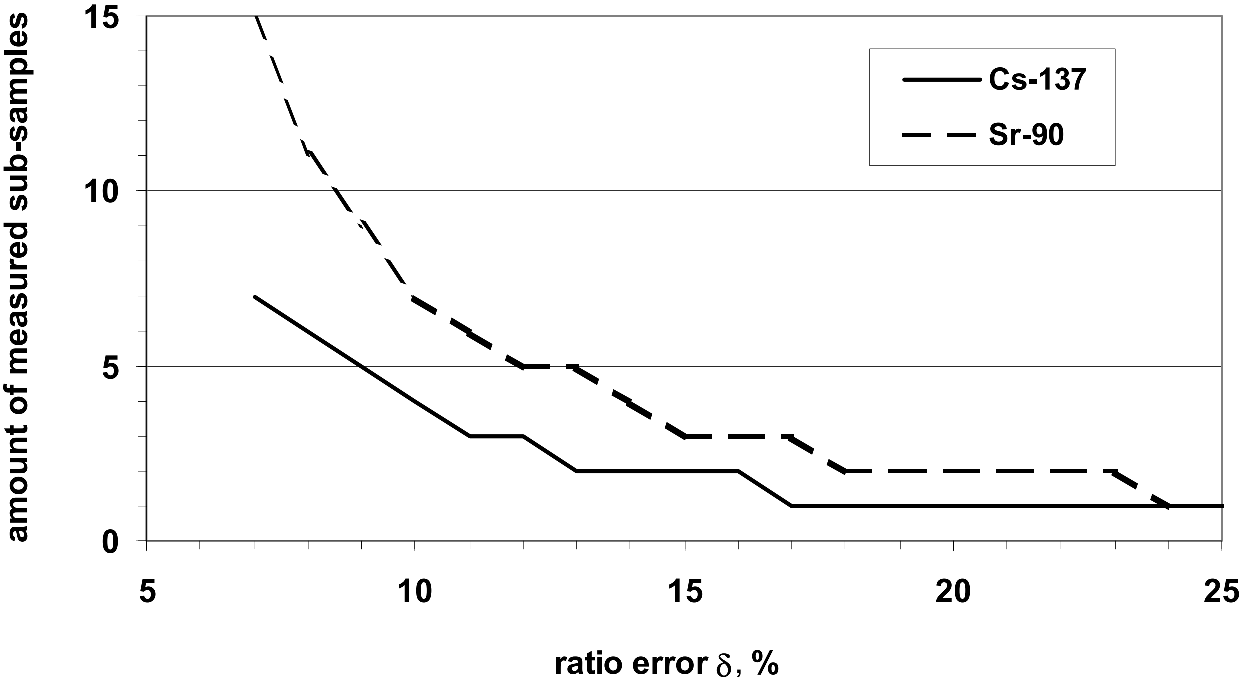


Fig. 5.13. The nomogram to account minimum necessary amount of measured sub-samples with volume of 100 cm^3 to determine median of radionuclide content in soil single sample with prescribed ratio error *δ*.

If the parameter of the volumetric contamination heterogeneity of the soil samples $s_{n.sa.}$ and relative error of the measurement of a radionuclide δ_{inst} are different from the accepted base values, and minimum necessary number of the measured soil sub-samples for the evaluation of the median of sample specific activity under the same confidence probability is determined on the base of recurrent ratio

$$n = n_{bas} \cdot \frac{s_{n.sa.}^2 + s_{inst}^2}{0.0125} \text{ для } ^{137}\text{Cs}, \qquad (5.5)$$

$$n = n_{bas} \cdot \frac{s_{n.sa.}^2 + s_{inst}^2}{0.025} \text{ для } ^{90}\text{Sr}.$$

The solution of a number of problems on planning forthcoming volumes of the measurements (dividing annual production of measuring laboratory for themes and subdivisions, accounts of the costs on forthcoming measurements) demands under stipulated conditions the accuracy of determination of the radionuclide content in the soil sample and absence of information about the portion of the fuel component to evaluate total volume of forthcoming measurements. When the concrete information about the portion of the fuel component in soil samples is absent, it is possible to use average conservative evaluations for $s_{n.sa.}$, for estimated accounts of volumes of the measurement. At the moment (2003) , such evaluation on ^{137}Cs is the $s_{n.sa.}^{Cs}$ magnitude ≈ 0.14, and on ^{90}Sr $s_{n.sa.}^{Sr} \approx 0.2$ for the soil samples selected within the 30- km zone of Ch NPP Having determined on the nomogram (Fig. 5.13) a number of sub-samples n_{bas}^{Cs} and n_{bas}^{Sr} for base parameters, and using the ratio (5.5), we discover the corresponding number of sub-samples of 100 cm^3 volume for the conservative evaluations of the parameter of the volumetric heterogeneity: $n_{con}^{Cs} = 1.8 n_{bas}^{Cs}$ and $n_{con}^{Sr} = 1.7 n_{bas}^{Sr}$. When ^{137}Cs measuring in the Marinelle flasks 1000 cm^3 the obtained values should be reduced in 1.3 times as little, i.e. $n_{con}^{Cs} = 1.1 n_{bas}^{Cs}$.

Example. It is required to calculate the minimum necessary number of the measured sub-sample from the single soil sample to determine the ^{137}Cs and ^{90}Sr content with the relative error δ equal 20 %, under confidence probability p=0.95 in them. It is planned to take the soil samples within the 30- km zone, however definite sampling places (the portion of the fuel component) are not determined. The measurements of the sub-samples are supposed to perform with the ratio error no more than 10 % at ±2σ level.

Using algorithm described above, we get the result: under average conservative evaluations of the parameter of the volumetric contamination heterogeneity of the soil samples ($s_{n.sa.}^{Cs} \approx 0.14$, ^{90}Sr $s_{n.sa.}^{Sr} \approx 0.2$):

- if ^{137}Cs is measured in the cylindrical samples 100 cm^3, then 2 of them should be taken 2;
- if ^{137}Cs is measured in the Marinelle flasks 1000 cm^3, 1 sample is enough;
- if for the ^{90}Sr measurement we take the samples 100 cm^3 each, then 3 of them should be taken.

It is necessary to note, if the sample is selected outside the 30- km zone of Ch NPP on a conditioned trace, then to evaluate the ^{137}Cs content in such sample under δ =20 % with the same demands to the error of the measurement in any of the considered geometry, one sample is enough.

Let's consider one more essential aspect of determination of the ^{137}Cs content in the single soil sample under specific demands to reliability of obtained results.

When realising γ-spectrometric measurements of the ^{137}Cs content in the samples, the number of registered decays (impulses) is proportional to the time of the measurement. The number of decays in a unit of time is subjected to the law of Poisson distribution, for which the dispersion is equal to mathematical expectation. It is easy from here to get the formula where the ratio between the times of one measurement, which ensure errors δ_{inst,t_1} and δ_{inst,t_2} looks like

$$\frac{t_2}{t_1} = \left(\frac{\delta_{inst,t_1}}{\delta_{inst,t_2}} \right)^2 . \qquad (5.6)$$

Using the obtained ratio, we analyse the example mentioned above for the ^{137}Cs measurement in the cylindrical sub-samples 100 cm^3 (the measurement of single soil sample in the Marinelle flasks 1000 cm^3, has no practical significance, as the sample should be very large in this case. As a minimum 2 sub-samples from the sample should be measured at the requests, stipulated in it for reliability of determination of the ^{137}Cs contents in single sample (**δ=20 %, p=0.95**) and relative 10 % at the ±2σ level error of the measurement of ^{137}Cs in the sample. If the measurements of ^{137}Cs in the sub-sample conduct with the error of 30 % at the ±2σ level, as a minimum 3 sub-samples should be measured, but it requires 9 times as little time on the measurements of every sub-sample in the correspondence with the ratio (5.6). From here, the total time which is minimum necessary for the measurement of one sample will be 6 times as much when measuring 2 sub-samples with the 10 % error, than 3 similar sub-samples with the 30 % error.

Thus, from the point of view of the temporary costs (costs of working hours) to achieve the same reliability of determination of the ^{137}Cs content in single sample is much more favourable to measure more sub-samples but with smaller exactness (the time on sample preparation for the measurement remains constant).

5.4 Conclusions on the chapter 5.

In the results of conducted researches It has been established:

- The soil samples selected within the 30-кm zone, because of the presence of the radioactive fallouts fuel component, have essential volumetric heterogeneity which is not eliminated when homogenising;
- The specific content of either radionuclide in such sample (containing the fuel particles), determined in the results of the measurement, is the random value and is satisfactorily described by the logarithmically normal law of the probability distribution;
- The average evaluations of the parameter of the volumetric heterogeneity of single soil sample when γ-spectrometric measurement of ^{137}Cs in two geometries and when radiochemical measurement of ^{90}Sr and $^{239+240}$Pu are obtained within the 30- km zone of Ch NPP for 1997 on the basis of the experimental data:
    - **0.14±0.01** (the average evaluation), **0.23±0.03** (the conservative evaluation) when measuring ^{137}Cs in the cylindrical sub-samples 100 cm^3 and

0.11±0.02 (the average evaluation), **0.17±0.03** (the conservative evaluation) when measuring in the Marinelle flasks 1000 cm^3;

- **0.20±0.05** (the average evaluation), when measuring ^{90}Sr and $^{239+240}$Pu in the sub-samples 100 cm^3 volume;
- **0.17±0.05** (the average evaluation), when measuring of the Cs/Sr ratio in the sub-samples 100 cm^3 volume;

- The parameter of the contamination heterogeneity of the sample $s_{n.sa.}^{Cs}$ (for both geometry) and $s_{n.sa.}^{Sr}$ when measuring the sub-samples 100 cm^3 volume is in the 0.05 -0.09 interval for condensation traces and global radioactive fallouts;
- The dependencies of the parameter of the volumetric heterogeneity of the soil sample when measuring ^{137}Cs, ^{90}Sr and their ratio from the portion in it of the fuel component in sub-samples 100 cm^3 volume, which allow to predict the value of the considered parameter for the soil samples within the 30-km zone of Ch NPP when measuring ^{137}Cs and ^{90}Sr ($^{239+240}$Pu) for any specific time after 1997 were obtained;
- The accounted method of minimum necessary number of the measured subsamples necessary for determination, with a specific relative error, the median of the specific content of ^{137}Cs, ^{90}Sr ($^{239+240}$Pu) and their ratio in the soil samples containing fuel particles was suggested;
- The nomogram was constructed for base significance of the parameter of the volumetric contamination heterogeneity of the soil samples with the radionuclides, which allows to evaluate fast and plan forthcoming volume of necessary measurements guaranteeing specific reliability of the median of the content of ^{137}Cs, ^{90}Sr (^{239,240}Pu) in the soil samples under any specific error of the measurement and the parameter of the volumetric sample heterogeneity;
- It was shown, that it is more favourable to take and measure more subsamples but with a smaller exactness from the single soil sample containing fuel particles, from the point of view of the working hours costs of a measuring laboratory to achieve the same reliability of determination of the ^{137}Cs content in the single sample.

6. Optimisation of soil, plant and milk sampling for radioecological monitoring

Samples of different objects of the environment for radioecological monitoring characterise their real contamination with radionuclide at either reliability. This property of the aggregate of samples (sampling) is called representativeness concerning radioactive contamination of the examined object. The quality and reliability of radioecological monitoring of objects of the environment are determined by representativeness of sampling aggregate selected for inspecting the objects. It is necessary to distinguish two kinds of representativeness: physical and statistical. The physical representativeness of sample is determined by the correct technique of single sample (single) sample during the time and space (for example, it is the account of vertical migration of radionuclides when the soil density contamination is evaluated; it is the account of seasonal dynamics when the milk contamination in the settlements is evaluated). The statistical representativeness of the sampling is determined by the amount of selected samples (sampling size) and by statistical variability of the inspected object contamination with radionuclides.

The reproducibility of obtained outcomes as any scientific research determines reliability and objectivity of radioecological monitoring. If inspected object contamination with radionuclides does not change during the period of time (long-lived radionuclides), the difference between the newly evaluated magnitude of either performance and the previous one should not be statistically significant after realization of the repeated investigation (sampling). Statistically significant difference is the evidence for unrepresentativeness of one of investigations. In this case there is a possibility to correct the committed earlier error. If the radioactive contamination of the investigated object has a brightly expressed dynamic character, it is not possible to eliminate the earlier committed errors completely when investigation is repeated. The investigator should always remember that all the modern analytical methods couldn’t compensate bad representativeness of the selected aggregate of samples.

There is inevitably a problem on statistical reliability of received conclusions and outcomes when we analyse outcomes of radioecological monitoring and other observations. Of course, the larger single sample and the amount of the selected samples, the more correctly the performance or characteristics of the object of the environment, which a researcher is interested in, could be evaluated. But then the investigator comes across the restrictions. Existing measuring toolkit and the techniques of measurement limit volume and weight of a measured sample. Economic, temporary and other restrictions do not allow to sample very large amount of samples. Therefore the investigator should set an acceptable (allowable) error for either performance of investigated object and ensure its achievement on the basis of selected samples.

The outcomes and conclusions obtained in the previous sections allow to proceed with the main purpose of researches – to optimise the amount of selected soil, plants and milk samples when radioecological monitoring and guarantee the specified reliability of monitoring outcomes (monitoring representativeness). The planning (account) of minimum necessary amount of samples for evaluating median of inspected object contamination with radionuclides or for other performances account connected with contamination level of inspected object with specified error is the purpose of the considered problem. This problem can be successfully solved on the basis of methods of interval estimation of unknown parameters [30–32]. The interval evaluations of unknown quantities characterise their accuracy (error) for specified reliability level in difference from dot valuations as one number. They connect an error of determining of unknown quantities with the amount of selected samples (carried out measurements).

U criterion (for a priori known value σ) or t criterion (for a priori unknown values m and σ) [30–32] is used for the normal law of distribution of a random variable with parameters m and σ in order to receive interval valuations of average value of this random variable. In other words when the normal law of distribution of a random variable is the statistics

$$U = \frac{\bar{x} - m}{\sigma} \cdot \sqrt{n} \qquad (6.1)$$

has a normal distribution probability with parameters 0 and 1, and the statistics

$$t = \frac{\bar{x} - m}{s} \cdot \sqrt{n} \qquad (6.2)$$

has a Student distribution probability with n-1 degree of freedom:
where $\bar{x}$ - sample average;
s^2 - selective variance,
n - amount of sampling.

Hence H appears to determine the amount of sampling, which guarantees no exceeding of the specified absolute error $\varepsilon = \bar{x} - m$ when confidence probability p, we get two ratios:

$$n \geq \frac{U^2(p) \cdot s^2}{\varepsilon^2}\ , \qquad (6.3)$$

when a priori known value σ (explicit expression for n) and

$$n \geq \frac{t^2(p, n-1) \cdot s^2}{\varepsilon^2} \quad \text{или} \quad n \geq \frac{t^2(p, n-1) \cdot \bar{w}^2}{\delta^2}\ , \qquad (6.4)$$

when a priori unknown values m and σ (implicit expression for n),

where $\bar{w} = \frac{s}{\bar{x}}$ - sample variation coefficient (as a result on the same sample);

$\delta = \frac{\varepsilon}{\bar{x}}$ - ratio error;

U (p) - fractile of normal distribution of level *p*.

t (p, n-1) - fractile of Student probability distribution of level *p* with n-1 degrees of freedom.

It is necessary to note, that different investigators use both ratio /39–45/ when sampling the deposits of minerals. However in practice the expression (6.3) has broader application and it is even recommended in [27] to account minimum necessary amount of samples for evaluation of average value of environment objects contamination with radionuclides in spite of the fact that the predicted amount of sampling on basis of (6.2) is much lower. The fact can be explained this way: the variance value of investigated random variable (parameter σ) is very seldom known a priori in practice. Therefore for using expression (6.3) preliminary researches to evaluate parameter σ, requiring additional costs are necessary.

While to use the formulas (6.4) for of planning the amount of samples, strictly speaking, is not absolutely correct, since the evaluations of distribution parameters, included in them, $\bar{x}$, s and $\bar{w}$ (they will be evaluated) depend on the planned amount of samples. Strictly speaking, these formulas allow to answer the problems:

- Whether there are enough measurements, which have already been performed to evaluate average value of a general population with specified error and confidence probability?
- What minimum error of evaluation of average value with specified confidence probability ensures the performed amount of measurements?

The practical application of ratio (6.4) for a priori planning amount of sampling, as a rule, is difficulty for investigators, as it is necessary to do either assumptions about the future value $\bar{\sigma}$ (or $\bar{w}$). Using some a priori specific value instead of evaluation $\bar{w}$ breaks a postulate about absence of any information about parameters of the distribution law, on the basis of which the formulas (6.4) have been estimated. Their formal using for a priori known coefficient of variation leads to setting the planned amount of samples too high Therefore the amount of samples planned on the basis of formulas (6.4) for a priori specified coefficient of variation should be considered as conservative (overstated) evaluation.

Depending on the above stated reasons, from our point of view, it is expediently to conduct the planning of samples amount for radioecological monitoring with using U criterion (expression (6.3)) if the distribution law of inspected magnitude is normal. In this connection we go on the preliminary costs concerning evaluation of parameter σ, which are compensated when carrying out radioecological researches by essentially smaller amount of samples and measurements.

Since the beginning of the 90-s we have been conducting researches on investigation and evaluation of parameters of variability of the radionuclides content in different objects of the environment [63,69], and also on appropriate distribution laws. The above mentioned generalisation of outcomes of these researches show that on the sites non-gradient with contamination, the soil density contamination with radionuclides, specific content in different vegetation are subjected to the logarithmically normal law of probability distributions. The logarithmically normal law describes also ^{137}Cs specific content in milk of the cows on private farms with the common pasture at the in definite moment. In the case of the logarithmically normal law of distribution of the radionuclide specific content (C) in samples of researched object, the upper ratio error of evaluation of average value $\overline{C}$, depending on (6.3) is equal

$$\delta = \frac{\overline{C}_{up} - \overline{C}}{\overline{C}} = exp\left(\frac{U(p)\cdot\sigma_{ln}}{\sqrt{n}}\right) - 1. \qquad (6.5)$$

where σ_{ln} - standard deviation of log of the radionuclide specific content in samples;

$\overline{C}_{up}$ - upper evaluation of average value of the radionuclide specific contents in samples.

The minimum necessary amount of samples for evaluating average value of the radionuclide content in samples of researched object equals

$$n \geq \left\{\frac{U(p)\cdot\sigma_{ln}}{ln(1+\delta)}\right\}^2 . \qquad (6.6)$$

As a rule, in practice the magnitude σ_{ln} is evaluated with some error. Instead of magnitude σ_{ln} the definite evaluation S_{ln}, obtained on the basis of some experimental information is undertaken. It leads to an error of calculation n stipulated by inaccuracy S_{ln}. The ratio error of minimum necessary amount of samples (coefficient of variation) w_n, at first approximation, can be evaluated as

$$w_n = 2w_{S_{ln}}, \qquad (6.7)$$

where $w_{S_{ln}}$ – coefficient of variation of an evaluation S_{ln} .

