METHOD FOR SIMULTANEOUS ^{90}Sr AND ^{137}Cs IN-VIVO MEASUREMENTS OF SMALL ANIMALS AND OTHER ENVIRONMENTAL MEDIA DEVELOPED FOR THE CONDITIONS OF THE CHERNOBYL EXCLUSION ZONE

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ABSTRACT

To perform in vivo simultaneous measurements of the ^{90}Sr and ^{137}Cs content in the bodies of animals living in the Chernobyl Exclusion Zone (ChEZ), an appropriate method and equipment were developed and installed in a mobile gamma beta spectrometry laboratory. This technique was designed for animals of relatively small sizes (up to 50 g). The ^{90}Sr content is measured by a beta spectrometer with a 0.1 mm thick scintillation plastic detector. The spectrum processing takes into account the fact that the measured object is "thick-layered" and contains a comparable quantity of ^{137}Cs, which is a characteristic condition of the ChEZ. The ^{137}Cs content is measured by a NaI scintillation detector that is part of the combined gamma beta spectrometry system. For environmental research performed in the ChEZ, the advantages of this method and equipment (rapid measurements, capability to measure live animals directly in their habitat, and the capability of simultaneous ^{90}Sr and ^{137}Cs measurements) far outweigh the existing limitations (considerations must be made for background radiation and the animal size, skeletal shape and body mass). The accuracy of these in vivo measurements is shown to be consistent with standard spectrometric and radiochemical methods. Apart from the in vivo measurements, the proposed methodology, after a very simple upgrade that is also described in the article, works even more accurately with samples of other media, such as soil and plants.

Key Words: Chernobyl, ^{90}Sr, ^{137}Cs, whole body counting.

INTRODUCTION

Soon after the nuclear accident that occurred at the Chernobyl Nuclear Power Plant (ChNPP) in 1986, ^{137}Cs and ^{90}Sr became the most ecologically significant radionuclides released because of their relatively long half lives (about 30 y) and because of their persistence in the environment. Many standard methods have been developed to measure the ^{137}Cs and ^{90}Sr content in liquid, gaseous, and solid matter (including biota). For example, the ^{137}Cs content is typically directly measured using a gamma-spectrometer to identify the 0.66 MeV gamma emission of its short-lived daughter - ^{137m}Ba. For ^{90}Sr, various methods are used for its radiochemical separation followed by measurements using either beta radiometers or direct radiometry in which case the ^{90}Sr content is assessed indirectly by measuring the daughter ^{90}Y product and rejecting ^{90}Sr, ^{137}Cs, and ^{40}K beta particles with a lower energy. However, these measurements are always separated in time and, in addition, ^{90}Sr measurements are time-consuming and costly. The only efficient method is the ^{90}Sr beta spectrometry method based on the daughter ^{90}Y measurements mentioned above. However, this method is intended for homogenous samples without significant quantities of "interfering" radionuclides (e.g. ^{137}Cs and ^{40}K).

The situation becomes even more challenging when measuring the ^{90}Sr and ^{137}Cs content in live animals and other biota. In vivo measurements of ^{137}Cs content have been conducted for the whole body of animals, specifically, in fish (Hadderingh et al. 1996; Belyaev 2001), birds (Brisbin et al. 1973; Straney et al. 1975; Levy et al. 1976; Domby et al. 1977; Fendley et al. 1977; Craig et al. 1979; Halford et al. 1983; George et al. 1990; Millard et al. 1990; Kennamer et al. 1993; Moss and Horrill 1996), and mammals (Richmond 1980). For this purpose, gamma spectrometric systems (or pulse counting devices) were developed and thoroughly calibrated,

which was a feasible but not necessarily easy task. In vivo ^{90}Sr measurements of animals using radiochemistry or radiometry have been conducted on a limited basis, but these measurements were only performed on a small amputated part (e.g. a tip of the tail or a toe of a mouse) (Ilyenko 1967). The development of in vivo ^{90}Sr measurements for whole body live animals is more difficult and these measurements are complicated by the facts that (1) body tissues heavily absorb beta particles and (2) the ^{90}Sr distribution within the body is extremely uneven (Brisbin 1991).

It would be difficult to overestimate the practical value of having the equipment and methods available for the rapid, in vivo, simultaneous measurements of ^{90}Sr and ^{137}Cs content in the whole body of animals. First, it would significantly improve the effectiveness of research studies due to the larger number of samples (animals) that could be analyzed in a given time. Second, it would make it possible to more adequately compare ^{90}Sr and ^{137}Cs exchange processes since they occur under the same conditions (in the same individuals). Third, such measurements would provide an opportunity for a long-term radioecological monitoring of animals under both laboratory and natural conditions. Finally, the presence of this method would, to a large extent, eliminate the need to kill the animals, thereby making it possible to perform radioecological research not just on common widespread species, but also on species protected by national and international conventions and laws [e.g., bat (*Chiroptera*) species].