6.1 Minimum necessary amount of samples to evaluate soil density contamination on non-gradient sites

The soil sample can be selected in two ways: by sampling in one place (single sample) and by combining few single samples (combined or composite sample). Sampling square of single sample is either single one non-separable area (in our case it is one "injection") or some areas directly adjacent to each other (in our case they are some adjacent "injections"). The sample square of combined sample it is some areas located on such distance from each other that the radionuclides content in them is mutually independent (single samples are located outside of area of influence on one another, see section 7.). It is naturally the representativeness of combined sample for performance of median contamination of non-gradient site is higher, than single sample. Accordingly, variability of the radionuclides content for depleted samples is less, than for single samples. We can see here perfect analogy to the deposits of mineral sampling in the loose mixed mineral masses [43–45]. In connection with above-stated we would consider planning of minimum necessary amount of soil samples for single samples and depleted samples.

6.1.1 Planning of single samples amount

In section 2.2.1.3 for sampling of single samples when sampling square is ≥ 0.0054 m^2 and the measurement error of activity in soil samples is ≤ 10 % at the level ±2σ we obtain evaluations of standard deviation of a log of soil density contamination with ^{137}Cs on non-gradient site s_n=0.30±0.09. We will to consider conditionally as a base: given value, and also parameters and conditions of soil sampling, measurement error of soil sub-samples for subsequent presentation.

In case of arbitrary but known measurements error (in particular at the level ±2σ), it is possible to present the variance of a log of soil density contamination with ^{137}Cs on non-gradient site as two components

$$s_{so}^2 = s_{so,0}^2 + s_{inst}^2 = (s_{so,0}^2 + 0.0025) + (s_{inst}^2 - 0.0025) \cong s_{bas}^2 + s_{add}^2 \qquad (6.8)$$

where s_{bas}^2 - main (basic) variance of log of the soil density contamination stipulated by the microheterogeneity of the site contamination, by the natural microheterogeneity of soil, volumetric heterogeneity of sample (availability of fuel particles), by the process of samples preparation to measurement and choosing of measured sub-sample, error of standard sub-sample and 10 % (base ±2σ) measurement error;

$s_{so,0}^2$ - variance of log of soil density contamination on site, without consideration the measurement error of soil samples;

$s^2_{add} = s_{inst}^2 - 0.0025$ - additional variance of log of soil density contamination stipulated by part of measurement error which is > 10 %;

$s_{inst}^2 = 0.25\delta_{inst}^2$ - variance of log of soil density contamination stipulated by measurement error (see ratio (2.5)).

With using this presentation of general variance of log of soil density contamination and ratio (6.6) we get the expression to determine minimum necessary amount of samples n, which must be sampling on site with non-gradient contamination site for evaluating median of soil density contamination with any radionuclides with specified ratio error δ and with confidence probability p=0.95

$$n \ge \left(\frac{U_p}{ln(1+\delta)} \right)^2 \cdot \left(s_{bas}^2 + s_{add}^2 \right) = n_{bas} + n_{add} \ . \qquad (6.9)$$

In accordance with outcomes of statistical conclusions stated above (6.6) for basic value s_{bas} the nomogram (**Fig. 6.1**) was constructed which allows to determine fast minimum necessary amount of single samples for evaluating median of soil density contamination with ^{137}Cs on non-gradient site with a specified ratio error δ and for confidence probability p=0.95. Possible errors for n (±σ) are also shown in the nomogram. These errors are stipulated by the scatter of evaluation of standard deviation of log of soil density contamination with ^{137}Cs as a result of influence of random factors for sampling single samples on different landscapes and traces of fallout. For calculations the value of amount of samples was approximated "up" to the nearest whole value, which has led to stepness of nomogram

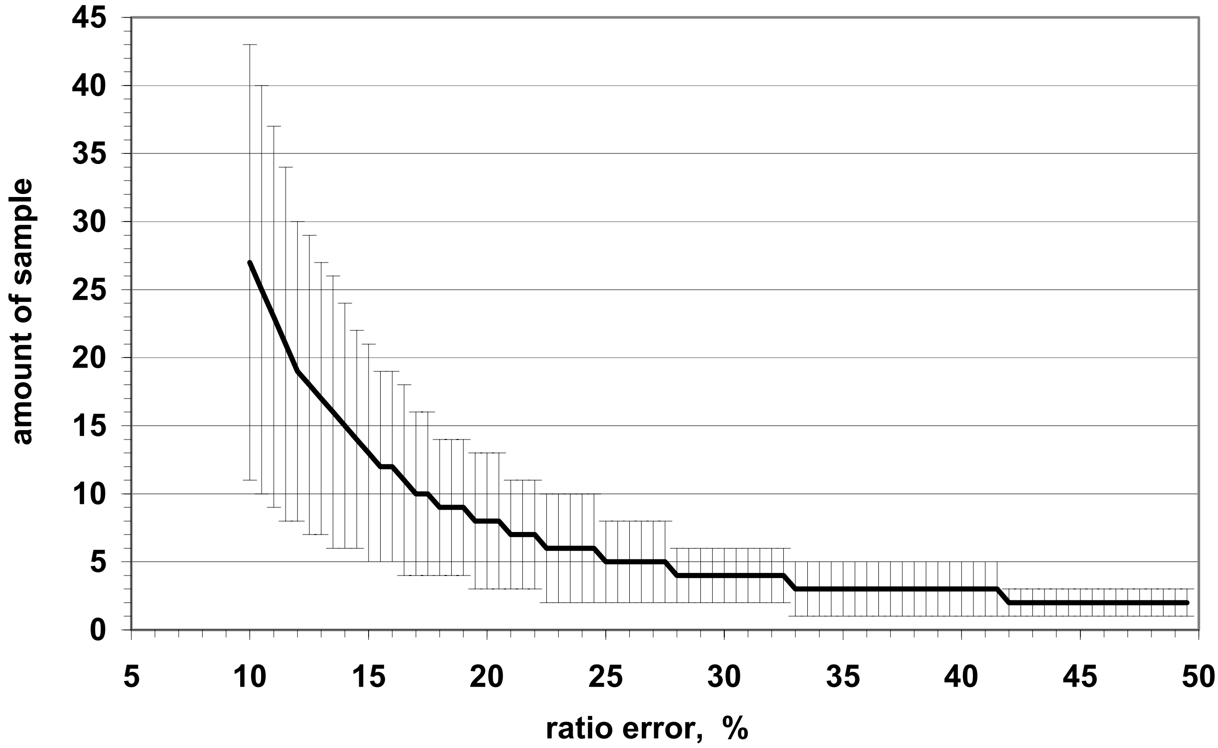


Fig. 6.1.– The nomogram to account minimum necessary amount of soil samples to evaluate the median of soil density contamination with ^{137}Cs on the non-gradient site when sampling and measuring base parameters.

If the ratio measurement error of the ^{137}Cs contents into soil at the level ±2σ exceeds 10 %, it is necessary to sample additional amount of samples to take into account (to compensate) this error for evaluating average value of soil density contamination with specified ratio error. In accordance with expansion **(6.9)** it is calculated by the formula

$$n_{add} = n_{bas} \frac{s_{add}^2}{s_{bas}^2} . \qquad (6.10)$$

Let's consider the following example. Let imagine it is necessary to define minimum amount of soil samples for median of soil density contamination with ^{137}Cs on the site non-gradient site with contamination when error is 30 %. The sampling and preparation samples to measuring will be made according to stipulated above base conditions and parameters. If the measuring ^{137}Cs activity in soil samples will be made with ratio error which does not exceeding 10 % at the level ±2σ, than it is necessary to select not less than 4 independent single samples (at the average) or 6 independent single samples (taking into account error) because of obtained nomogram (**Fig. 6.1**) If measuring ^{137}Cs activity in soil sub-samples will be made with a ratio error which is 30 % at the level ±2σ, because of ratio (6.10) it is necessary to select 1 independent single sample.

Thus, the technique for account of minimum necessary amount of single samples for evaluating median of soil density contamination on site non-gradient with radionuclides contamination with specified ratio error is suggested.

6.1.2 Planning the amount of integrated samples

As mentioned above the composite or combined sample is formed by combining of several single samples located on such distance from each other, when the content of radionuclides in them is mutually independent (single samples are located outside of influence area on one another). Here we will consider only the case, when all point soil samples have the same sampling square and volume, and also when all mass of the sampled soil is included into combined sample (the homogenisation of sample and selecting some part from it is not executed under field conditions).

The variance of log of soil density contamination with ^{137}Cs on sites non-gradient with contamination has two components

$$s_{so}^2 = s_{n.sit.}^2 + s_{n.p.sit.}^2 \qquad (6.11)$$

where $s_{n.p.sit.}^2 = s_{n.sa.}^2 + s_{inst.}^2$ is the dispersion of the log of soil density contamination stipulated by the contamination variability determining at the point of site; $s_{n.sit.}^2$ is the dispersion of the log of soil density contamination stipulated by the microheterogeneity of site contamination (variability among the points of the site).

Let every integrated soil sample form from **m** point soil samples. Since the single samples are independent than the dispersion for combined samples stipulated by microheterogeneity of site contamination (variability among sampling points of combined samples) will be equal $s_{n.sit.com.}^2 = \frac{s_{n.sit.ind.}^2}{m}$ (the dispersion of the average value). Owing to averaging the virtual equalisation of evaluation of sample contamination by radionuclides happens. The dispersion of the point soil sample $s_{n.sit.ind.}^2$ generally depends on square of a sampler and the kind of radioactive fallout. However with sampling squares ≥ 0.005 m^2 this dependence is practically absent. It can be evaluated on the basis of the sampling results and the expression (6.11) $s_{n.sit.ind.}^2 = s_{so.ind.}^2 - s_{n.sa.ind.}^2 - s_{inst.}^2$.

With the $s_{n.sa.com.}^2$ dispersion, stipulated by the volumetric heterogeneity of the combined soil sample, the things are more complicated. In the combined sample the volumetric heterogeneity of the single samples included in its structure are averaged, but it is not clear how this process depends on number and parameters of "hot" particles which in the single samples consists of. Therefore we will consider at the first approximation, that the volumetric heterogeneity of the combined soil sample on the average is the same, as the single sample (the conservative evaluation of the heterogeneity). Then at the first approximation $s^2_{n.sa.com.} \cong s^2_{n.sa.ind.}$.

With due regard for above-stated date the standard deviation of the log of soil density contamination with ^{137}Cs for the sample non-gradient on contamination can be evaluated for combined samples by the magnitude

$$s_{so.com.} \cong \sqrt{\frac{s^2_{n.sit.ind.}}{m} + s^2_{n.sa.ind.} + s^2_{inst.}} \qquad (6.12)$$

The minimum necessary amount of the combined samples for an evaluation of average value of soil contamination with ^{137}Cs for the non-gradient sample is also evaluated under the formula (6.6). Under required ratio error d the minimum necessary amount of the integrated samples can be determined through appropriate volume of single samples on the ratio

$$\frac{n_{ind.}}{n_{com.}} = \left(\frac{s_{so.ind.}}{s_{so.com.}}\right)^2 \qquad (6.13)$$

In the section 2. the evaluations of the dispersion (the standard deviation) of the log of the soil density contamination with ^{137}Cs of the non-gradient on contamination samples s^2_{so} when measuring the cylindrical sample 100 cm^3 are obtained. If the combined sample volume is more than 1000 cm^3, the measurement of the ^{137}Cs activity in it can be made both in the cylindrical samples 100 cm^3, and in the Marinelle flasks 1000 cm^3. When measuring in the Marinelle flask1000 cm^3, the dispersion s^2_{so} will be naturally another one. However, proceeding from the ratio (6.11) and taking into account that the dispersion $s^2_{n.sit.}$ does not depend on the measured sample, under approximately identical errors of the measurement of the dispersion of the log of soil density contamination with ^{137}Cs of the samples non-gradient on contamination when measuring the sample in the Marinelle flask1000 cm^3 can be evaluated by the magnitude

$$s^2_{soMMar} = s^2_{so} - s^2_{n.sa.\ cil.} + s^2_{n.sa.Mar.} \qquad (6.14)$$

For the conservative valuations of magnitudes $s^2_{n.sa.}$ (see section 5.) we have

$$s^2_{soMar.} = s^2_{so} - 0.024 \qquad (6.15)$$

In the table (Table 6.1) the corresponding values of the standard deviations of the log of the soil density contamination with ^{137}Cs of the non-gradient samples for the combined samples (m=5), formed from the single samples on the base of the sampler with a sampling square 0.00107m^2 under the conservative evaluations of $s^2_{n.sa.}$ magnitudes are presented.

Table 6.1– Standard deviations of the log of the soil density contamination with ^{137}Cs of the non-gradient samples when selecting the point and combined samples and the ratio between minimum necessary amount of the samples

Sampling square of the single sample	Condensation traces of fall-out			Fuel traces of fallout		
	$s_{so.ind}$	$s_{so.com.}$	$n_{ind.}/n_{com.}$	$s_{so.ind.}$	$s_{so.com.}$	$n_{ind.}/n_{com}$
Measured sample – cylinder of 100 cm^3 volume						
0.001m^2 (1 injection)	0.34	0.27	1.57	0.51	0.32	2.54
0.002m^2 (2 injections)	0.31	0.26	1.37	0.40	0.29	1.93
0.003m^2 (3 injections)	0.30	0.26	1.30	0.35	0.27	1.65
0.004m^2 (4 injections)	0.29	0.26	1.26	0.33	0.27	1.49
0.005 m^2(5 injections)	0.29	0.26	1.24	0.31	0.26	1.38
Measured sample - the Marinelle flask of 1000 cm^3 volume						
0.004m^2 (4 injections)	0.25	0.21	1.41	0.29	0.22	1.73
0.005m^2 (5 injections)	0.24	0.21	1.38	0.27	0.21	1.58

As it is seen in the table, the sampling square since the square 0.005 m^2 (5 "injections") practically does not influence the standard deviation of the log of soil density contamination with ^{137}Cs of the non-gradient samples when measuring the point and combined samples in both geometries. When using the combined samples (m=5) the minimum necessary amount of selected samples required for maintenance of the specific exactness of evaluation of the average value of the soil density contamination of non-gradient sample, is reduced 1.2-1.4 time when measuring the cylindrical sample 100 cm^3 and 1.4-1.6 time, when measuring the Marinelle flask1000 cm^3.

6.2 The minimum necessary amount of samples to evaluate the ^{137}Cs specific content in plants

In the results of conducted researches and statistical analysis (section 2.3) it was established, that the dispersion of the log of the specific ^{137}Cs content in plants within the limits of the non-gradient samples does not depend on the density contamination, types of fallout and kind of vegetation. Under selected basic ratio error of the measurement of ^{137}Cs in the samples of point vegetative samples ≤ 10 % at the level ±σ, the standard deviation of the log of specific content is at the first approximation evaluated by magnitude s_p=0.37±0.11 for all kinds of plants.

The nomogram (Fig. 6.2) was constructed in accordance with the obtained results and the ratio (6.6). It allows to determine quickly the minimum necessary amount of samples when evaluating the median of the ^{137}Cs specific content in plants within the non-gradient sample with the specific ratio error δ and confidence probability p=0.95 under basic error of the measurement ($\delta_{inst} \leq 10$ % at the level ±σ).

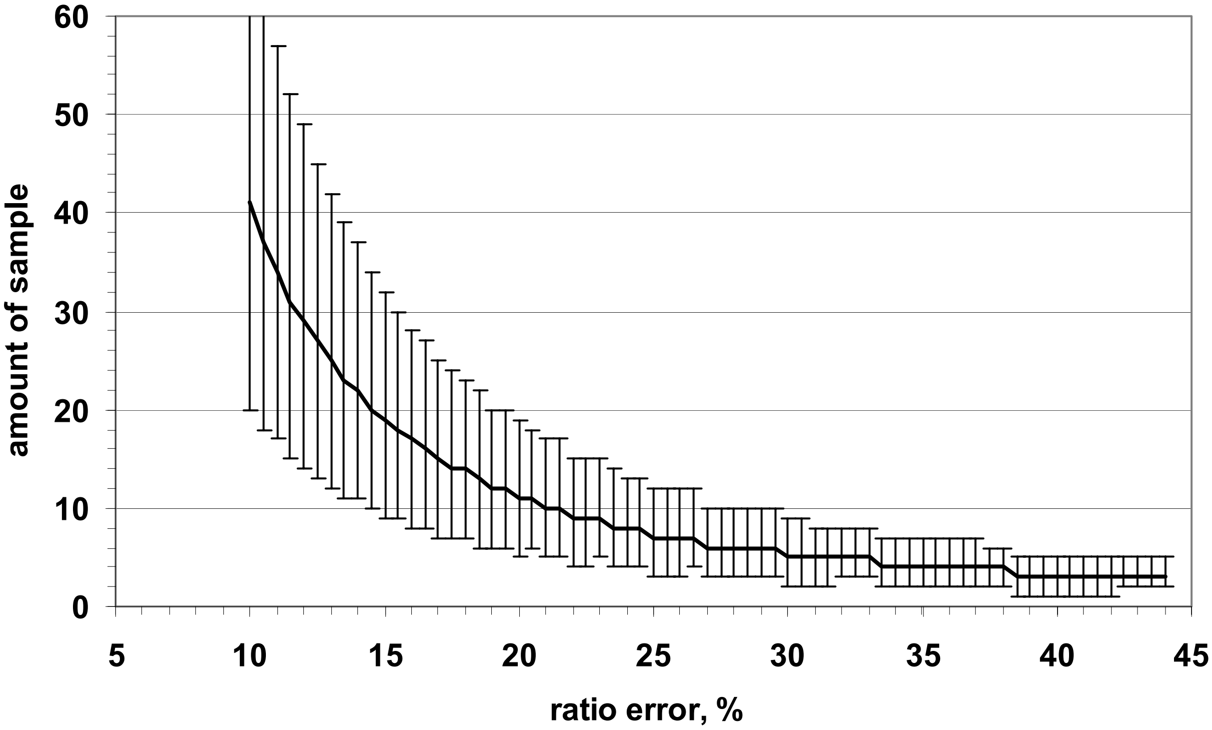


Fig. 6.2.– The nomogram to account minimum necessary amount of plant samples to evaluate the median of ^{137}Cs specific content in the plant on the non-gradient site when base parameters for soil sampling and measuring.

Possible errors for n (interval containing 90 % of all possible values - the significance level 0.05) were shown there. These errors are stipulated by scattering of the evaluation of the standard deviation of the log of the ^{137}Cs content, stipulated for casual factors, which are not taken into account. When calculating the forecasted amount of the samples it was rounded off "up" to the nearest integer value, that has led to steppedness of the nomogram.

If the ratio error of the measurement of ^{137}Cs activity in plants exceeds the basic value (10 % at the ±σ level), it is necessary to select additional amount of samples. It is calculated in accordance with the ratio (6.8) and (6.10).

Let's consider the following example. Let us imagine it is necessary to determine the minimum amount of soil samples for the median of the specific ^{137}Cs contents in plants on some non-gradient sample with the ratio error 25 %. The sampling and preparation of the samples for the measurement will be executed under stipulated above basic conditions and parameters. If the measurement of ^{137}Cs activity in the plant samples will be executed with the ratio error which does not exceed 10 % at the ±σ level, it is necessary to select not less than 7 (on the average) or 12 (with due regard to the errors) independent samples on the base of obtained nomogram (Fig. 6.2). If the measurement of ^{137}Cs activity in soil samples will be executed with the ratio error 20 % at the ±σ level, it is necessary to select 2 single samples on the base of the ratio (6.10) with due regards to (6.8).

Thus, the method of calculation of the minimum necessary amount of the plant samples for evaluation of the median of the ^{137}Cs content in the plant crop within the non-gradient sample with given ratio error is offered.

The conducted comparison has shown, if use the ratio (6.4) formally, to calculate the minimum necessary amount of soil and plant samples then under the same standard deviations (for soils s_{so} = 0.30, for plants s_p =0.37) the amount of soil and plant samples will be approximately twice more under the same requests to sampling, as well as the measurement of ^{137}Cs activity in them and exactness of the estimated magnitude (the median).

6.3 The minimum necessary amount of the samples to evaluate the transfer factors of ^{137}Cs in "soil - plant" chain

The problem of obtaining reliable valuations of the transfer factors in the "soil - plant" chain in definite conditions is one of the most important problems when forecasting the contamination by radionuclides both of agricultural crops and growing wild plants. Its solution is connected with the selection and measurement of great amount of the conjugate soil-plant samples. Limited nature of means provided for radioecological researches, the substantial labour expenditures on sampling and high price of measurements make the problem on optimisation of selected amount of the samples when maintaining necessary exactness of obtained valuations for the transfer factors of radionuclides from soil into plants, urgent.

From the point of view of the ultimate goal of radioecological researches it is possible to select two problems, which are the most frequent in practice:

- Determination of the minimum necessary amount of the conjugate samples for evaluation of the median of the transfer factor in the soil – plant chain with the specific ratio error;
- Determination of the minimum necessary amount of the conjugate samples for evaluation of authentic difference between the transfer factor in the soil – plant chain under different conditions.

In given section it was established, that the dispersion of the log of the transfer factor of ^{137}Cs in plants does not depend on fallout density, as well as on vegetation type and its different parts within the limits of the samples which are homogeneous on agrochemical performances of soil. Under the considered parameters of the sampling of the conjugate soil-plant samples (the sampling square of the plant sample ≤1m^2, soil > 0.005 m^2) and the error of the measurement of the radionuclide content in the soil samples do not exceed 10 % at the ±2σ level, and in the plant samples it does not exceed 10 % at the ±σ level, at the first approximation equal s_k **≅0.49±0.06**.

We will consider given sampling parameters, s_k value and exactness of the measurement of the sub-samples as conventionally basic factors. The given evaluation of the standard deviation of the log of the transfer factor of ^{137}Cs into plants allows to solve the formulated above problems.

6.3.1 The minimum necessary amount of the conjugate samples to evaluate the median of the transfer factor with the specific error

Using the basic value of the standard deviation of the log of the transfer factor and the ratio (6.6), it is easy to determine the minimum necessary amount of the conjugate soil-plant samples n, which needs to be selected for the evaluation of the median of the transfer factor with the specific ratio error δ. Below in Figure (Fig. 6.3) the nomogram is presented which allows to determine fast the minimum necessary amount of the conjugate soil-plant samples which is necessary for the evaluation of the median of the transfer conversion factor of radionuclide with the specific ratio error δ and confidence probability p=0.95 when accepting the basic parameters of sampling and exactness of the measurement of the sub-samples. Possible errors for n ($\pm\sigma$) are given there. These errors are stipulated by scatter of the evaluation of the standard deviation of the log of the transfer factor and are connected with casual, uncontrollable factors. Forecasted amount of samples is approximated to the nearest integer value, that has led to steppedness of the nomogram.

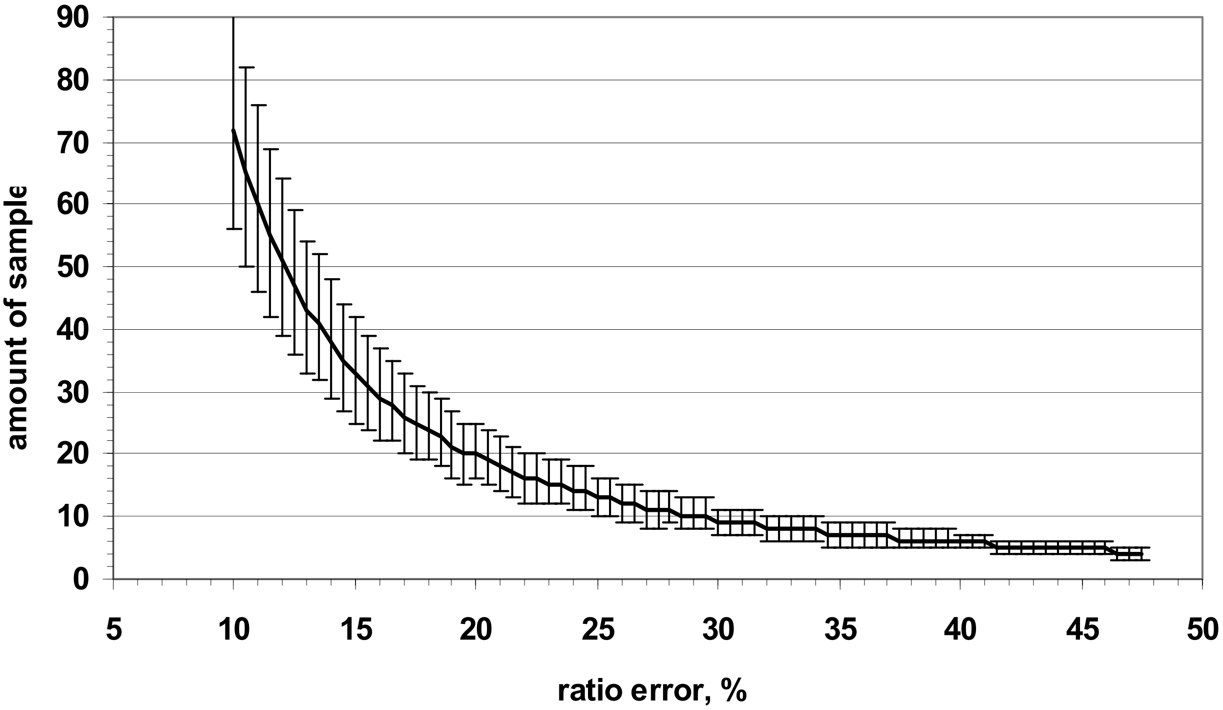


Fig. 6.3– The nomogram to account minimum necessary amount of conjugate samples "soil - plant" to evaluate the median of transfer factor with prescribed ratio error when sampling and measuring base parameters and accuracy if confidential probability is p=0.95.

If the errors of the measurement of the radionuclide content in soil or plants will exceed the basic values, it is necessary to select additional amount of samples to compensate this exceeding when accounting n. In that case it is convenient to use the conservative evaluation of the dispersion of the log of the transfer factor

$$s_k^2 = s_{k,bas}^2 + s_{k,add}^2 \qquad (6.16)$$

where $s_{k,bas}^2$ is the dispersion of the log of the transfer factor under basic values $\delta_{inst,so}$ ($\delta_{inst,p}$);

$s_{k,add}^2 = s_{inst.so}^2 + s_{inst.p}^2 - 0.0125$ is the dispersion of the transfer factor stipulated by an additional error of the measurement of activity in soil and plant samples;

$s_{inst.so}^2$ is the dispersion of soil density contamination stipulated by the error of the measurement of activity in the soil samples $\delta_{inst.so}$;

$s_{inst.p}^2$ is the dispersion of the log of specific contamination of plants stipulated by the error of the measurement of activity in vegetative samples $\delta_{inst.\,p}$;

The formula for additional amount of the conjugate samples looks like

$$n_{add} \geq \left(\frac{U_p}{ln(1+\delta)} \right)^2 \cdot s_{k,add}^2 \ . \qquad (6.17)$$

Let's consider the following example. If it is necessary to determine the minimum amount of conjugate soil-plant samples for the evaluation of the median of the transfer factor of ^{137}Cs with the ratio error 25 % under confidence probability p=0.95.It is necessary to select not less than 13 (on the average) or 16 (with due regard errors) independent conjugate samples on the base of obtained nomogram (Fig. 6.3) if the measurements of the ^{137}Cs activity in samples of soil and vegetation will be conducted with the ratio error which does not exceed the basic value. If the ^{137}Cs activity in the soil and plant samples will be measured with the ratio error 30% at the $\pm 2\sigma$ level, then it is necessary to select 2 conjugate samples in addition. In total it is necessary to select on average not less than 15 (on average) or 18 (with due regard errors) independent conjugate samples. A selection of the conjugate soil-plant samples and the measurement of ^{137}Cs in them should be executed under the stipulated above basic parameters and conditions.

6.3.2 The minimum necessary amount of conjugate samples to confirm the difference between the transfer factors

The agricultural rehabilitation of the territories, polluted with radionuclides, to minimise their influence on radioactive contamination of production and, in the end, the population dose load, demands different counter-measures realisation [62]. Agrotechnical and agrochemical counter-measures (applying higher dozes of mineral fertilisers, liming of soils, alkalisation and realkalisation etc.) reduce radionuclide transition into the crop. Information about reliability of differences between the transfer factors in the soil – plant chain under definite conditions is necessary for evaluation of economic and radiological efficiency of such counter-measures.

It is known [32,67] from the theory of statistical conclusions, if two totalities have normal distributions with parameters N(m_x;σ_x) and N(m_y;σ_y), and the extracts by volumes n_x, n_y are made from them, and corresponding selected sample average $\bar{x}$ *and* $\bar{y}$, are obtained then the statistics

$$\Theta = \frac{\bar{x} - \bar{y}}{\sqrt{\frac{\sigma_x^2}{n_x} + \frac{\sigma_y^2}{n_y}}} \qquad (6.18)$$

also has normal distribution with parameters N(0;1).

The evaluation of the least allowable differences between average values of two random values can be used as an example. In our case the logarithms of the transfer factors have the normal distribution and their common standard deviation s_k is known, therefore the statistics

$$\frac{\overline{ln(Tf1)} - \overline{ln(Tf2)}}{s_k \cdot \sqrt{\frac{1}{n_1} + \frac{1}{n_2}}} = \frac{ln\left(Me_{Tf1} / Me_{Tf2}\right)}{s_k \cdot \sqrt{\frac{1}{n_1} + \frac{1}{n_2}}} \qquad (6.19)$$

has normal distribution with parameters N(0;1). It follows the multiplicity of the ratio of the selective medians of two transfer factors Me_{Tf1} *u* Me_{Tf2} $\left(Me_{Tf1} > Me_{Tf2}\right)$ will be significant with the confidence probability *p*, if the condition is executed

$$ln\left(\frac{Me_{Tf1}}{Me_{Tf2}}\right) \geq U_P \cdot s_k \cdot \sqrt{\frac{1}{n_1} + \frac{1}{n_2}} \quad . \qquad (6.20)$$

The obtained ratio allows to calculate the optimum connected pair (n_1; n_2) - two series of the conjugate soil-plant samples which is necessary to confirm the assigned multiplicity of difference $d = \frac{Me_{Tf1}}{Me_{Tf2}}$. The ratio between this pair and the similar pair (n_{21}; n_{22}), obtained to confirmation the selected multiplicity of difference *d* with that confidence probability but the other value of the standard deviation $s2_k$, is determined by formula

$$\left(\frac{s_k}{s2_k}\right)^2 = \frac{\frac{1}{n2_1} + \frac{1}{n2_2}}{\frac{1}{n_1} + \frac{1}{n_2}} \quad . \qquad (6.21)$$

For quick determination of the connected pair (n_1; n_2) of the minimum necessary amounts of the conjugate soil-plant samples to confirm the assigned multiplicity of differences between the medians of two transfer factors with the basic s_k value and the basic parameters of selection and measurement of the conjugate soil-plant samples for the confidence probability *p*=0.95, the nomograms were constructed as under arbitrary ratio between n_1 and n_2, (thin lines), and under n_1=n_2, (thick line)

(Fig. 6.4). When calculating, the forecasted amount of the samples was approximated to the nearest integer, which has led to steppedness of the nomogram. The possible errors for n_1, which are stipulated by the evaluation error s_k, are not shown on the nomogram with the purpose do not litter it.

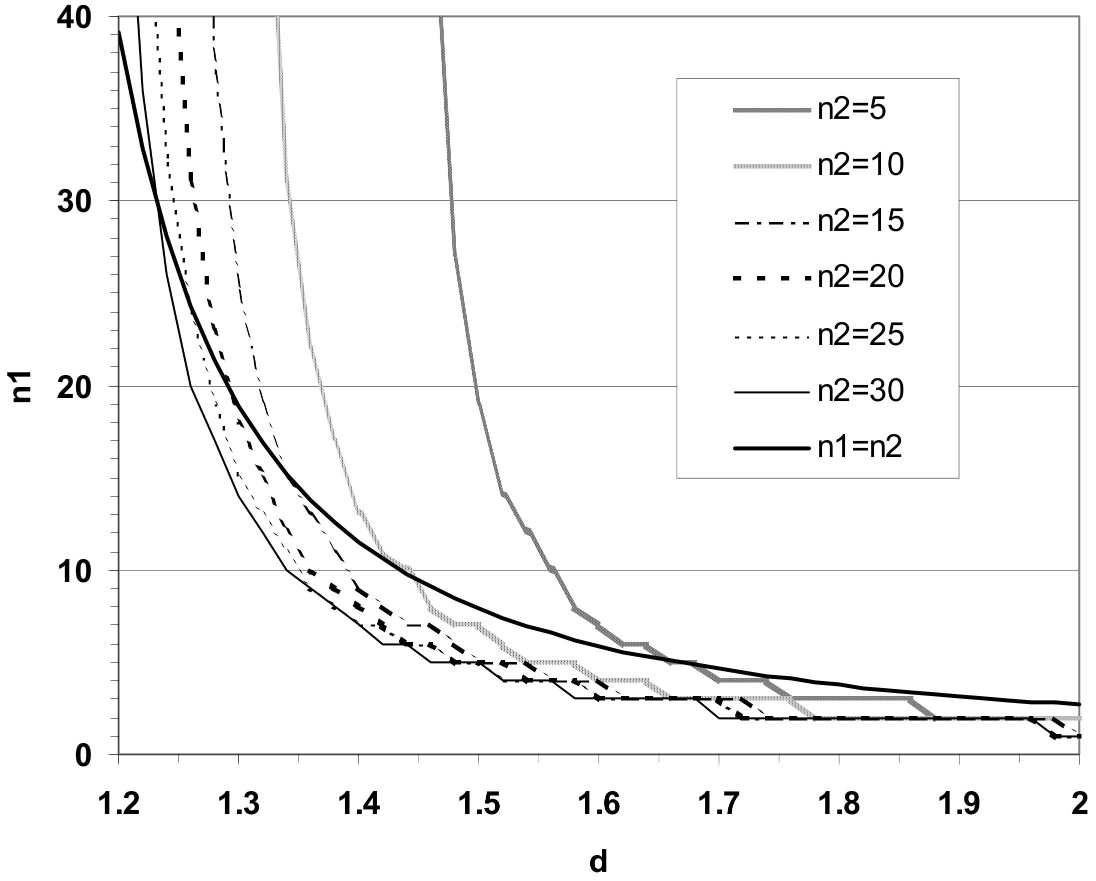


Fig. 6.4 – The nomograms to account minimum necessary amount of conjugate samples "soil – plant" to confirm the prescribed multiplicity of the ratio between transfer factors medians.

Not to clutter up the nomogram, the possible errors for n_1, which are stipulated by the error of the evaluation s_k are not shown in it. These errors are easy to calculate separately. In accordance with the rule of the evaluation of the dispersion of function of random variables [30–32] at the first approximation the ratio error for *n* (δ_n, supposing that $\delta_{n1}=\delta_{n2}$) at the ±σ level is calculated under the formula

$$\delta_n = 2 \cdot \delta_{s_k} \cdot \left(\frac{ln(d)}{U_p \cdot s_k} \right)^2 \cdot \sqrt{\frac{n_1^2 \cdot n_2^2}{n_1^2 + n_2^2}}, \qquad (6.22)$$

where δ_{s_k} is the ratio error s_k equals 0.122 in this case.

If the ratio error of the measurement of radionuclide activity in the soil-plant samples is more than basic values ($s_{inst.so}$>0.05, $s_{inst.p}$>0.1), then the dispersion s_k^2

will change. It is possible to use the conservative evaluation (6.16) to correct $\hat{s}_k^2$ value. For the corrected $\hat{s}_k^2$ value the minimum necessary amount of the conjugate samples of soil – plants $n_1(n_2)$ will change to the magnitude $\Delta n_1(\Delta n_2)$. Let's consider only proportional modifications i.e. $\frac{\Delta n_1}{n_1} = \frac{\Delta n_2}{n_2} = \eta$. With this assumption, when substitute the values $n2_1 = n_1 + \Delta n_1 = (1+\eta)\cdot n_1$ and $n2_2 = n_2 + \Delta n_2 = (1+\eta)\cdot n_2$ in the ratio (6.21) for base value s_k =0.49, after simple transformations we will receive the simple formula for calculation η

$$\eta \cong 4.16 \cdot \hat{s}_k^2 - 1. \qquad (6.23)$$

Thus, with certain basic values n_{1b} and n_{2b} for the new dispersion $\hat{s}_k^2$ the minimum necessary amount of the conjugate soil-plant samples (n_1; n_2) is corrected really simply with due raged for the magnitude η

$$n_1 = n_{1b} \cdot (1+\eta) \text{ and } n_2 = n_{2b} \cdot (1+\eta). \qquad (6.24)$$

These ratios are also possible to use for the determination of the upper valuations of the amount of the conjugate soil-plant samples n_1 and n_2, which is necessary to confirm the specific multiplicity of the ratio between the medians of two transfer factors with due raged for the errors of basic value s_k =0.49±0.06. Maximum (with due regard for the error) the basic values are calculated this way

$$n_{1max} \cong 1.26 n_1 \text{ and } n_{2max} \cong 1.26 n_2\ , \qquad (6.25)$$

where n_1 and n_2 are the values taken from the nomogram (Fig. 6.4).

The upper evaluations of the values n_1 and n_2, calculated with the use of the ratio error δ_n **(6.22)** and the ratio **(6.24)** coincide with the exactness of approximation to the nearest integer

Let's consider the following example. It is necessary to make a comparison of the transfer factors of ^{137}Cs in the soil – plant chain with two different conditions (for example, under application of two different counter-measures) and with the confidence probability p=0.95. The determination of minimum volumes of the conjugate soil-plant samples (identical ones to determine every factor) is necessary to confirm the multiplicity of their ratio not less than 1.5. The sampling and measurement of the conjugate samples is planned to conduct with the basic sampling parameters and exactness of the measurement of the sub-samples. On the base of the nomograms (**Fig. 6.4**) we define, that it is necessary to select on average not less than 8 conjugate soil-plant samples or 11 samples (with due regard for the error). Each soil and plant sample is measured separately. If the measurements of the ^{137}Cs activity in the soil and plant samples are conducted with ratio error 20 % at the ±σ level, it is necessary to select additionally 2 more conjugate samples. If the $d = \frac{Me_{Tf1}}{Me_{Tf2}} \geq 1.5$ ratio is calculated on the base of obtained results, then such difference will be statistically significant with the confidence probability not less than 0.95.