Since theoretical prerequisites for developing such equipment existed and the total ^{90}Sr to ^{137}Cs ratio in the bodies of animals, plants and soils from the Chernobyl Exclusion Zone (ChEZ) varied from 10^{-1} to 10^{1}, the following tasks were identified: (1) develop and implement a method for in vivo ^{90}Sr concentration measurements in the whole body of animals; (2) develop and

implement a method for in vivo ^{137}Cs concentration measurements in the whole body of animals; and (3) deploy both methods in a single system that can be utilized in the field.

As discussed, task 2 has already been addressed for various biota, including humans. For the first task, beta spectroscopy was chosen for ^{90}Sr content measurements. However, the heterogeneous distribution of ^{90}Sr in animals (primarily in skeletons) and the presence of a comparable ^{137}Cs quantity in animals of the ChEZ present significant problems for in vivo ^{90}Sr beta spectrometry. To solve these problem, the following facts were taken into consideration: ^{137}Cs is a beta emitter (514 keV E_{max}), as is ^{90}Sr (546 keV E_{max}), and the beta-spectra of these two radionuclides are essentially the same (Browne and Firestone 1986). In addition, ^{137m}Ba, a daughter product of ^{137}Cs, is a source of gamma (661 keV) and characteristic x ray radiation (31.8-37.5 keV). This task is facilitated by the presence of beta irradiation from ^{90}Y (2270 keV E_{max}), a daughter product of ^{90}Sr. Its beta energy characteristics significantly differ from those of ^{137}Cs and ^{90}Sr, making it possible to separate the ^{90}Y component from the total spectrum and use it as the ^{90}Sr indicator. In this situation, gamma and x ray radiation appear as background interfering factors. These studies resulted in the development of a combined and mobile (vehicle deployed) gamma/beta spectrometry system.

METHODS AND MATERIALS

Mobile gamma/beta spectrometry system

The developed spectrometry system includes the following major components:

1) Counting chamber with shielding 100–50 mm thick lead walls and a steel pull-out drawer (100 × 300 × 100 mm) as a container for the animal to be measured in (item

1 in Figs.1a and 1b). The pull-out drawer has a lateral 90 mm diameter opening on the side, opposite to the location of the gamma detector (Fig. 1c);

2) beta-detector (scintillation plastic thinfilm 0.1 mm detector, 60 mm in diameter) installed vertically above the counting chamber, 11 cm away from the bottom of the pull-out drawer (item 2 in Figs. 1a and 1b);

3) Canberra NaI scintillation detector (Tl-activated, 63 mm in diameter) installed horizontally to the counting chamber (item 3 in Figs. 1a and 1b); the energy resolution of the detector is 50.1 keV for 661 keV, the spectrum was collected from 1,024 channels;

4) Canberra ASA-100 pulse height analyzer with «Beta+» software application for beta-spectra processing (Institute of Nuclear Research, Kiev, Ukraine) (item 4 in Fig. 1a);

5) Canberra InSpector-2000 Multi Channel Analyzer, and Canberra Genie-2000 software application for gamma-spectra processing (item 5 in Figs. 1a and 1b); and

6) Personal computer (item 6 in Figs. 1a and 1b).

The spectrometry system was installed in a bus and powered from a gasoline generator (Fig. 2), making it possible to deploy it in the field. Apart from live animals, this spectrometry system also was capable of measuring the total ^{90}Sr and ^{137}Cs content in pretreated soil and plants samples.

Spectrometers utilized for the comparative analysis

For evaluation of the characteristics of the beta spectrometer included into the mobile gamma/beta spectrometry system, same-sample comparisons were made between the

measurements performed using the beta spectrometer in the mobile system and the stationary beta spectrometers installed at the Chernobyl Center International Radioecology Laboratory (IRL). The technical specifications of all beta spectrometers used are provided in Table 1.

To assess the quality of the ^{137}Cs measurements using the mobile gamma/beta spectrometry system, we compared these measurements results with those obtained by using the IRL stationary gamma spectrometer system (Canberra-Packard HPGe detector, GC 3019).

Sample pretreatment for spectrometric measurements

For the comparative analysis, samples of animals (small passerine and murine species), soil, and plants that were already available in the IRL samples bank from other earlier studies in the ChEZ were used. The external covering (feathers and hair) and the internal organs of the animals were not removed to make sure that the measurements results would match the actual field results obtained for live animals.

For the spectrometric measurements, the animals were placed into a disposable cardboard box (one of the three dimensions depending on the animal body size: 40×35×70, 50×35×100, and 60×50×170 mm) with an upper plastic 0.1 mm thick wall. These containers had been developed for the spectrometry analysis of live animals from a pigmy shrew (*Sorex minutus*) or soprano pipistrelle bat (*Pipistrellus pygmaeus*) (3–8 g) to a yellow-necked field mouse (*Sylvaemus flavicollis*) or blackbird (*Turdus merula*) (40–90 g). Empirically, it was established that birds survive this procedure without any negative consequences if the measurements do not exceed 30–60 min, depending on the ambient temperature. Amphibians, reptilians and mammals can survive in the container for several hours. However, if rodents stay in the container for longer than an hour, they may destroy the container.