6.4 The minimum necessary amount of the samples to evaluate the median of the specific ^{137}Cs content in cow's milk in the settlements

The cow's milk in the settlement is one of the main objects of radioecological monitoring. In the most cases it is a critical product from the point of view of formation of dose loads for the population within the territories, polluted with radionuclides. In the present part the problem is considered to determine the minimum necessary amount of samples for evaluation, at definite moment of the pasturable and stabling periods, as well as the median of the specific ^{137}Cs content in cow's milk of the settlement with the set ratio error.

In section 4. in result of statistical analysis of observation it was established, that within the limits of the animals' group, which is homogeneous on keeping conditions, the standard deviation of the log of the specific ^{137}Cs content in cow's milk of the settlement equals s_m^{st} = **0.67±0.19** during the stabling period and s_m^{pas} =**0.56±0.19** during the pasturable period. These results are obtained with the instrument error of the measurement, which does not exceed 15 % at the $\pm 2\sigma$ level, and they are average for the settlements located in regions of Ukraine polluted by radionuclides of Chernobyl fallouts. Henceforth we will consider as basic value the value s_m^{pas} = **0.56±0.19** for the pasturable period and exactness of the measurement of radionuclides in milk samples, under which it was obtained.

For the basic value of standard deviation of the log of the specific ^{137}Cs content in cow's milk of the settlement on the base of the ratio (6.6) the nomogram was constructed (**Fig. 6.5**).

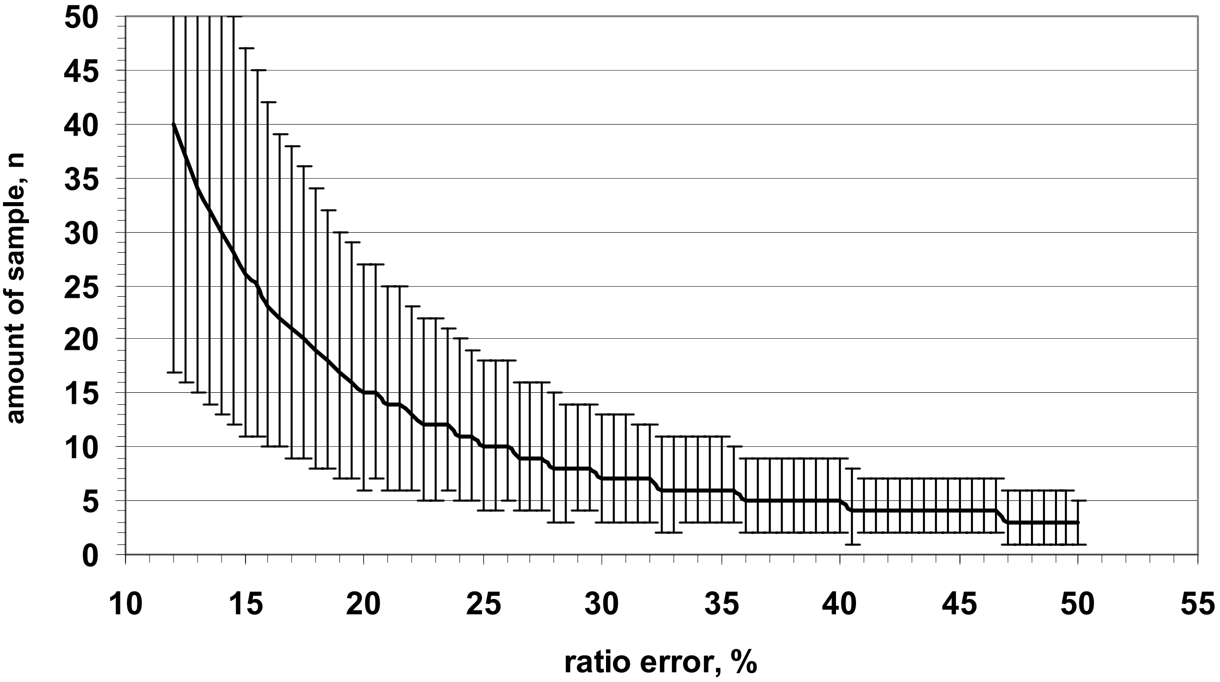


Fig. 6.5. The nomogram to account minimum necessary amount of samples to evaluate the median of milk contamination with prescribed ratio error in the definite moment of grazing period when p=0.95 and δ_{meas} is no more than 15 % at the level ±2σ.

It allows to determine operatively the minimum necessary amount of samples to calculate the median of the specific content of this radionuclide in cow's milk in the definite settlements (for the animals' group –one herd, which is homogeneous on keeping conditions) at any moment of the pasturable period with the specific ratio error δ with confidence probability p=0.95. Possible errors for n, stipulated by casual variability s_M in the settlement are shown there too. When constructing the nomogram the forecasting amount of the samples was approximated to the nearest integer value.

The minimum necessary amount of samples to calculate the median of the specific ^{137}Cs content in cow's milk of the settlement at the definite moment of the stabling period proceeding from the formula (6.6) is determined by the ratio

$$n_{st} = n_{nac}\left(\frac{s_m^{st}}{s_m^{pas}}\right)^2 \qquad (6.26)$$

and it will be **1.42** times as large than in the pasturable period. With such significant magnitude of the standard deviation of the log of specific content of ^{137}Cs and ^{90}Sr in cow's milk in the settlements (s_m^{st} = **0.67±0.19** during the stabling period and s_m^{pas} =**0.56±0.19** during the pasturable period) increase of the instrument error of the measurement of radionuclides in the milk samples up to 20 % at the ±2σ level does not influence much the minimum necessary amount of samples. If the ratio error of the measurement of the specific radionuclide content in the milk samples at the ±2σ level is more than 20 %, then it is necessary to correct the s_m^{pas} magnitude, using approximate evaluation of dispersion

$$(s_m^{pas})^2 \approx 0.56^2 - 0.075^2 + s_{inst}^2 \approx 0.308 + s_{inst}^2 \,. \qquad (6.27)$$

It is possible to use the dependence $s_m(t)$ (Fig**. 4.7**) when planning the sampling in the settlement at some moment of time to determine the s_M value. The minimum necessary amount of samples in this case is determined by direct calculation under the formula(6.6).

It is easy to determine, when using the constructed nomogram, that it has to be selected from **7** (on average) up to **13** samples (to exemene 7-10 animals) during the pasturable period or **10** (on the average) up to **19** samples during the stabling period to evaluate the median of the specific content, for example ^{137}Cs, in the animals' group which is homogeneous on keeping conditions, with the ratio error 30 %, and confidence probability p=0.9.

It is necessary to note that the solution found above concerns to so-called returnable extracts (the amount of cows in the herd is endless). If the amount of cow's herd **N** is small, then the obtained volume *n* must be recounted (evaluated) to the conditions of irrevocable extracts [32].

$$n' = \frac{1}{1/n + 1/N} . \qquad \textbf{(6.28)}$$

If there are some herds (groups) of animals in the settlement, that use different pastures and haymakings, and these pastures and haymakings considerably differ by the level of contamination of vegetation, in particular by ^{137}Cs, then the density function of the specific content of ^{137}Cs in cow's milk of PSF at some moment of time as a whole for the settlement $f(C_m(t))$ will be multimodal. To obtain an objective radioecological picture that reflects the dose loads of all groups of the settlement inhabitants, the minimum necessary amount of samples to calculate the median of the specific ^{137}Cs content in milk, every group of animals according to its number is planned independently (6.28). Total minimum of necessary amount of milk samples, which must be selected in such settlement, equals the sum of samples for all groups of animals.

6.5 Planning milk sampling to evaluate dose loads on the settlement inhabitants

The evaluation of average value of radionuclides penetrating into human organism, defining the doze of internal radionuclide irradiation is one of the practical problems, where the value of annual average ^{137}Cs concentration in milk of the settlement is used. According to [85]it is calculated as

$$D_{Cs}^{vill} = \alpha \cdot \overline{C}_{M}^{Cs} \qquad (6.29)$$

where, $\overline{C}_m^{Cs} = \int_0^1 \overline{C}_m(t)dt$ is the annual average value of ^{137}Cs concentration in milk of the settlement; $\overline{C}_m(t)$ is the average value of ^{137}Cs concentration in milk at moment t during a year ($0 \le t \le 1$); α is a coefficient of proportionality.

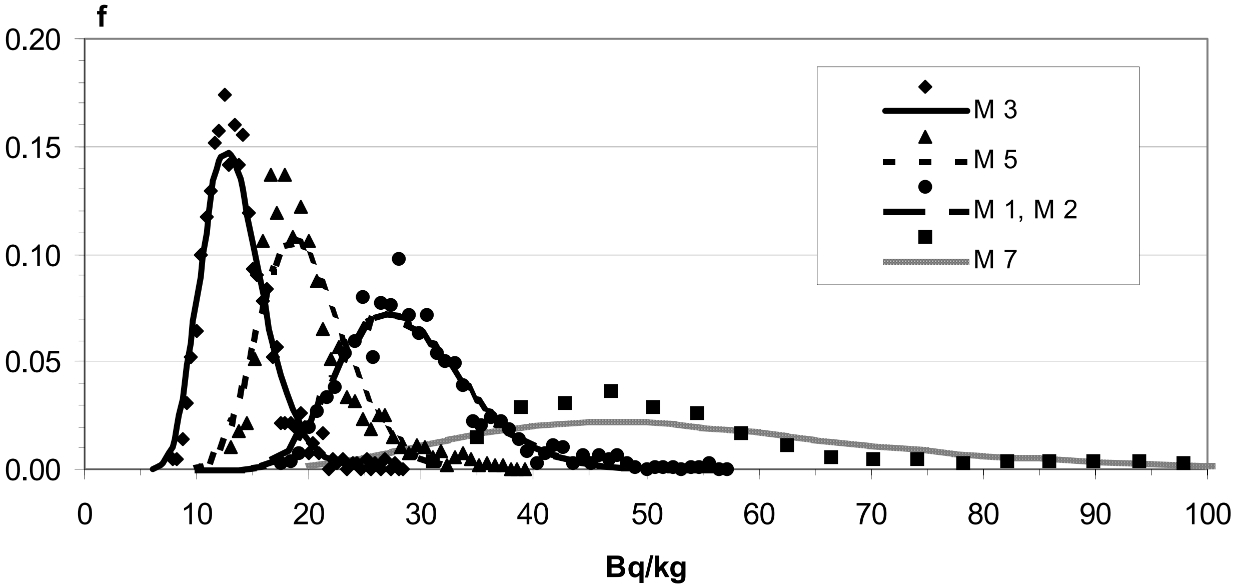
In this case $\overline{C}_m(t) = exp\left(\mu_m(t) + 0.5 \cdot s_m^2(t)\right)$. The ratio error of determination of the D_{Cs}^{vill} magnitude in the definite settlement, which is designated by the letters *δD*, will be determined by a scatter of the annual average value of ^{137}Cs concentration in milk of the given settlement $\overline{C}_m^{Cs}$. For the selected regulations of sampling (the amount of sampling moments and extracts volumes) $\overline{C}_m^{Cs}$ is a random value, as function of casual arguments: a disposition of sampling moments during the stabling and pasturable periods; evaluations of the average value of ^{137}Cs concentration in milk at the sampling moment; variability of the value of ^{137}Cs concentration in milk during the stabling and pasturable periods (Table 4.2).

To determine the minimum necessary amount of samples ensuring the ratio error δD, we will simplify the sampling regulations. Let's consider that the amount of selected samples during the stabling period is constant at any moment of sample selection and it equals N_{st}. The amount of selected samples during the pasturable period is also constant at any moment of selection and accordingly equals N_{pas}. The statistical modelling of the magnitude $\overline{C}_m^{Cs}$ for the different settlements and different regulations of sampling has shown that its probability distribution has the right asymmetry and it is well approximated by the logarithmically normal law. The densities of distribution of the $\overline{C}_m^{Cs}$ magnitudes for the settlements, where the basic radiological samples of UIAR were located, when sampling on one of the possible regulations are given as example below in Figure (Fig. 6.6). The amount of realisations during modelling was accepted equal 5000. If during the stabling or pasturable period the sampling of milk was simulated more than one time, then the corresponding period was divided into equal parts, in each of them the sampling moment (with the exactness up to a month) is selected accidentally (equal probability). The maximum (upper) ratio error of determination of the D_{Cs}^{vill} magnitude for the definite settlement covering 100p of % of a possible dispersion D_{Cs}^{vill} is determined as

$$\delta D(p) = \frac{D_{Cs}^{vill}(p) - D_{Cs}^{vill}}{D_{Cs}^{vill}} = exp(s_m \cdot U_p) - 1. \qquad (6.30)$$

where U_p is the fractile of normal distribution of the level p; s_m is the standard deviation of the log $\overline{C}_m^{Cs}$.

On the base of the results obtained by the method of statistical modelling under the different possible regulations of sampling milk sub-samples, the dependence $N_{pas} = f(\delta D)$ was constructed with p=0.9 for the settlements, where the basic radiological points of UIAR were located. As it was already noted, when creating the network of the basic radiological points of UIAR, the basic farms had been selected to include all possible regional features causing contamination of agricultural production (including milk) by radionuclides within the territory, suffered from the catastrophe on Ch NPP. Thus, at the first approximation it is possible to consider, that the obtained variability of the ratio error of determination of the magnitude D_{Cs}^{vill} is characteristic as a whole for all the territory, which has undergone radioactive contamination after the catastrophe on Ch NPP. Proceeding from this, obtained for the different settlements of the dependences $N_{pas} = f(\delta D)$ were averaged. The averaged dependencies are represented below as nomograms to use conveniently the obtained results in practice purposes.



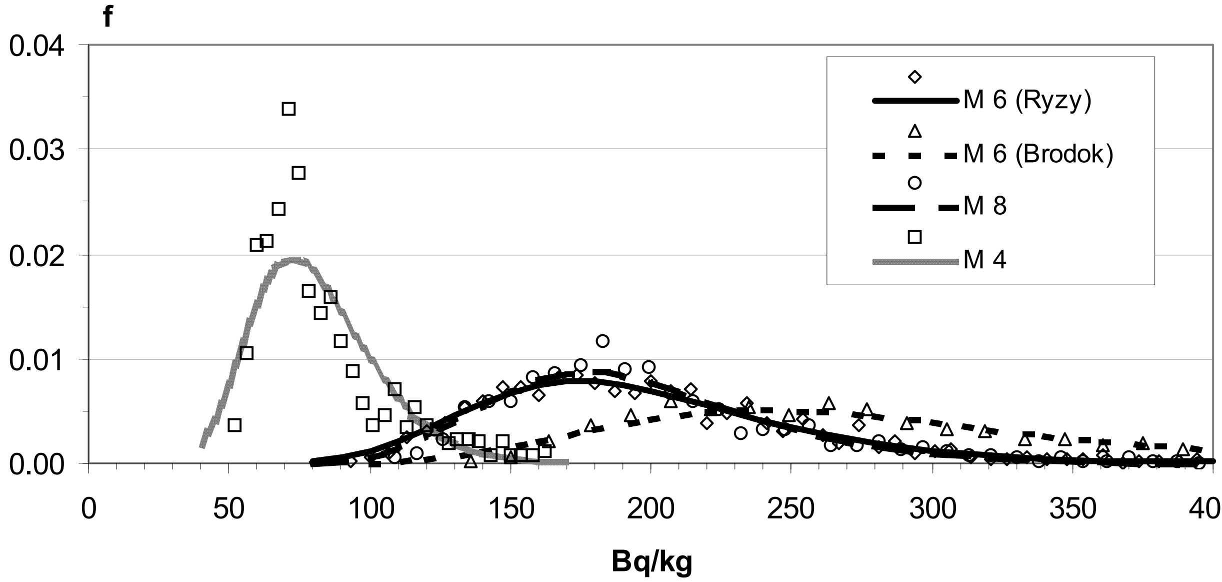


Fig. 6.6.- The distribution of possible values of ^{137}Cs average annual concentration in milk in the PSFs when milk sampling is three times a year: once in a stabling period - 5 samples, and twice - of 10 samples in a grazing period.

In nomograms (**Fig. 6.7**–**Fig. 6.10**) the amount of selected samples during the pasturable period N_{pas} under every examination is shown on ordinate axis, and every curve corresponds to the amount of the selected samples during the stabling period N_{st}. The possible variability of the dependencies $N_{pas} = f(\delta D)$ for the different settlements is shown in nomograms too. On the other hand this variability can be interpreted as a possible disposition of these dependencies during different years for one settlement.

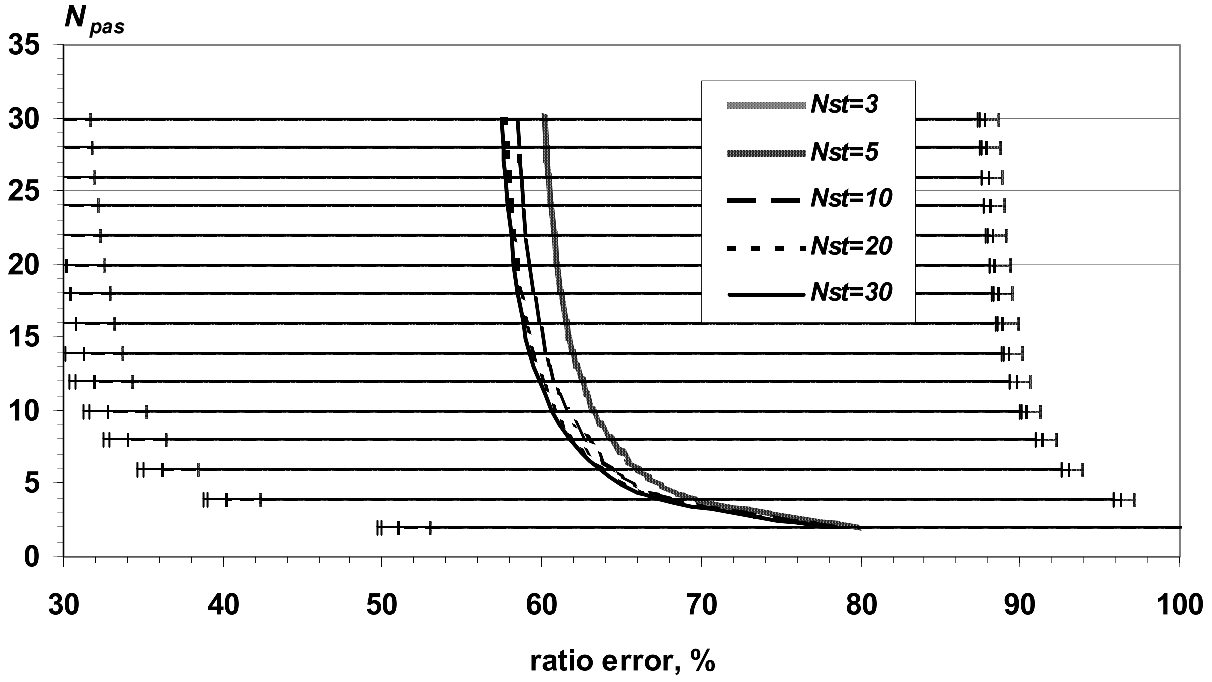


Fig. 6.7. The nomogram to account minimum necessary amount of milk samples, that guarantee the prescribed ratio error to evaluate average annual individual doze of an internal exposure when sampling twice a year (**once in a stabling period and once in a grazing period**)

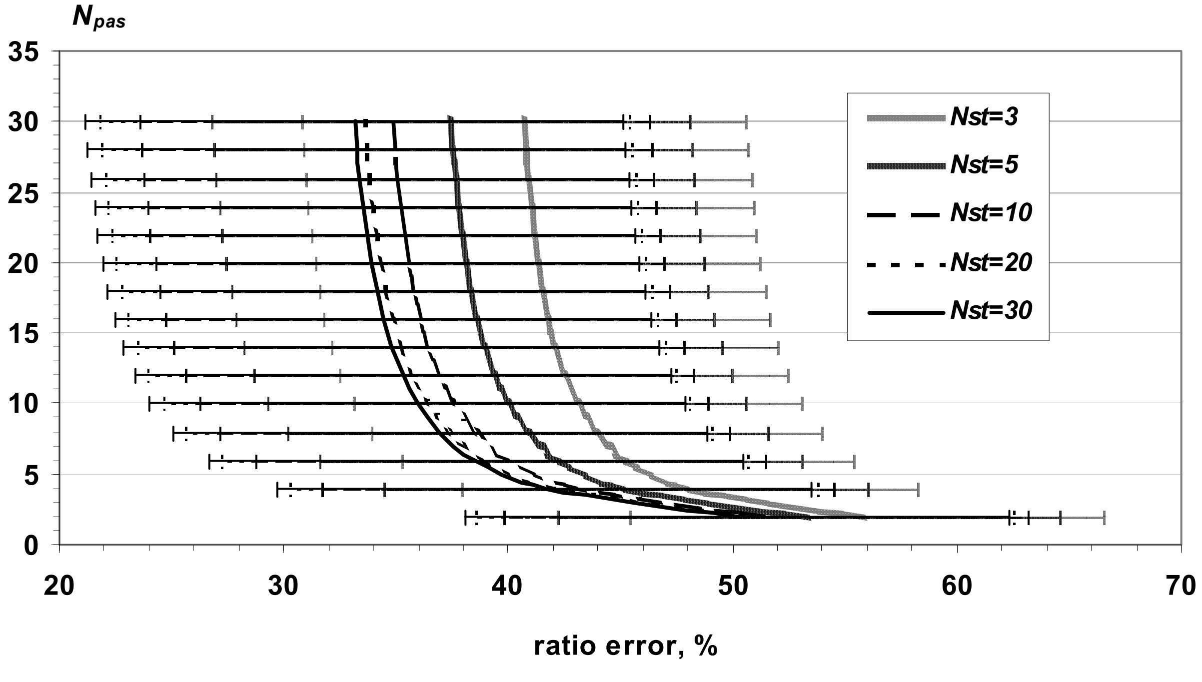


Fig. 6.8. The nomogram to account minimum necessary amount of milk samples, which guarantee the prescribed ratio error, to evaluate average annual individual doze of an internal exposure when sampling three times a year (**once in a stabling period and twice in a grazing period**)..