Method of radiochemical assessment of ^{90}Sr content

The ^{90}Sr concentration in the samples of mice (as well as in soils and vegetation) was measured by using the radiochemical oxalate method based on the daughter ^{90}Y isotope (Marey et al. 1980). The chemical yield for ^{90}Y was at least 65%. The sample was measured using the low background gas flow radiometer Eberline FHT-770T6. The relative uncertainty of the ^{90}Sr measurements in the samples ranged from 12 to 45% for various levels of activity.

The ^{90}Sr concentration in the samples was separated radiochemically using the selective ion exchange Sr-resin (IAEA-TECDOC-1092 1999). The ^{90}Sr activity was then measured using a liquid scintillation counter (LSC) Packard 2500TR.

Calibration standards

A standard OISN-16 source (Applied Ecology Laboratory of 'Environmental Safety Centre', Odessa, Ukraine) was used for calibrating the stationary HPGe detector. The source consisted of epoxy resin granules (< 3.0 mm), with a bulk density of 1 g cm^{-3}, and ^{152}Eu concentration of about 158 kBq kg^{-1} (8 October 2001).

Calibration of the mobile gamma/beta spectrometry system was performed using a series of six phantoms with known quantities of ^{90}Sr and ^{137}Cs. The phantoms presented packages of elliptical cross section, manufactured from plastic film (0.1 mm thickness), and having the following linear dimensions (length x smaller diameter x larger diameter, mm): (1) 6 g: 45 × 10 × 18; (2) 8 g: 47 × 11 × 20; (3) 12 g: 50 × 13 × 24; (4) 14 g: 52 × 14 × 25; (5) 16 g: 55 × 15 × 26; and (6) 20 g: 61 × 15 × 29. The packages were filled with 6, 12, 14, 16, and 20 g of the standard OISN-3 source (Applied Ecology Laboratory of 'Environmental Safety Centre',

Odessa, Ukraine), respectively. The source consisted of 0.5–1.0 mm epoxy resin granules with the density of 1 g cm^{-3}, ^{90}Sr concentration of 112 kBq kg^{-1} and ^{137}Cs concentration of 115 kBq kg^{-1} (as of 22 February 2007). The sizes selected for the phantoms reflect a typical size range for the vertebrate species that are the most frequently studied species in the ChEZ, specifically: pigmy shrews – 7.5 g on the average (range: 3–17 g); rodents (mice and voles) – 23.8 g on the average (range: 6–58 g), chiropterans – 13.8 g on the average (range: 3.4–41 g), small birds – 24 g on the average (range: 8–102 g), and amphibians (frogs and toads) - 8.9 g on the average (range: 2–32 g).

One 20 g phantom was used to perform daily checks of the gamma/beta spectrometry system. All six phantoms were used to calculate the beta particles self-absorption coefficient in objects with various linear sizes. Parameters of the exponential equation introducing adjustments for the animal body mass were then calculated. The NaI gamma detectors were checked with the same sources with no adjustments needed for self-absorption.

RESULTS AND DISCUSSION

The measurements of the total ^{137}Cs content using the mobile gamma/beta spectrometry system did not cause any difficulties. The differences between the measurement results obtained by using this mobile system and those obtained by using the stationary gamma spectrometer for the same samples ranged from 3 to 15%. This depended on the activity of the samples, the duration of the measurements, and the external gamma background in the area where the mobile laboratory was deployed. Due to the same circumstances, the minimum detectable activity (MDA) for the mobile system ranged widely, from 1 to 100 Bq per sample. For the optimal

conditions (primarily, if the measurement time was at least one hour) the MDA did not exceed 1–20 Bq per sample.

The biggest problem for the ^{137}Cs spectrometric measurements was associated with the external gamma background in the location of deployment of the mobile laboratory. In some cases, the lead 100–150 mm thick shielding did not appear sufficient. Empirically, it was determined that reliable ^{137}Cs measurements were only made possible if the radiation background in the area of the mobile laboratory was at least 3–4 times lower than in the area where the animals had been sampled. In view of this, prior to the measurements, the best location for deploying the mobile laboratory was determined and, where necessary, an additional 50 mm thick lead shielding layer was used. The highest background values at which the laboratory operated ranged from about 3–5 μSv h^{-1}.

Technical problems associated with the ^{90}Sr measurements in animals

Theoretically, the thicker the electron recording layer in the beta detector is, the higher the efficiency of this detector (and the associated efficiency of the ^{90}Sr measurements) is. However, in the presence of ^{137}Cs either in the sample or as a high external radiation background, there is a higher probability for recording both gamma rays (661 keV) and x ray radiation, thereby complicating the beta spectrum analysis. For these cases, a thinner detector is more appropriate, i.e., a detector with the thickness of the recording layer being adequate to absorb enough beta energy, but with a low probability to absorb much gamma or x ray energy. equal to the ^{90}Y beta absorption length. Figs. 3 and 4 show the beta/gamma spectra of the samples measured by the detectors with the scintillation layers of 1 mm and 0.1 mm, respectively.

The "joint peak" of gamma rays (661 keV) and the barium conversion electrons visible in the 1 mm scintillator was no longer seen if the scintillator was 0.1 mm thick. In addition, the ^{90}Sr (^{90}Y) and ^{137}Cs beta spectra acquired a more defined look and they could be separated easier. For this reason the beta detector with a 0.1 mm thick scintillator was selected for the mobile gamma/beta spectrometry system. Nevertheless, even with a 1 mm thick scintillator, the effectiveness of gamma recording (661 keV) was thirty times lower than the effectiveness of the recording of electrons of the same energy. Therefore, the measurements using different beta spectrometers provided similar results. For example, measurements of the total ^{90}Sr content in the bodies of animals (birds) showed that the 0.1 mm device agrees to within approximately 20% and 10% with the >1mm and 1 mm devices (Fig. 5).

Further processing of the experimental spectrum was conducted by comparing this spectrum with the calibration spectrum, specifically, with the spectra obtained using the same spectrometer with standard ^{137}Cs and ^{90}Sr+^{90}Y (phantoms) sources. To assure a high statistical reliability of the ^{90}Sr measurements, the calibration standards were measured for 1-2 hours daily in the field conditions and for 24 hours monthly in the stationary laboratory. The background was measured during the same time and with the same frequency.

The spectra of the calibration sources and background were described using cubic splines and were subsequently used for description of the experimental spectra. This process was implemented in the Beta+ software application as follows.

In a general case, for approximation of the experimental data $\{y_i\}$, $1 \le i \le i_0$, using the function $f(i, \mathbf{X})$ where $\mathbf{X}$ is a vector of the adjustable parameters $\{X_j\}$, $1 \le j \le j_0$ (j_0 is the quantity of the adjustable parameters), the functional has to be minimized using the least square method shown in eqn (1):

$$S(\mathbf{X}) = \sum_{i=1}^{i_0} w_i \left[f(i, \mathbf{X}) - y_i \right]^2 , \quad (1)$$

where w_i is a weight coefficient that is usually selected as equal to the reverse square of the experimental error.

To determine the functional minimum, the $\partial S/\partial X_j$ derivatives are assumed to be equal to zero and the generated system of equations is solved relative to the required X_j parameters.

The obtained $\mathbf{X}_0$ value corresponds to the minimal value of the functional $S(\mathbf{X}) = S_0$. To determine the errors of the parameters, the equation $S(X_{0k} + \delta X_k, X_{0j}) = S_0 + 1$ is solved separately for each of them relative to δX_k, i.e. to determine which deviation of the k^{th} parameter from the optimal value (if the remaining parameters are unchanged) increases the S functional value by 1 in comparison with the minimum. The error of the parameters will be equal to the results of eqn (2):

$$\Delta X_k = \delta X_k \cdot R, \quad (2)$$

where $R = \max\ [1, S_0/(i_0 - j_0)]$ is the parameter that takes into account the quality of the adjustment, i.e., «χ^2 times the degree of freedom».

The beta spectra are processed as follows.

First, the background spectrum (taking into account the live times) is deducted as shown in eqn (3) from the processed spectrum and the spectra of the calibration sources, each of which consists of i_0 channels with the N_i number of counts in the i^{th} channel, for $1 \le i \le i_0$ measured during the live time t and for $0 \le j \le 2$ where the j index corresponds to the following: 0 – to the processed spectrum, 1 – to the ^{137}Cs calibration spectrum and 2 – to the ^{90}Sr calibration spectrum:

$$y_{j,i} = N_{j,i} - t_j/t_{\text{background}} \cdot N_{\text{background},i}. \quad (3)$$