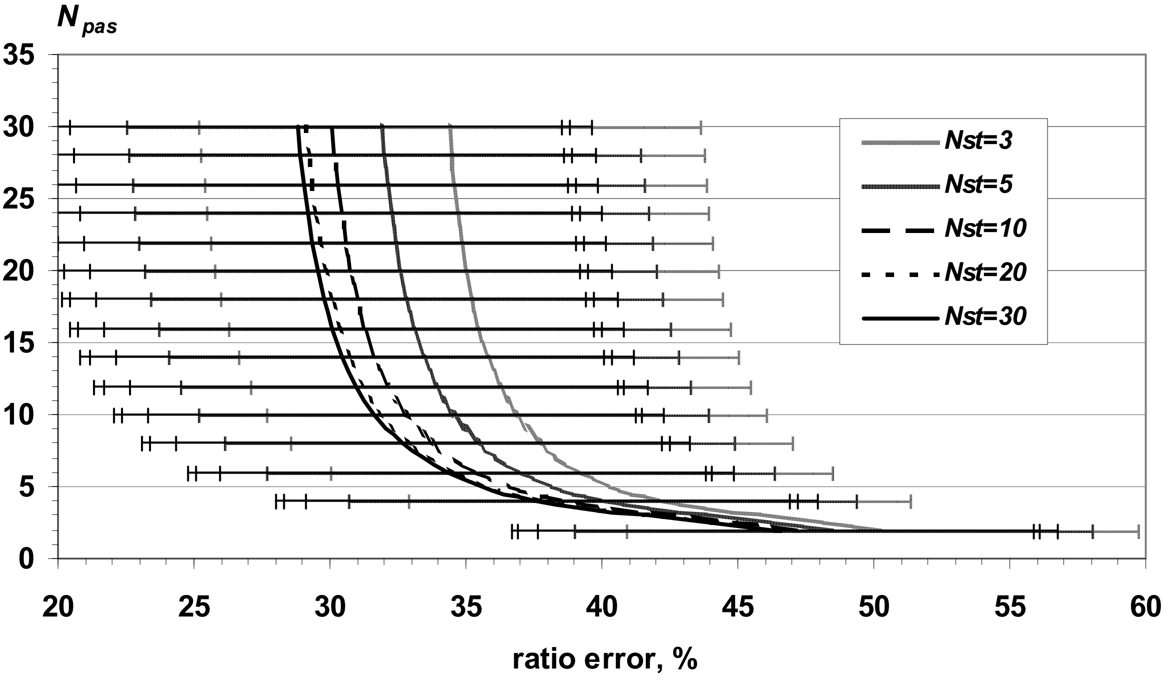


Fig. 6.9. The nomogram to account minimum necessary amount of milk samples, that guarantee the prescribed ratio error to evaluate annual average individual doze of an internal exposure when sampling fourfold a year (**twice in stabling period and twice in grazing period**).

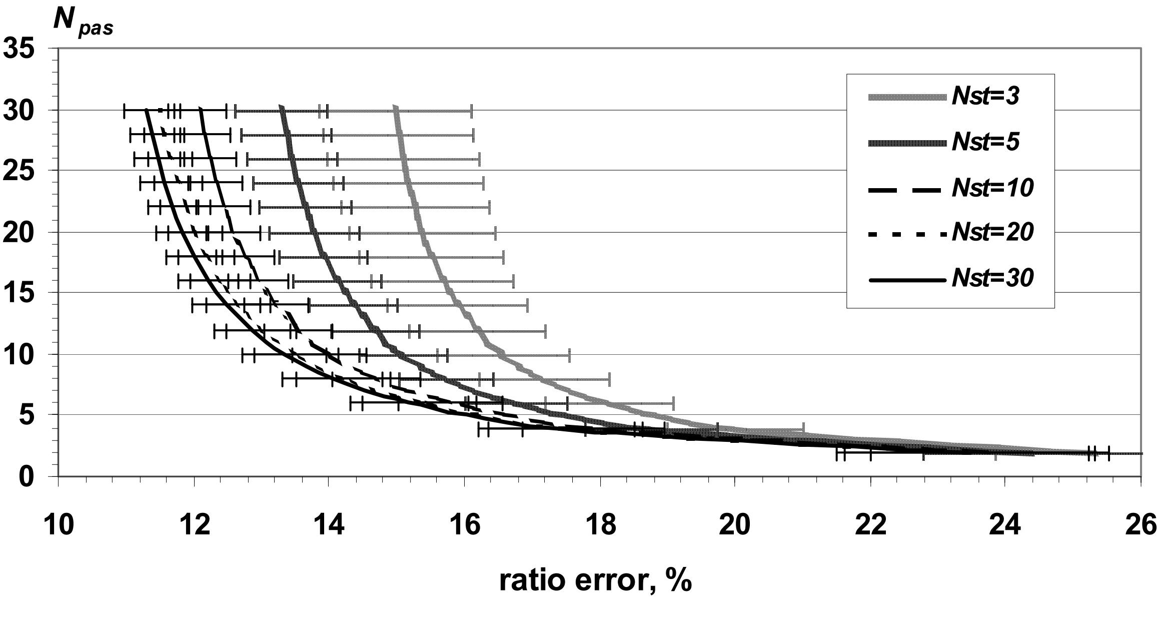


Fig. 6.10. The nomogram to account minimum necessary amount of milk samples, which guarantee the prescribed ratio error, to evaluate average annual individual doze of an internal exposure when sampling during the year (**monthly**).

It is easy to determine the minimum necessary amount of the extracts (to plan the sampling regulations) using the given nomograms for selected sampling frequency For example, to provide the ratio error of the evaluation of the annual average individual dose of internal radioactive irradiation which equals 50 % and defined under the formula (6.29) when sampling milk in any settlement three times per year (1 time during the stabling period and two times during the pasturable period (Fig. 6.8) it is enough to select 5 samples during the stabling period and 10 samples during the pasturable period at every sampling moment. The definite moments (time) of sampling are not regulated. They are casual for every settlement, one time during the stabling period (October - March), two times during the pasturable period (April - June, July September).

6.6 Conclusions on chapter 6.

The main results of the given chapter are the following.

- The methods for calculating the minimum necessary amount of extracts to evaluate with the set ratio error have been developed:
  - the medians of soil and vegetation density contamination with radionuclides on sites non-gradient on contamination;
  - the medians of the transfer factor in the "soil - plant" chain and reliable difference between the transfer factors in the "soil - plant" chain;
  - the medians of the specific contamination of milk with ^{137}Cs with the specific ratio error in the settlement at definite moments of the pasturable and stabling periods.
- The methods for defining the minimum necessary amount of the milk samples extracts during the pasturable and stabling periods have been developed, which guarantee (when calculating the dose of internal radioactive irradiation) the specific ratio error of evaluation the annual average individual dose of the internal radioactive irradiation under different frequency of sampling..
- The corresponding nomograms have been constructed to use conveniently the suggested methods in practice.

7. Geostatistical characteristics of sites non–gradient on contamination

Geostatistical methods have been wide spread and fruitfully used in geology when searching mineral deposits and analyzing selected samples [58,86,87,88,89]. It allows to estimate general characteristics of dispersion and distribution of either minerals attributes. In the given chapter some geostatistical characteristics of soil and plants contamination with ^{137}Cs on the sites in the exclusion zone and adjoining territories have been evaluated by these methods.

Soil density contamination or vegetation contamination with either radionuclide on the experimental site, can be described mathematically by some continuous function from points coordinates on a site $f(x, y)$. The form of this function for some non–gradient sites is shown in section 2. (Fig. 2.6–Fig. 2.7; Fig. 2.16–Fig. 2.20). The main structural function, describing variability of either impurity content in soil (geological rock) in some territory is variogram $\gamma(z)$, which is defined by the following equivalence

$$2\gamma(z)=\frac{1}{n(z)}\sum_{i=1}^{n(z)}\left[f(x_i,y_i)-f(x_i,y_i,z)\right]^2 , \qquad (7.1)$$

where z – distance between sampling points;

$f(x_i,y_i)$ – the impurity content in a point (x_i,y_i) (in sample);

$f(x_i,y_i, z)$ – the impurity content in a point on distance z from a point (x_i,y_i);

$n(z)$ – the pairs of samples number selected on distance z from each other.

As far as soil contamination in a point and on non–gradient site has logarithmically normal law of probability distribution and the subject under consideration is variability of logarithm soil density contamination, then the corresponding natural logarithms (the logarithm of soil density contamination or the logarithm of ^{137}Cs content in plants) instead of function values $f(x_i,y_i)$ have been used to construct the variogram.

Variogram is universal and convenient mean to describe soil contamination variability (or the objects connected with soil, for example, vegetation growing on it) with either impurity (radionuclide) on some area. It is the function of vector argument, i.e. it depends on distance and a direction. In our case variogram shows an average variability of density contamination (of the specific content) on a researched site with either radionuclide and interdependence degree (correlation) of this density (specific content) between the sampling points when changing distance between them in the set direction. In an ideal case it is positively determined growing function starting with zero. If with $z=0$, variogram differs from zero, we can observe display of so-called effect of "nuggets", i.e. sudden modifications of contamination cover short distances. In other words, it’s a degree of distinction between the results of contamination measurements when sampling repeatedly at the same place. It is either consequence of low accuracy of the selected samples measurement, or strong variability of the measured magnitude, or both of them. In the considered case, you can find both of these reasons, especially the second one, caused by availability of fuel particles in soil.

If variogram starting with some *z,* becomes flatter (goes parallel to axes *x*), then the value *z* in the set direction determines a zone (radius, interval) of sample influence. Comparison of influence radiuses for different directions, determines anisotropy of a considered attribute. In our case, at the first approximation, we will consider that there is no anisotropy in soil and plants contamination on non–gradient sites. It means that variability contamination of soil and plants does not depend on a direction on the site.

For comparison and the variogram analysis, received empirical variograms for various sites, as a rule, are approximated by either theoretical model [58,86-89]. For our purposes we will use one of the most spread – spherical model of the following form:

$$\begin{cases} \gamma(z) = G \cdot \frac{1}{2}\left(3 \cdot \left(\frac{z}{R_0}\right) - \left(\frac{z}{R_0}\right)^3\right) + G_0 & z \le R_0 \\ \gamma(z) = G + G_0 & z > R_0 \end{cases} \qquad (7.2)$$

where R_0 – normalising parameter;

$G+G_0$ - a variogram threshold (the magnitude of the general dispersion of contamination on the whole site, received as a result of selection of dot samples);

G_0 - effect of “nuggets” (the part of the general dispersion, caused by effect of "nuggets").

The choice of this model was caused because of following: the parameter for it R_0, coincides with the radius of sample R_{sa} influence. The location of sample radius influence in this case is reduced to parameter R_0 estimation on empirical variogram values. Parameter G_0 is possible to consider identified, it is nothing else than a dispersion logarithm of the density contamination in a point (a general logarithm dispersion of the specific sample $s^2_{sum.sa.}$ activity**)**.

7.1 Minimum necessary distance between sampling points of sites non–gradient on contamination

When doing radio ecological research of either site, selected dot samples of soil and plants should be representative and on sites non–gradient on contamination, statistically independent, especially when forming incorporated samples. So, when the contamination gradient is absent as a whole on a site, the radionuclide content in soil dot sample should not correlate with the same radionuclide content in the other similar samples. It says that the distance between sampling points of soil dot samples selection, providing statistical independence of samples, should exceed some quite certain magnitude, probably the one, which depends on either features of the site. The zone of sample influence on such site ends at distance, starting with which variogram becomes flat, and the dependence between samples disappears outside of this zone. These reasons we will take as a principle of practical definition of radius sample influence.

Variogram is connected with correlation function [58] by ratio $\gamma(z)=s^2\cdot(1-r(z))$, where s^2 – dispersion of logarithm of soil density contamination (specific plants contamination) with ^{137}Cs on the site. Further we will use this ratio for errors estimation of empirical variogram values, and the other known statistical criteria and conclusions [30–32].

7.1.1 Radius of soil samples influence

To get more correct estimations of sample radius influence on parts of experimental non–gradient sites, located on various traces of radioactive Chernobyl fallout, additional ground samples (25-100 samples) have been selected. The relative coordinates of selection places were connected to the basic sampling grid. When sampling by a cylindrical sampler 3.7 cm in a diameter. a step was equal 5 cm. (on fallow lands) and 10 cm. (on arable lands), when ring sampling 13.2 cm in a diameter – 0.2 m (on fallow land). All measurements of ^{137}Cs in soil samples were carried out in a cylindrical vessel 100 cm^3 in volume with error measurements of radionuclide content ≤ 10 % at a level ±2σ. It allowed to construct empirical variogram of logarithm of soil density contamination with ^{137}Cs (Fig. 7.1), using model (7.2) to estimate radius of the sample influence (**Table 7.1**). The value of parameter G_0 was considered identified. At the moment of sampling (1999) it was accepted equal to the average size of experimental sites magnitude 0.02 ($G_0=s^2_{sum.sa.}\approx 0.14^2+0.05^2$ for more information see section 5).

In the result of the realised statistical analysis significant dependence of radius influence, when sampling by a cylindrical sampler 3.7 cm in a diameter, on the type of radioactive fallout, landscape features and cultivation of the land, has not been revealed. There are no significant distinctions between radiuses of samples influence, ring sampling (it was possible to estimate only the top limit of radius influence for them) and a sampler..

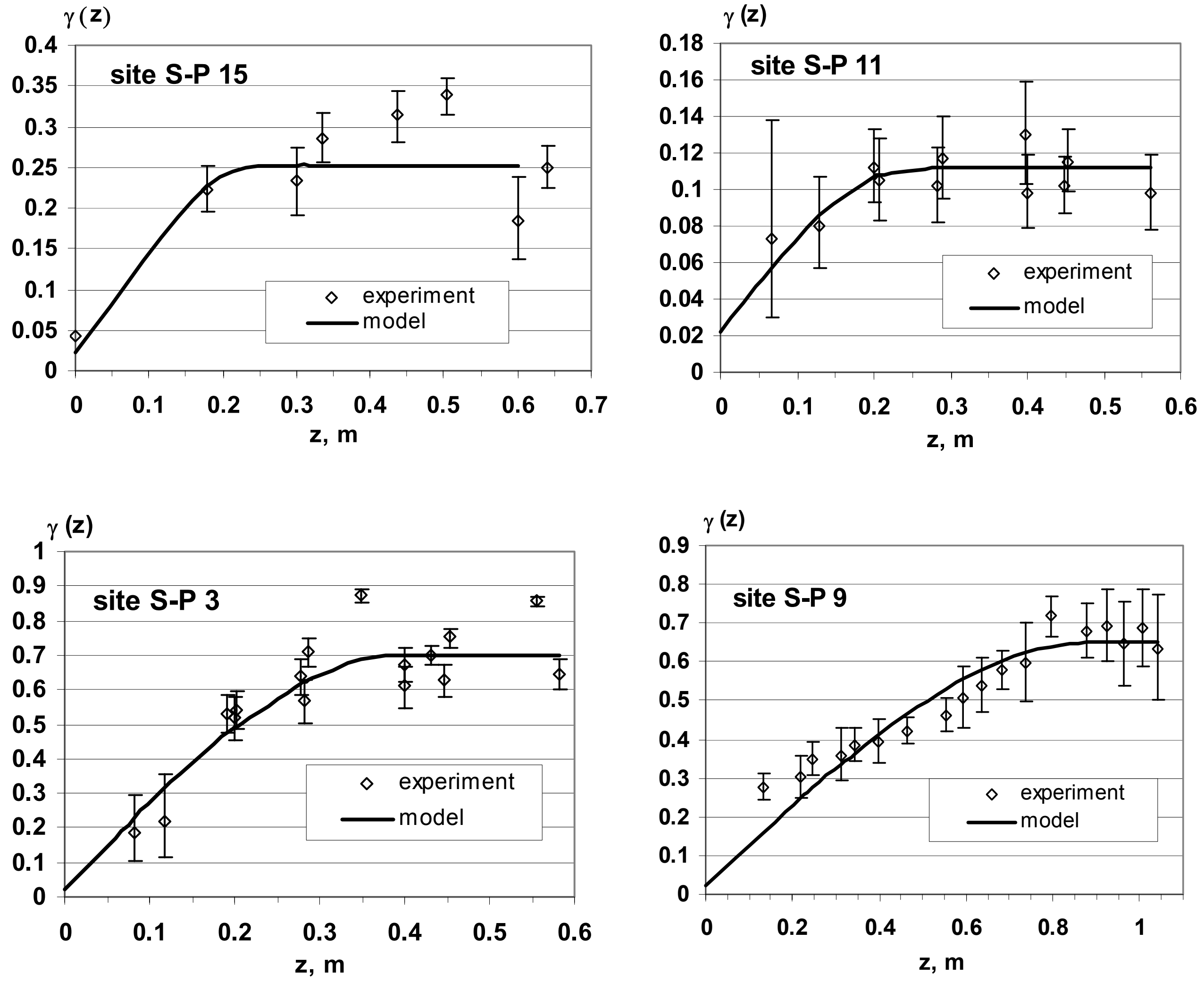


Fig. 7.1. Typical variograms on ^{137}Cs when sampling by cylindrical sampler 3.7cm in diameter.