The deduction results in a "pure" ^{137}Cs and ^{90}Sr calibration spectra and spectrum of the analyzed sample. The latter contains unknown ^{137}Cs and ^{90}Sr activities and, possibly, a small fraction of other activities. To make up for this fraction, the quadratic background is added as shown in eqn (4):

$$\sum_{k=0}^{2} a_k i^k \quad . \tag{4}$$

Another adjustment is introduced due to the fact that if the equipment operates for a long time, the spectrometer gain may slightly "drift" and the energy calibration in the samples spectra and calibration spectra may differ, i.e. the energy E_0 will correspond to the i^{th}-channel in the calibration spectrum and the energy $(1+c)\cdot E_0$, with c as a minor adjustment will correspond to the i^{th}-channel in the processed spectrum. To take this effect into account, the calibration spectra are modified as follows. The i^{th} channel of the processed spectrum is assumed to correspond to the $\hat{\imath}^{th}$ channel of the calibration spectra, $\hat{\imath} = (1+c)i$. Obviously, in the general case, the $\hat{\imath}^{th}$ value is not an integer. Therefore, a linear interpolation is used to determine the number of counts of the calibration spectrum in the $\hat{\imath}^{th}$ channel as shown in eqn (5):

$$y_{\hat{\imath}} \approx y_{\hat{\imath}0} + (y_{\hat{\imath}0+1} - y_{\hat{\imath}0})\cdot(\hat{\imath} - \hat{\imath}_0), \tag{5}$$

where $\hat{\imath}_0$ is the integer part of $\hat{\imath}$.

Since the c adjustment is usually a small value, i.e., $|ci| < 1$, it can be assume that $\hat{\imath}_0 = i$ and, consequently the final value is determined by eqn (6):

$$y_{\hat{\imath}} \approx y_i + (y_{i+1} - y_i)\cdot ci. \tag{6}$$

If the channel domains from i_1 to i_2 are identified in the processed spectrum, the functional will be determined by eqn (7):

$$S(\mathbf{a},\mathbf{b},c)=\sum_{i=i_1}^{i_2} w_i\left[\sum_{j=1}^{2} b_j\cdot\left(y_{j,i}+(y_{j,i+1}-y_{j,i})\cdot c\cdot i\right)+\sum_{k=0}^{2} a_k i^k-y_{0,i}\right]^2 . \quad (7)$$

The functional minimization results in obtaining the b_j values, i.e., relative ^{137}Cs and ^{90}Sr activities. To obtain the absolute activity *A,* the b_j values have to be multiplied by the activity of the calibration sources with the live times ratio taken into account and shown in eqn (8):

$$A_j = b_j\cdot\beta_j\cdot t_0/t_j. \quad (8)$$

Although the Beta+ software application was developed for both ^{90}Sr and ^{137}Cs measurements, the use of a thin scintillation detector (0.1 mm) significantly reduced effectiveness and accuracy of the ^{137}Cs measurements. A varying external gamma background in the locations of deployment of the mobile laboratory was also an interfering factor. These two factors significantly increased the error of the ^{137}Cs measurements in comparison with a simple activity measurement based on the 661 keV gamma line. Therefore, a separate scintillation (NaI) detector included into the mobile gamma/beta spectrometry system was used for measuring ^{137}Cs in the samples.

For ^{90}Sr, the quality of the results obtained using the mobile gamma/beta spectrometry system was assessed by comparing them with the results of the standard radiochemical analyses of the same samples.

Specifically, the comparison of the measurements data for small murine rodents with the body mass ranging from 14.1 to 45.1 g and with the ^{137}Cs concentration ranging from 0.7 to 393 Bq g^{-1} showed that the beta spectrometry results differed from the radiochemical analysis results by not more thatn 25%, on the average, without a systematic deviation (Fig. 6). The absolute

domination of the total ^{137}Cs activity over the ^{90}Sr activity (by factors of 2.6-7.8) did not affect the accuracy of the measurements. However, if the ^{90}Sr activity was lower than 10 Bq g^{-1}, we observed the trend of the error increase by 16% on the average, reaching 40–60%.

A similar comparison was made for sampled birds ranging from 10.2 to 23.2 g with the ^{137}Cs concentration from 1.1 to 8.9 Bq g^{-1} in the body. A consecutive evaluation of the ^{90}Sr total content measured by the beta spectrometry and radiochemical methods showed a satisfactory correlation between the results obtained by using these two methods (Fig. 7). There was no detectable impact of ^{137}Cs on the measurements results. However, unlike the results obtained for murine rodents, the radiochemical ^{90}Sr results for birds appeared to be systematically lower than the beta spectrometry results, i.e., the beta spectrometry results were only about 50% of the radiochemical results. Such a difference might have resulted from differences in the spatial distribution of strontium-containing structures (bones) in birds in comparison with mammals. Specifically, the bones in bird extremities constitute a much larger fraction of the total mass of the skeleton than do bones in rodent extremities. In addition, if animals are considered as a geometric ellipsoid-shaped source, the bones of bird extremities are located near the upper surface of the ellipsoid being less protected (shielded) by tissues than do the bones in rodent extremities. This will increase the effectiveness of beta particle recording. Consequently, an adjustment factor was introduced that reflected a systematic difference between the beta spectrometry and radiochemical results for ^{90}Sr content measurements using the field beta spectrometer. After the adjustment factor was introduced for the given set of samples, the average deviation of the beta spectrometry results from the radiochemical results became unsystematic and equal to about 21.7% (range: 4–40%).