The question about dependence of radius influence of sample on the sampling square should not be considered closed .The fact is that the radius of sample influence cannot be less than a sampler radius. Therefore, when increasing the sampler working area, the radius of influence of the selected samples should also increase. However, according to existing measurement technique of the radionuclide content in soil samples, the same measured sample is selected out of any sample (big or small). This fact and also “hot” particles in samples can level expected effect. But this question was not the subject of the presented work and it demands separate consideration.

Nevertheless, it is possible to consider as established fact, that irrespective of density and type of radioactive fallout, landscape features and cultivation of the land, the average value of radius influence of soil sample, selected from the area at least no more than **0.014 m^2** at first approximation and taking into account error determination, radius of sample influence R_{sa} on ^{137}Cs does not exceed **1** m. (**Table 7.1**).

Table 7.1. Radiuses of sample influence ^{137}Cs on different sites, when sampling by a cylindrical sampler 3.7 cm in a diameter

The site	**The type of fallout**	**The type of land use**	R_{sa} **, м**
Site S-P 3	fuel, ***DFP***≈25%	fallow land	0.4
Site S-P 9	fuel, ***DFP***≈25%	ploughed field	0.9
Site S-P 7	fuel, ***DFP***≈25%	fallow land	0.15
Site S-P 11	fuel, ***DFP***≈25%	fallow land	0.25
Site S-P 13	condensate, ***DFP***<10%	ploughed field	0.4
Site S-P 15	condensate, ***DFP***<10%	ploughed field	0.25
Site S-P 16	condensate, ***DFP***<10%	waterless valley	0.7
Average value			0.44

Thus, radionuclide content in dot samples (including the samples consisting of several located nearby samplings made by a sampler 3.7cm in a diameter), selected on non–gradient site on distance (centre–to–centre sampling) > **1** m from each other at the sampling square no more than **0.014 m^2,** should be considered as statistically independent magnitudes (statistically independent samples).

7.1.2 Radius of vegetative samples influence

For the geostatistical analysis and estimation of the radius influence of plants sample, the same experimental sites and selected plant samples were used. Statistical characteristics of specific vegetation contamination have been estimated. As a result of the analysis received with the help of empirical plants contamination variogram, experimental sites have been divided into three groups. Sites, where the land is regularly worked or has been worked (ploughed) since 1986, in variograms behaviour practically there are no distinctions (they differ only with a threshold – a dispersion contamination of plants ^{137}Cs on a site).

This group (we will call it the first) included sites **S-P 13** (avena (sativa)), **S-P 15** (oenothera biennis (L)), **S-P 7** (secale (cereale)) and **S-P 17** (triticum (clurum vulgare)). Variograms of contamination of the plants selected on various former arable and fallow lands, experimental sites (which were not worked after accident) have been divided in two groups. Sites **S-P7, S-P11**, **S-P16**, **S-P18** are in the second group where calamagrostis epigeios (L) Roth was selected. In the third group there is one site **S-P 3** where elytrigia repens (L) Nevski grew. This site is characterised by the greatest radius influence of plants samples that, apparently, it is connected with the features of elytrigia repens (L) Nevski root system. Variogram of each group were normalised on corresponding dispersions and are incorporated into one, characterising a group as a whole (Fig. 7.2–Fig. 7.4).

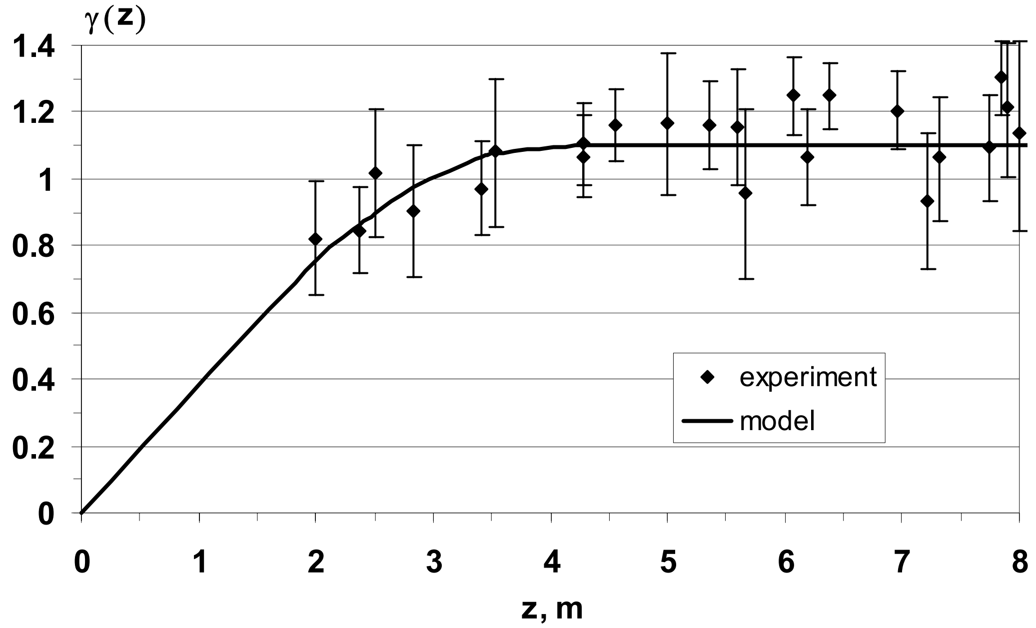


Fig. 7.2. The normalised generalised variogram of plants contamination with ^{137}Cs on the arable experimental sites (**S-P 13** – avena (sativa), **S-P 15** – oenothera biennis (L), **S-P 9** – secale (cereale), **S-P 17** – triticum (clurum vulgare)).

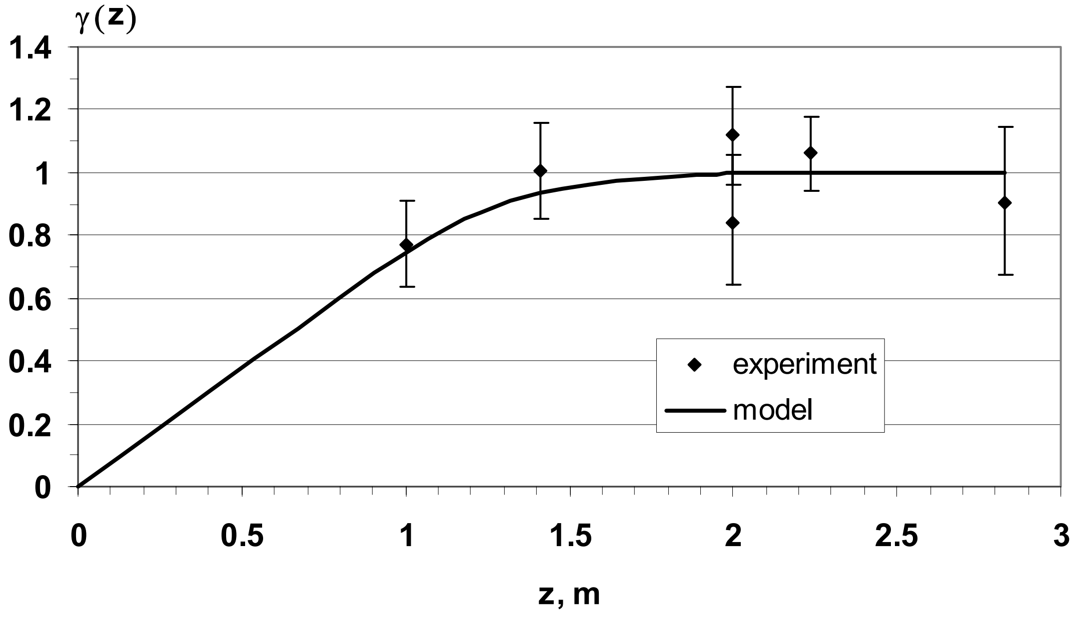


Fig. 7.3. The normalised generalised variogram of calamagrostis epigeios (L) Roth contamination with ^{137}Cs on the fallow and virgin land experimental sites (**S-P 7, S-P 11, S-P 16, S-P 18**).

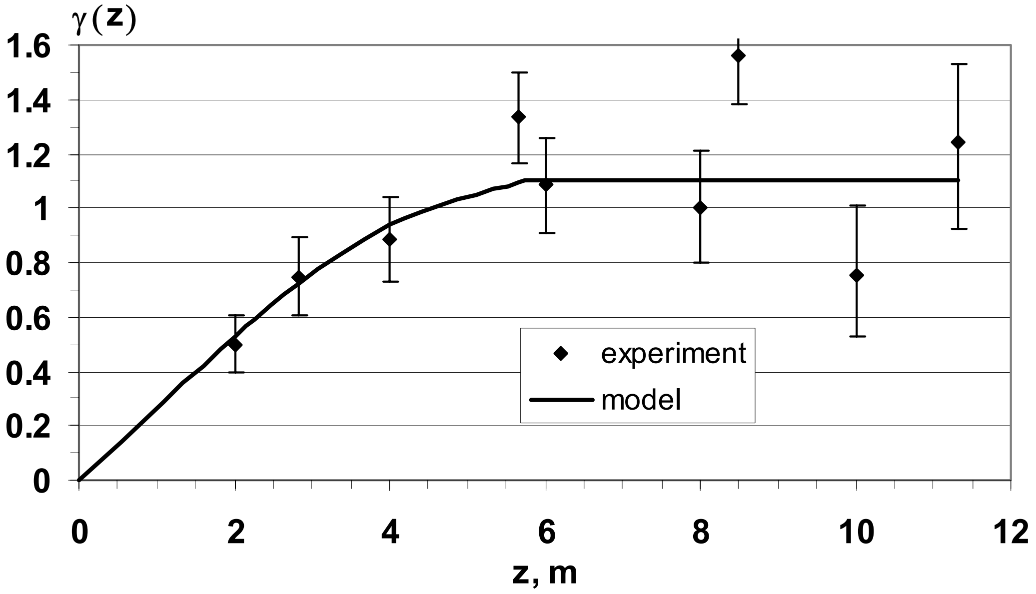


Fig. 7.4. The normalised generalised variogram of elytrigia repens (L) Nevski contamination with ^{137}Cs on the virgin land experimental site **S-P 3**.

Each sample of plants is selected, as a rule, on the area no more than 1 m^2 (a square with the side 1m), for a guaranteed estimation of radius influence of plants samples to distance on which modelling variogram is levelled, it is necessary to add 1 more meter (distance between borders of plants sampling). The radiuses influence of plants samples received by this method is shown in the table (Table 7.2). As well as at soil sampling, for statistical independence maintenance of plants samples in the selected set, and it is necessary to use conservative estimations of radius of sample influence in every sample.

Table 7.2. Estimations of radius influence of plants sample for various experimental sites at definition of the ^{137}Cs content.

№	Sites group	Radius of plants sample influence when defining ^{137}Cs content R_p, **m**
1	Arable lands (*triticum (clurum vulgare), secale (cereale), avena (sativa), oenothera biennis (L)*)	5
2	Former arable lands and fallow lands (*calamagrostis epigeios (L) Roth*)	2.8
3	Former arable lands and fallow lands (*elytrigia repens (L) Nevski*)	7

The following conclusion is possible to draw on the basis of the received results: the centre–to–centre distance of plants sampling not less than **8-10** m in all cases provides their statistical independence of the selected samples on ^{137}Cs content.

7.1.3 Minimum necessary centre to centre distance of conjugated samples selection when estimating transfer factors

The estimations received in the previous sections for radiuses of soil and plants samples influence, allow to estimate the minimum of permissible centre- to- centre distance of conjugated samples “soil – plant” and define the concept of samples conjugation more precisely.

Soil and plants sample selected on the same place at the same area is implied under conjugated sample “soil- plant”. The size of the sampling square is not indicated precisely. As a rule, soil sampling square is smaller than the plants sampling square.

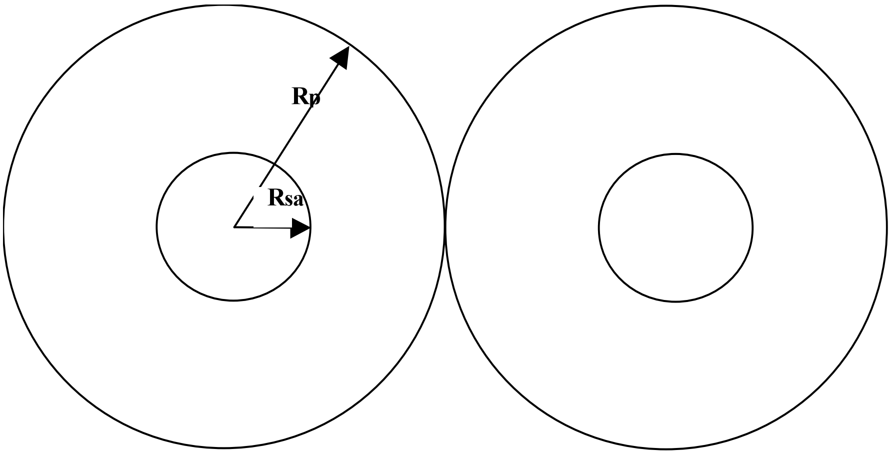


Fig. 7.5– The ratio of the radiuses to the influence zones of soil and plants samples scheme.

This scheme (Fig. 7.5) allows to define the rule for selection of the conjugated samples “soil - plant" more precisely and it is the following:

- When selecting conjugated samples “– plant”, the soil sample should completely characterise the area, where the plants are selected.
- If the vegetation is plentiful in researched territory and has high radionuclide content, then the conjugated samples can be selected on the basis of dot samples (the sampling area of soil samples > 0.001 m^2). Plants should be selected on the area limited to a circle with radius 0.4 m or in a square with the side 0.7 m with the center in the place of soil dot sample selection. In the other cases a plants sample is selected on some area (for example, 1m^2 or more), and soil contamination on this site is estimated on measurement of the incorporated sample, generated on the basis of soil dot samples, which areas of influence cover the chosen site, without crossing.
- The centre-to-centre distance of selection of independent conjugated samples should be not less than two radiuses of plants sample influence. As first approximation for the majority of plants it should be not less than **12 m**.

7.2 Local and global geostatistical characteristics of radioactive soil contamination in 30 km Ch NPP zone

At strongly pronounced spottiness of radioactive Chernobyl fallout, especially in 30 km zone, either non–gradient site occupies a small area, characterises a local place. Geostatistical contamination characteristics of such sites, caused by local heterogeneity of radioactive fallout, later will be called local as well as variograms corresponding to them.

Geostatistical contamination characteristics of big territories, which contamination variability is also caused by the gradients of radioactive fallout, will be called global, as well as variograms corresponding to them. The typical kind of local variogram, which are received on the basis of dot sampling, is shown in figure (7.1).

Local variograms parameters of contamination with radionuclide depend not only on micro heterogeneity of contamination of non–gradient site itself, but also on sampling methods and measurement of the radionuclide content in a dot sample.

The results shown in the previous sections of the present work, allow to receive models of local variogram of soil contamination with radionuclide (geostatistical characteristics of non–gradient sites) for various methods of dot sample selection and measurements. The variogram threshold $G+G_0$ – depending on the sampling square, is recalculated according to expression **(2.9**). At the sampling square > 0.005 m^2 it practically does not depend on type of radioactive fallout and its value at first approximation for logarithms of soil density contamination with ^{137}Cs, in case of radionuclide measurement error variogram ≤ 10 % at level $\pm 2\sigma$, now (calculated values for 2003) is **0.096±0.01** ($G+G_0=s^2_{5,n}$). For parameter G_0 (the general dispersion of the contamination logarithm in a point caused by effect of "nuggets" –by the presence of fuel particles) also the estimations in view of various physical and chemical features of radioactive fallout have been received and their dynamics has been researched (see section 5).

More than 1300 incorporated soil samples were selected when mapping 30-km Ch NPP zone with^{90}Sr contamination [52]. Samples were selected by a sampler 3.7cm in diameter to a depth of 30 cm in five points by the method of "envelope" with the side of 2-5 m in every sampling place. The uniform incorporated sample, with a weight of about 3 kg was also formed. The measurements of the ^{137}Cs content in a sample were carried out by γ–Spectrometer ADCAM-300 in four samples (3 cylindrical vessels in volume of 100 cm^3 and a Marinelle flask in volume of 1000 cm^3). The results of the measurements, kept in a databank, allow constructing variograms for the whole 30 km zone, and its separate sites at any chosen direction. Examples of global variogram, received on the basis of this sampling for various parts of 30 km zone (traces of radioactive fallout) and directions, for the logarithm of soil density contamination with ^{137}Cs are shown below in figures (Fig. 7.6, Fig. 7.7).

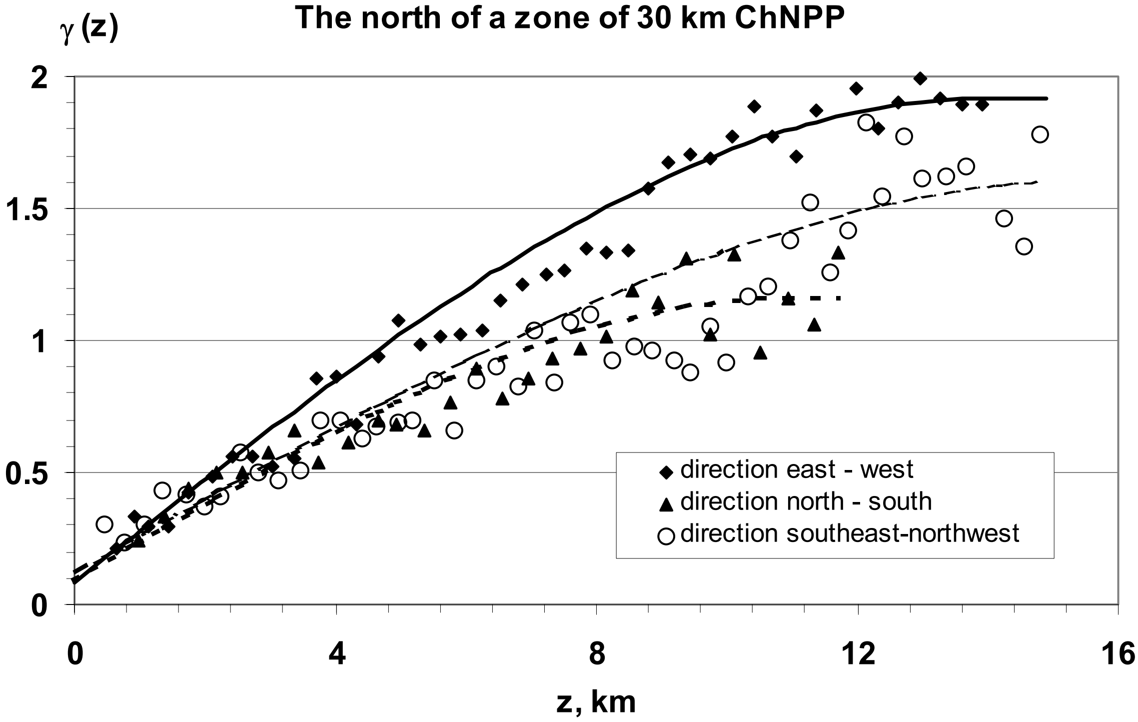


Fig. 7.6. The variograms of soil contamination with ^{137}Cs in the northern part of the ChNPP 30-km Zone.

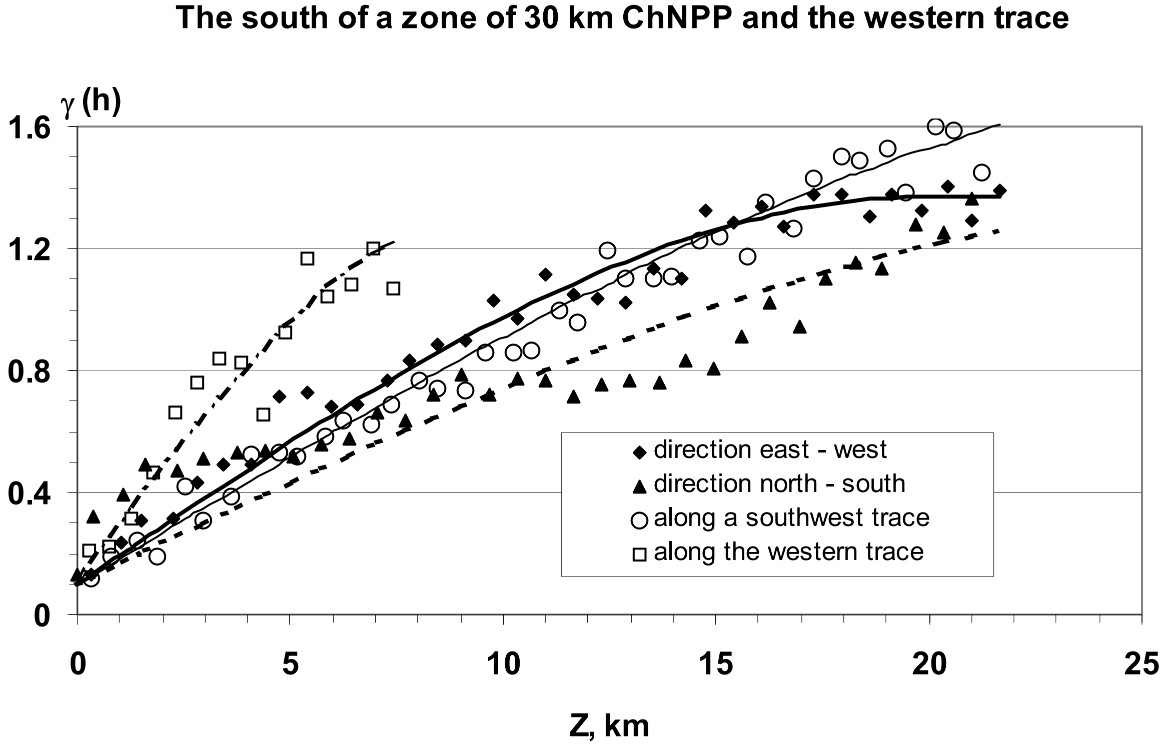


Fig. 7.7. The variograms of soil contamination with ^{137}Cs in the southern part of the ChNPP 30-km Zone and on the western trace.

It is visible in them, that global variogram parameters depend on a direction, anisotropy of territory contamination with radionuclide, caused by the generated traces of radioactive fallout is observed. This factor, and also significant gradients and fallout spottiness cause specific character and a global variogram variety of soil contamination with ^{137}Cs for various parts of 30-km zone Ch NPP and directions. However these variograms have one common feature, behaviour. All of them have one and the same value of parameter G_0 – effect of "nuggets". For given variograms this value equals **0.1**±0.02.

As the effect of "nuggets" is a dispersion of the logarithm contamination of sample at the point of selection (in this case the incorporated sample), it characterises a dispersion of logarithm of soil contamination on the area of chosen "envelope". In view of insignificant sizes of the "envelope", it could be considered as non–gradient on contamination site. Using the results of soil samples measurements kept in UIAR databank, which was created at mapping 30-km zone Ch NPP contamination with ^{90}Sr and a parity (6.12), an average on 30-km zone estimation of logarithm dispersion of soil contamination with ^{137}Cs on the area of such envelope at sampling by a sampler 3.7 cm in a diameter (a dispersion of the incorporated sample) for the moment of selection (1997) was received. This estimation equals **0.07**±0.025. In view of available errors it practically coincides with the value of the "nuggets" effect received on the basis of the empirical variogram analysis.

Thus, it is possible to prove, that a threshold of local soil contamination variograms with ^{137}Cs $G+G_0$ (a general dispersion of the logarithm contamination of non–gradient site) at the first approximation is the estimation for dispersion, describing the effect of "nuggets" of global variogram. That is to say there is a uniform geostatistical picture of territory radioactive contamination, realised in consequence of Ch NPP accident and in structure of which non–gradient on contamination sites play the role of elementary components. Their statistical characteristics are a basis for construction of the generalised geostatistical models of big territories radioactive contamination.

7.3 Conclusions of chapter 7.

As a result of the carried out analysis it has been established:

- Irrespectively of density and type of radioactive fallout, landscape features and a kind of working the land, the average value of radius influence of soil sample selected from the area no more **than 0.014 m^2** at a first approximation, do not exceed **1 m**. The content of radionuclide in the samples, selected from such areas, on homogeneously polluted site at a distance more **than 1 m** from each other at first approximation will be statistically independent values.
- The radius of plant samples influence depends on a kind of plants and by results of the carried out researches is estimated by **5 – 7** m distance, depending on a kind of plants.
- Minimum necessary distance between sampling points of the plants, provide statistical independence of plants samples equal **8-10** m.

8. THE CONCLUSION

Radioactive contamination of ecological monitoring objects has the statistical nature. Therefore, studying statistical characteristics of various objects radioactive contamination of the environment (territory, vegetation, animals) and agricultural grounds and production in particular is the important and necessary condition of the correct understanding and the description of sources of person's dose loads. The knowledge of statistical characteristics of various objects radioactive contamination of the environment allows to plan, organise and carry out their radio ecological researches and ecological monitoring most precisely

Sampling and measurement are determining bases of ecological monitoring. The results of analysis presented in the monograph and long-termed researches generalisations, carried out in UIAR during the liquidation of the consequences of Ch NPP accident, allow to improve successfully the methodology of carrying out experimental works on selection of representative dot samples and defining radionuclide content in them in view of monitoring objects contamination with "hot" (fuel) particles. This, in its turn, allows to form the representative sets of sample describing adequately radionuclide contamination of either monitoring object. The unique results, describing contamination of non–gradient sites (fields, arable lands, areas) on various traces of Chernobyl fallouts; various plants growing on them, milk of cows in settlements is represented in the monograph.

On non–gradient with contamination sites, density of soil contamination with **^{137}Cs**, its content in vegetation and corresponding transfer factors submit to the law of logarithmically normal probability distribution. The law also describes ^{137}Cs content at the specific moment of time in milk of cows on private farms on common pastures. Corresponding parameters of probability distribution characterising dispersion of soil, plants and milk contamination are estimated.

- The standard deviation of the logarithm soil density contamination with^{137}Cs on non–gradient with contamination sites when sampling area > 0.005m^2 does not depend on density contamination, types of fallout, features of a landscape and at the first approximation when the error of measurement of the radionuclide content in soil samples ≤ 10 % at a level ±2σ is estimated by magnitude 0.30±0.09.
- The standard deviation of the logarithm of the specific content ^{137}Cs in samples of plants does not depend on density contamination, types of fallout, kind of vegetation. At first approximation when the plants sampling square ≤ 1m^2 and a ratio error of measurement ^{137}Cs content in vegetative samples ≤ 10 % at a level ±σ is estimated by magnitude 0.37±0.11.
- The standard deviation of the logarithm of ^{137}Cs transfer factor into plants does not depend on density contamination, types of fallout, kind of vegetation and its different parts; at first approximation when selecting conjugated samples "soil - plant", when sampling square for plants ≤ 1m^2 and > 0.005 m^2, for soil

and ratio error of measurement ^{137}Cs content in soil and plants samples do not exceed10 % at a level ±σ , it is estimated by magnitude 0.49±0.06.

- The standard deviation of the logarithm of the specific content ^{137}Cs in milk of cows does not depend on settlement location; for homogeneous on pasture conditions (maintenance) of animal herds, when error of measurement, does not exceed 15 % at a level ±2σ it equals 0.67±0.19 during the stabling period and 0.56±0.19 during the pasturable period.

The received results have underlain a lot of scientific - methodical UIAR elaboration with great practical value when carrying out ecological monitoring of soil, plants and milk in settlements. The following should be mentioned:

- stochastic model, describing specific milk contamination with ^{137}Cs on private farms during a year;
- the calculation method of minimum necessary samples number for estimating the median of soil density contamination with^{137}Cs on non–gradient site with the set ratio error;
- the calculation method of minimum necessary samples number for estimating the median of the specific content ^{137}Cs in vegetation on uniformly polluted site with the set ratio error;
- the definition method of minimum necessary samples number for estimating the median of the specific milk contamination with ^{137}Cs with the set ratio error, during both, and stabling periods;
- the definition method of minimally necessary volumes of milk sampling during the pasturable and stabling periods, which guarantee the set ratio error of estimation average-annual individual dose of internal irradiation with different frequency of sampling;
- the calculation method of minimum necessary measured samples number for defining, with the set ratio error, the median ^{137}Cs, ^{90}Sr ($^{239+240}$Pu) content in the individual sample of soil containing fuel particles.

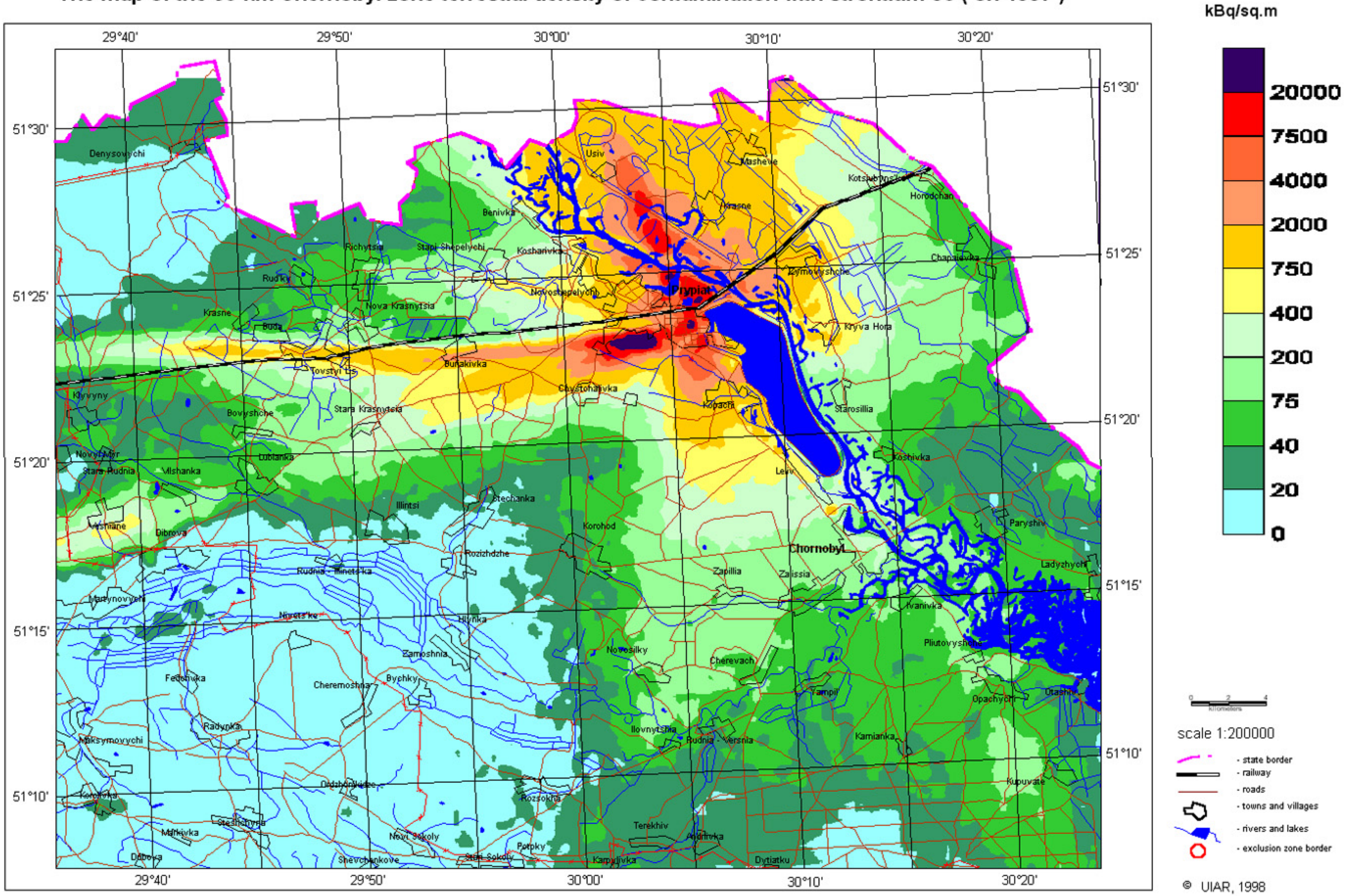
The researches to define average values of radiuses influence of soil and various plants have been carried out. The influence of density and types of radioactive fallout, landscape features, working the land, kind of plants has also been annualised. Irrespective of density and type of radioactive fallout, landscape features and a kind of working the land, the average value of radius influence of soil sample selected on the area no more **than 0.014 m^2** at first approximation does not exceed **1 m**. The radionuclide content in the samples selected on such areas, on homogeneously polluted site on the distance more **than 1** from each other **m** at first approximation will be statistically independent values. The radius influence of plants samples depends on kind of plants and is estimated by magnitudes **5 – 7** m by the results of released researches, depending on kind of plants. Minimum necessary distance between the plants sampling centres, providing statistical independence of plants samples equals **8-10** m, depending on kind of plants. When selecting statistically independent conjugated samples, the centre–to–centre distance between plants should not be less than **10 m.**

The analysis of long-termed results of measurement of radionuclide content in soil samples selected on various traces of Chernobyl fallout, allowed to establish the following fact. Soil samples selected on fuel traces (in particular in 30 km zone), owing to the presence of "hot" (fuel) particles have essential volumetric heterogeneity, which can not be eliminated when standard process of homogenisation. ^{137}Cs content in individual soil sample, with volumetric heterogeneity, is a random variable and is well described by the logarithmically normal law of probability distribution. Standard deviation of the logarithm specific activity of dot (individual) sample ^{137}Cs, caused by its volumetric heterogeneity, for 30 km zone Ch NPP for 1997 when measuring by γ–Spectrometer is characterised by values: **0.14±0.01** (an average estimation), **0.23±0.03** (a conservative estimation) for a cylindrical sample of 100 cm^3 in volume; **0.11±0.02** (an average estimation), **0.17±0.03** (a conservative estimation) for Marinelle flask of 1000 cm^3 in volume. The corresponding average estimation of standard deviation when measuring samples of 100 cm^3 in volume for ^{90}Sr ($^{239+240}$Pu) equals **0.20±0.05** (an average estimation) and **0.17±0.05** (an average estimation), when measuring ratio Cs/Sr in samples of 100 cm^3 in volume. The calculation method of minimum necessary measured samples number, to define, with the set ratio error, the median ^{137}Cs and ^{90}Sr ($^{239+240}$Pu) content in individual soil sample containing fuel particles has been suggested.

The received results allow to plan more precisely the number of selected samples and measurements when ecological monitoring in the exclusion zone and in adjoining territories and to carry out it with the minimal expenses, guarantee of the set estimations accuracy of controllable parameters.

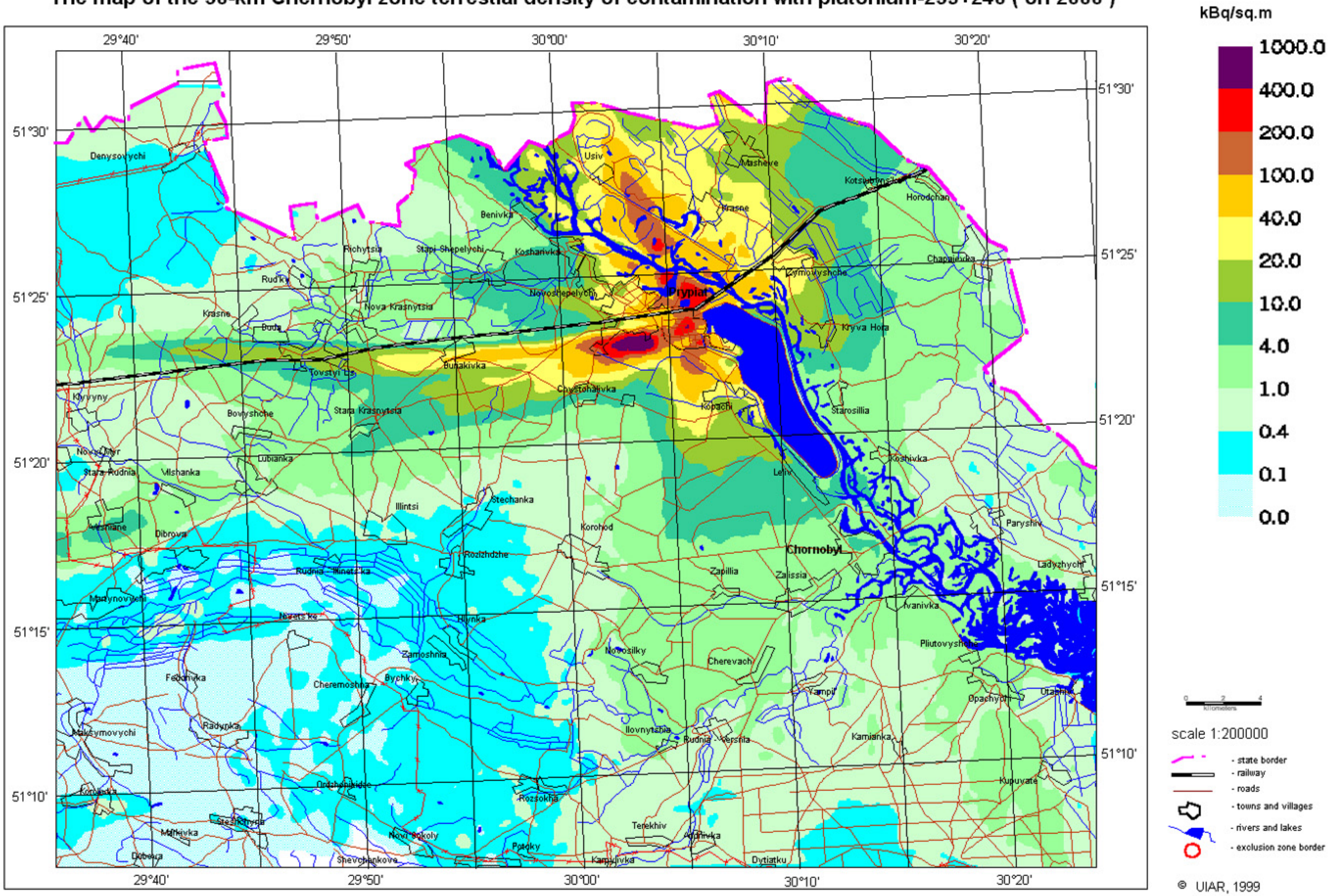
Figures on a cover

The map of the 30-km Chernobyl zone terrestial density of contamination with strontium-90 (on 1997)