Apart from the fact that animals from various systematic groups may have a different topography of their skeletons affecting results of the in vivo ^{90}Sr beta spectrometry, researchers have to deal with species of various sizes in their field studies. Obviously, the ^{90}Y electrons self-absorption in bodies of small species differs from that in large species. Thorough measurements of phantoms with the known ^{90}Sr quantity showed that the effectiveness of electrons recording drastically increased if the body size decreased (Fig. 8). Since, to save time, the equipment was calibrated using the phantom of only one size (a 20 g source), an adjustment factor *k* for the body mass (*m*) was introduced for calculating ^{90}Sr activity in animals of various sizes. The adjustment factor *k* was expressed by the parameters of the following exponential eqn (9):

$$k = 0.949 + 1.24e^{-\frac{m}{6.28}}. \quad (9)$$

For this purpose, the initial value was divided by *k*.

In the process of the field measurements another technical problem was encountered associated with the use of the gamma/beta spectrometry system. The accuracy of both ^{90}Sr and ^{137}Cs measurements is directly related to the duration of the measurements. However, the duration of the measurements is limited by both the measured object (for example, a high sensitivity of birds to being measured) and a number of measurements that can be realistically performed within one work day. The total activity of radionuclides in bodies of animals may vary within the same site by factors of tens, and the ^{90}Sr to ^{137}Cs ratio from species to species may also vary significantly, occasionally changing to the directly reverse value. Consequently, the settings of the measurements parameters to assure a high accuracy of ^{90}Sr measurements do not guarantee a high accuracy of ^{137}Cs measurements and vice versa. Therefore, for each specific area and object, specific optimal requirements and assumptions have to be identified.

This method is satisfactory for animals of small sizes, however, the uncertainties associated with the skeleton topography, body size and measurements conditions remain to be addressed. Measuring a group of species with a different anatomy has to be preceded by a preliminary assessment of deviations of the ^{90}Sr beta spectrometry results from the radiochemical results. For this assessment, a small sized animal is defined as one with the body size up to 40 – 50 g. Some indirect data show that, for a certain skeleton topography, this method may provide satisfactory results for animals of up to 100 g (e.g., birds and small snakes). In large sized animals, the role of poorly monitored processes of beta particle self-absorption inside the measured species will grow, thereby making this methodology unacceptable in principle for large sized animals.

For inanimate objects (soil, vegetation), ^{90}Sr beta spectrometry of their thoroughly homogenized samples is acceptable for all sample sizes. This method will require an appropriate accounting for beta electrons self-absorption, which is a function of the sample mass and density, For this we use a HPGe spectrometer with a beryllium window, since the NaI- scintillation detector included into the gamma/beta spectrometry system cannot resolve the low energy photon to be measured. The electron self-absorption correction factor can be determined by evaluating the self-absorption of the ^{137m}Ba characteristic irradiation. About 8% of ^{137}Cs disintegrates by means of the internal conversion of the M4 gamma transition in the K-layer followed by x ray radiation K_x with the energy of 31.8–37.5 keV. Since the transmission of such radiation approximately matches the transmission of electrons with the energy of 3–4 MeV (Browne and Firestone 1986), the self-absorption of ^{90}Y beta particles will be approximately the same as the self-absorption of the characteristic x ray radiation of barium. The self-absorption coefficient for barium is easy to calculate by measuring barium K_x-radiation intensity and the

associated higher-energy gamma radiation (661 keV). Since the self-absorption of 661 keV gamma radiation is not significant for thin low density samples (Browne and Firestone 1986), the ^{90}Y electrons self-absorption coefficient is calculated from the ratio of the K_x x ray radiation intensity to the 661 keV gamma radiation intensity for "thin" standard sources and measured samples. A thin source in this context is a source for which the average electron path is significantly smaller than the thickness of the source.

The measurements of thoroughly pretreated samples of plants and soils in their standard geometry can serve as examples for illustrating the effectiveness of this methodology for measuring homogenized samples. The results of ^{90}Sr beta spectrometry measurements of plant samples differed from their radiochemical measurements results by 8.6% on the average (range: 2–12%) (Fig. 9) and the results of ^{90}Sr beta spectrometry measurements of soil samples differed from their radiochemical measurements results by 12.2% on the average (range: 0.1–34%) (Fig. 10). The ^{90}Sr specific activities of the soil samples differed by more than by three orders of magnitude: from 0.5 to 3,700 Bq g^{-1} (Fig. 10, Table 2). The density of the soil samples varied from 0.7 to 1.7 g cm^{-3} because the soil had been sampled from various depths.

CONCLUSION

The mobile gamma/beta spectrometry system has a number of technical limitations. First, it is intended for measurements of animals (and other environmental media) from the ChEZ where the ^{90}Sr to ^{137}Cs concentration ratio in the entire specimen is at least 0.02–0.05 and other "interfering" radionuclides are either present in minimum quantities or do not exist. Second, an external gamma background affects the operation of the equipment in the field, therefore, it is

desirable that the laboratory be deployed in less contaminated areas than those areas where the animals are sampled, or the lead shielding of the detectors must be made much thicker. Third, this method is intended for measuring animals of up to 40–50 g. The applicability of this method for animals of up to 100 g requires additional studies. Fourth, a body mass adjustment factor has to be introduced for the initial ^{90}Sr beta spectrometry results because the effectiveness of the beta particles recording for animals of various sizes significantly and regularly changes. Fifth, some systematic groups of animals have an essentially different skeleton topography, thereby requiring additional adjustment factors.

Nevertheless, this method and equipment provide vast opportunities for researchers in the ChEZ. The advantages of this method and equipment (rapid measurements, capability to measure live animals directly in their habitat, and the capability of simultaneous ^{90}Sr and ^{137}Cs measurements) far outweigh the existing limitations, and the accuracy of these measurements is consistent with the requirements applicable to most radioecological studies. Many studies have been conducted using this technology, specifically, radioecology of birds (Gashchak et al. 2008, 2009), murine rodents and pigmy shrews (Maklyuk et al 2007a,b), chiropterans (Gashchak et al 2010) and amphibians (Gashchak et al 2009) was studied. Without resorting to radiochemical methods, the spatial and vertical distributions of radionuclides in soils were measured in the "Red Forest", the most contaminated area of the ChEZ (Bondarkov et al. 2006). The mobile gamma/beta spectrometry system was heavily used for the dose exposure measurements of ChEZ biota (Maklyuk et al. 2008) and for the radioecological characterization of the experimental sites around the ChNPPP Cooling Pond (Oskolkov et al. present issue). After some additional modifications and calibrations, it should be possible to use this method and equipment in other radioecological conditions.

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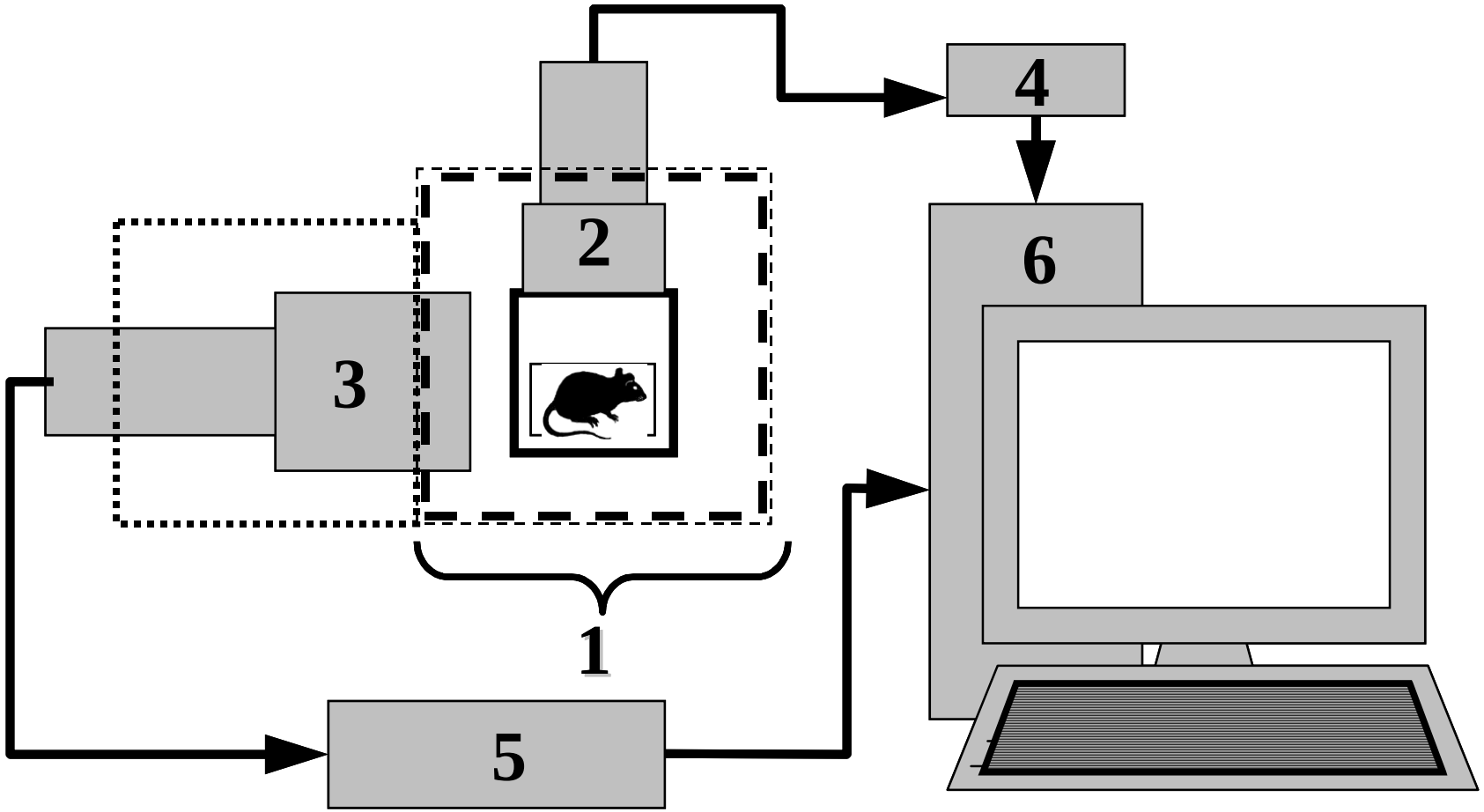