© UIAR, 1998

The map of the 30-km Chernobyl zone terrestial density of contamination with plutonium-239+240 (on 2000)



9. References

---

1. Baryakhtar V.G., 1997. Chernobyl Catastrophe. Kiev, “Export Publishing House”, p. 572.
2. Kuriny V.D., Ivanov Yu.A., Kashparov V.A., Loschilov N.A., Protsak V.P., Yudin E.B., Zhurba M.A., Parshakov A.E. Particle Associated Chernobyl Fall-Out in the Local and Intermediate Zones. //Annals of Nuclear Energy. -1993, v.20, N.6, p.415-420.
3. Kashparov V.A. Hot Particles at Chernobyl //Environmental Science and Pollution Research, v.10/1, 2003, pp.21-30.
4. Kashparov V.A., Ivanov Yu.A., Zvarich S.I., Protsak V.P., Khomutinin Yu.V., Kurepin A.D., Pazukhin E.M. Formation of Hot Particles During the Chernobyl Nuclear Power Plant Accident. //Nuclear Technology. - 1996, v.114, N.1, p.246-253.
5. Cambray R.S., Cawse P.A., Garland J.A., Gibson J.A.B., Johnson P., et al. Observations on radioactivity from the Cernobyl accident // Nuclear Energy. – 1987. - Vol. 26, N 2. – P. 77-101.
6. Joshi S.R. Early canadian results on the long-range transport of chernobyl radioactivity // The Science of the Total Environment. - 1987. - 63. – P. 125-137.
7. Costa Charles, Kuroda P.K. Fission Products in Air over the United States after the 1986 Chernobyl Event // Radiochimica Acta. - 1989. - 47. – P. 199-208.
8. Gudiksen P.H., Harvey T.F., Lange R. Chernobyl Source term, atmospheric dispersion, and dose estimation // Health Physics. – 1989. - Vol. 57, N5. – P. 697-706.
9. Joshi S.R. The Fallout of Chernobyl Radioactivity in Central Ontario, Canada // Journal Environ. Radioactivity. - 1988. - №6. – P. 203-211.
10. Pollanen R., Valkama I., Toivonen H. Transport of radioactive particles from the Chernobyl accident //Atmospheric Environment.- 1997, 31/21, p. 3575-3590.
11. ApSimon H.M., Wilson J.J.N., Simms K.L. Analysis of the dispersal and deposition of radionuclides from Chernobyl across Europe // Proceedings of Radiation Society. - London. – 1989. -A 425. – P. 365-405.
12. Izrael Yu.A. Ecology and monitoring the Environmental situation.– Leningrad, Gidrometeoizdat Publishing House, 1979.–375 p. (In Russian).
13. Lundin S.M., Kashparov V.A., Khomutinin Yu.V., Kadygrib A.M. Problem of optimal sampling in the radioactive contaminated agricultural lands and the ways of its solution // Problems of agricultural radiology. Collection of scientific works by UIAR, V.3, Kiev, 1993, pp.17-30. (In Russian).
14. Kashparov V.A.,Yoshchenko V.I.,Levtchuk S.E.,Tschiersch J.,Wagenpfeil F. Application of the method of repeated mixing to non-uniformly contaminated bulky samples.// Journal of Radioanalytical and Nuclear Chemistry, 2000, vol.246, No.1, pp.165-172.

15. Khomutinin Yu., Kashparov V., Zhebrovska K. Sampling optimisation at the radioecological monitoring //Materials of “Ecorad 2001” – France, 2001. – P6/14(288).
16. Khomutinin Yu.V., Kashparov V.A., Zhebrovskaya E.I. (2001): Optimization of sampling and measurements in radioecological monitoring. Kyiv, “VIPOL Publishing House”, 160 p (in Russian).
17. Khomutinin Yu.V. Optimization of sampling for assessment of the terrestrial density of radioactive fallout // Collection of scientific works by Institute of Nuclear Researches, V.1(9), Kiev.–2003.–p.145-155. (in Russian).
18. Kashparov V.A., Oughton D.H., Zvarich S.I., Protsak V.P., Levchuk S.E. Kinetics of fuel particle weathering and ^{90}Sr mobility in the Chernobyl 30-km exclusion zone // Health Physics, 1999, vol.76, N.3, p.251-259.
19. Kashparov V.A., Ahamdach N., Zvarich S.I., Yoschenko V.I., Maloshtan I.N., Dewiere L. Kinetics of dissolution of Chernobyl fuel particles in soil in natural conditions. //Journal of Environmental Radioactivity, v.72, Issue 3, 2004, p.335-353.
20. Kashparov V.A., Protsak V.P., Ahamdach N., Stammose D., Peres J.M., Yoschenko V.I., Zvarich S.I. Dissolution kinetics of particles of irradiated Chernobyl nuclear fuel: influence of pH and oxidation state on the release of radionuclides in contaminated soil of Chernobyl //Journal of Nuclear Materials, v. 279, 2000a, p.225-233.
21. Shestopalov V.M., Kashparov V.A., Ivanov Y.A. Radionuclide Migration into the Geological Environment and Biota Accident // Environmental Science and Pollution Research, v.10 Special (1), 2003, pp.39-47.
22. Methodical recommendations for the complex radiological monitoring in the territories of NPPs disposal. – Odessa, 1986.– 21 p. (in Russian).
23. Methodical guideline for survey in 1991-1992 of the agricultural areas in the zone contaminated with the radionuclides. – Kiev, 1991.–11 p. (in Russian).
24. Methodical recommendations for assessment of the radiation situation in the settlements. – Moscow, 1990.– 118 p. (in Russian).
25. Express-method for assessment of the terrestrial density of the agricultural lands contamination with cesium isotopes using the data of gamma-survey taking into account radionuclide composition and radionuclides distribution in soil profile. – Kiev, 1989.–12 p. (in Russian).
26. Guideline for soil sampling at the radiation survey of the contaminated territory. – Moscow, Inter-branch commission, 1987. – 10 p. (in Russian).
27. Makhon’ko K.P., Silantiev A.N., Shkuratov I.G. Monitoring of the radioactive contamination of the Environment in the NPPs vicinity. – Leningrad, Gidrometeoizdat Publishing House, 1985.–131 p. (in Russian).
28. Yakovlev Ye.A., Ostavnenko A.I., Bondareva N.M. et al. Temporary methodical recommendations for radioecological researches in the NPP influence zone. – Kiev, Ministry of Energy of USSR, 1990.–98 p. (in Russian).

29. Temporary requests for complex monitoring and evaluation of radiological quality of the contaminated territories in various landscape and geochemistry zones. Methodical recommendations. – Kiev, 1996. – 9 p. (in Russian).
30. Smirnov N.V., Dunin-Barkovsky I.V. Probability theory and mathematical statistics for technical applications. – Moscow, Nauka Publishing House, 1965.–511 p. (in Russian).
31. Kendall M.G. and Stuart A. The Advanced Theory of Statistics, Vol. 2, Hafner, New York, 1973.
32. Hahn G., Shapiro S. Statistical Models in Engineering, Wiley, New York, 1967.
33. Afifi A., Eisen S. Statisitical analysis. PC-based approach. – Moscow, Mir Publishing House, 1992. – 488 p. (in Russian).
34. Military Standart 105D. Sampling procedures and tables for inspection by attributes.–US Government Printing Office, Washington, 1963.
35. Knowler L., Howell J., Gold B., Coleman E., Moan O., Knowler W. Quality Control by Statistical Methods.– McGraw-Hill Book Company, New York , 1969.
36. GOST 27.502–83. Reliability in techniques. System of acquisition and treatment of information. Planning the observations. – Moscow, Publishing House for Standards, 1984. – 23 p. (in Russian).
37. Belyaev Yu.K. Probabilistic methods for test sampling.– Moscow, Nauka Publishing House, 1975.– 406 p. (in Russian).
38. Reliability and efficiency in techniques. Reference book in 10 volumes. V.7. Quality and reliability in industries. Ed. by I.V.Appolonov, 1989. – 280 p. (in Russian).
39. Kogan R.I. Integral estimates in the geological researches. Reference book. – Moscow, Nedra Publishing House, 1986. – 160 p. (in Russian).
40. Tkachev Yu.A., Yudovich Ya.E. Statistical treatment of the geochemical data. Methods and problems. – Leningrad, Nauka Publishing House, 1975. – 233 p. (in Russian).
41. Usikov Yu.T. Reliability of geological survey information. – Moscow, Nedra Publishing House, 1988.– 120 p. (in Russian).
42. Gavrishyn A.I. Assessment and quality monitoring of the geochemical information.– Moscow, Nedra Publishing House, 1980.–287 p. (in Russian).
43. Al’bov M.N. Tests of the mineral deposits. – Moscow, Nedra Publishing House, 1975.– 232 p. (in Russian).
44. Kazhdan A.B. Exploration of the mineral deposits. – Moscow, Nedra Publishing House, 1984. – 285 p. (in Russian).
45. Chetverikov L.I. Theoretical basis for depths exploration.– Moscow, Nedra Publishing House, 1984. –156 p. (in Russian).
46. Guide for creation of soil and agrochemical maps. Ed. by A.V.Sokolov. – Moscow, Kolos Publishing House, 1964. – 384 p. (in Russian).
47. Agrochemical methods of soil analysis. Guide for field and laboratory researches. – Moscow, Publishing House of the USSR Academy of Science, 1960. – 555 p. (in Russian).

48. Manual for soil analyses and agrochemical maps creation. – Moscow, Rosselhozizdat Publishing House, 1965. – 330 p. (in Russian).
49. GOST 17.4.3.01–83. Conservancy. Soils. General requests to the sampling. – Moscow, Publishing House for Standards, 1984. – 4 p. (in Russian).
50. GOST 28167–89. Soils. Sampling – Moscow, Publishing House for Standards, 1989.–7 p. (in Russian).
51. GOST 17.4.4.02–84. Conservancy. Soils. Methods of sampling and sample preparation for chemical, bacteriological and helminthologic analyses. – Moscow, Publishing House for Standards, 1985. – 11 p. (in Russian).
52. Kashparov V.A., Lundin S.M., Khomutinin Yu.V., Kaminsky S.P., Levtchuk S.E., .Protsak V.P., Kadygrib A.M., Zvarich S.I., Yoschenko V.I., Tschiersch J. Soil contamination with ^{90}Sr in the near zone of the Chernobyl accident // Journal of Environment Radioactivity, v.56, № 3, 2001, p.285-298.
53. Kashparov V.A., Lundin S.M., Zvarich S.I., Yoschenko V.I., Levtchuk S.E., Khomutinin Yu.V., Maloshtan I.N., Protsak V.P. Territory contamination with the radionuclides representing the fuel component of Chernobyl fallout //The Science of The Total Environment, vol.317, Issues 1-3, 2003, pp. 105-119.
54. Zaidel' A.N. Elementary estimates of the measurement uncertainties. – Leningrad, Nauka Publishing House, 1967. – 88 p. (in Russian).
55. Taylor J. Introduction into theory of errors. – Moscow, Mir Publishing House, 1985. – 272 p. (in Russian).
56. Novitsky P.F., Zograf I.A. Estimate of measurement uncertainties. – Leningrad, Energoatomizdat Publishing House, 1985. – 248 p. (in Russian).
57. Rudzit Ya.A., Plutalov V.N. Basics of metrology, accuracy and reliability in instrument-making. – Moscow, Mashinostrojenije Publishing House, 1991. –304 p. (in Russian).
58. Aronov V.I. Methods of mapping the geologic and geophysical indexes and of geometrization of oil and gas deposits using PC. – Moscow, Nedra Publishing House, 1990. – 304 p.(in Russian).
59. Atlas of the Radioactive Contamination of Ukraine, CD of the Ministry of Emergencies of Ukraine, 2002.
60. Molchanova I.V., Karavayeva Ye.N., Pozolotina V.N., Yushkov P.I., Mikhailovskaya L.N. Regularities of radionuclides behavior in flood-land landscapes of Techa river in Ural // Ecology, V3, 1994, pp.43-49. (in Russian).
61. Karavayeva Ye.N., Molchanova I.V. Radionuclides behavior in over-wetted soils in the zones impacted by the nuclear enterprise in Ural // Ecology, V.3, 1997, pp.191-194(in Russian).
62. Agriculture in conditions of the radioactive contamination of Ukrainian territory as a result of the Chornobyl accident. Methodical recommendations. – Kiev, Ministry of Agriculture of Ukraine, Ministry of Emergencies of Ukraine, UIAR, 1998. – 103 p. (in Ukrainian).
63. Prister B.S., Khomutinin Yu.V., Perepelyatnikova L.V. Assessment of the "guaranteed" transfer factors of radioactive cesium in the agronomic crops using the

data of soil agrochemistry // Problems of agricultural radiology. Collection of scientific works by UIAR. – Kiev, 1991. V.1. – pp. 132-141. (in Russian).

64. Fesenko S.V., Chernyaeva L.G., Sanzharova N.I., Alexakhin R.M. Probabilistic approach to prognosis of the radioactive contamination of the agricultural production // Atomnaya energiya (Nuclear Energy). – V.74, Issue 6, 1993. – pp. 507–513. (in Russian).
65. Geras'kin S.A., Fesenko S.V., Sanzharova N.I., Chernyaeva L.G. Statistical methods of analysis of the empirical distributions of radionuclides concentration ratios in plants // Agricultural biology, Ser. Biology of Plants.– V.1, 1994. – pp. 130-137. (in Russian).
66. Huber P.J. Robust Statistics. – N.-Y.: Wiley, 1981.-308p.
67. Hampel F., Ronchetti E., Rousseeuw P., Stahel W. Robust Statistics. The Approach Based on Influence Function.– New York, 1986.
68. Nosov V.N. Computer biometry. – Moscow, Publishing House of MSU, 1990.– 232 p. (in Russian).
69. Bondar' P.F. Influence of soil-climate conditions on ^{89}Sr accumulation from soil in plants and prognosis of levels of the harvest contamination // Agrochemistry, V.7, 1983. – p.69. (in Russian).
70. Bondar' P.F., Loshchilov N.A., Tereshchenko N.R., Maslo A.V. Numerical characteristics of radiocesium accumulation in agricultural crops from soddy-podzolic loamy soil of Ukrainian Polessie // Problems of agricultural radiology. Collection of scientific works by UIAR. – Kiev, V.3, 1993. – pp.83-93. (in Russian).
71. Bondar' P.F., Shmatok I.O. Assessment of bioavailability of radiocesium and radiostrontium and its influence on radionuclides accumulation in harvest depending on agrochemical properties of soils // Problems of agricultural radiology. Collection of scientific works by UIAR. – Kiev, V.4, 1996. – pp. 124-143. (in Russian).
72. Ivanov Yu.A. Radioecological substantiation of the long-term prognosis of radiation situation in agricultural lands in a case of the large nuclear accidents (on example of the accident on the Chernobyl NPP). Abstract of the thesis for scientific degree of Doctor of Biology. – Obninsk, 1997. – 50 p. (in Russian).
73. Annenkov B.N., Yudintzeva Ye.B. Principles of agricultural radiology. – Moscow, Agropromizdat Publishing House, 1991. – 287 p. (in Russian).
74. Prister B.S., Loshchilov N.A., Nemetz O.F., Poyarkov V.A. Principles of agricultural radiology. – Kiev, Urozhay Publishing House, 1991. – 474 p. (in Russian).
75. Agricultural radioecology / Alexakhin R.M., Vasiliev A.V., Dikarev V.G. et al. Eds. Alexakhin R.M. and Korneev N.A. – Moscow, Ecology Publishing House, 1992. – 400 p. (in Russian).
76. Astasheva N.P., Romanov N.P., Kostuk D.M., Khomutinin Yu.V. Dynamics of accumulation and clearance of radionuclides in organisms of agricultural animals // Problems of agricultural radiology. Collection of scientific works by UIAR. V.1. – Kiev, 1991. – pp. 160-170. (in Russian).
77. Methodical guidelines for determination of ^{90}Sr and ^{137}Cs activities in soils and plants. – Moscow, 1985. – 64 p. (in Russian).

78. Kashparov V.A., Lundin S.M, Kadygrib A.M., Protsak V.P., Levtchuk S.E., Yoschenko V.I., Kashpur V.A., Talerko N.M. Forest fires in the territory contaminated as a result of the Chernobyl accident: radioactive aerosol resuspension and exposure of fire-fighters //Journal of Environmental Radioactivity, v.51, 2000, p.281-298.
79. Kashparov V.A., Protsak V.P., Ivanov Yu.A., Nicholson K.W. Resuspension of Radionuclides and Contamination of Village Areas Around Chernobyl. //J. Aerosol Science. –1994, v.25, No.5, p.755-761.
80. Kashparov V.A., Protsak V.P., Yoschenko V.I., Watterson J.D. Inhalation of Radionuclides During Agricultural Work in Areas Contaminated as a Result of the Chernobyl Reactor Accident. //J. Aerosol Science. –1994, v.25, No.5, p.761-767.
81. K.Bunzl. Probability for Detecting Hot Particles in Environmental Samples by Sample Splitting. //Analyst. 1997. Vol.122. pp. 653-656.
82. K.Bunzl. Detection of radioactive hot particles in environmental samples by repeated mixing.// Appl. Radiat. Isot. Vol. 49 (1998) p.1625.
83. Kashparov V.A.,Yoshchenko V.I.,Levtchuk S.E.,Tschiersch J.,Wagenpfeil F. Application of the method of repeated mixing to non-uniformly contaminated bulky samples.// Journal of Radioanalytical and Nuclear Chemistry, 2000, vol.246, No.1, pp.165-172.
84. Recommendation of ^{90}Sr and ^{137}Cs content in soil and plant samples, Moscow, 1985.-64p. (in Russian).
85. Radiation-dosimetrical passportization of Ukrainian settlements subjected to radioactive contamination as a result of the ChNPP accident, including thyrodosimetry. Instruction-methodical guidelines. – Kyiv, Ministry of Public Health of Ukraine, AMS of Ukraine, Ministry of Emergencies of Ukraine, 1996. – 73 p. (in Ukrainian).
86. Rodionov A.R., Kogan R.I., Golubeva V.A., Smirnov B.I., Sirotiskaya S.V. Manual for mathematical methods in geology.– Moscow, Nauka Publishing House, 1987. – 334 p. (in Russian).
87. Matteron G. Principles of applied geostatistics. – Moscow, Mir Publishing House, 1968. – 408 p. (in Russian).
88. David M. Geostatistical methods for assessment of ore contents. – Leningrad, Nedra Publishing House, 1980. – 325 p. (in Russian).
89. Handbook of soil science./ Editor Malcolm E. Summer.– CRC PRESS, 1999.– ISBN 0-8493-3136-6.