a

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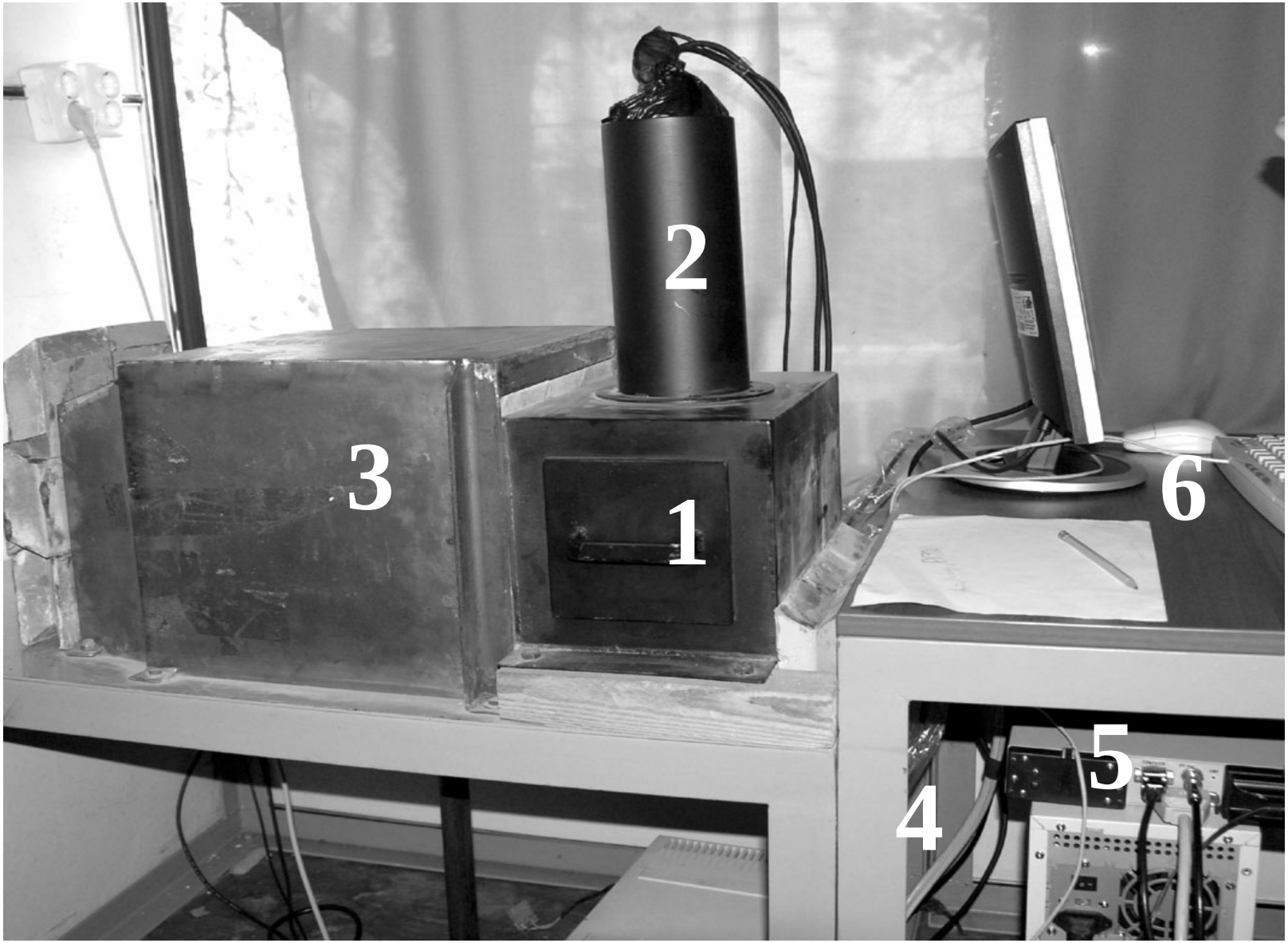

b

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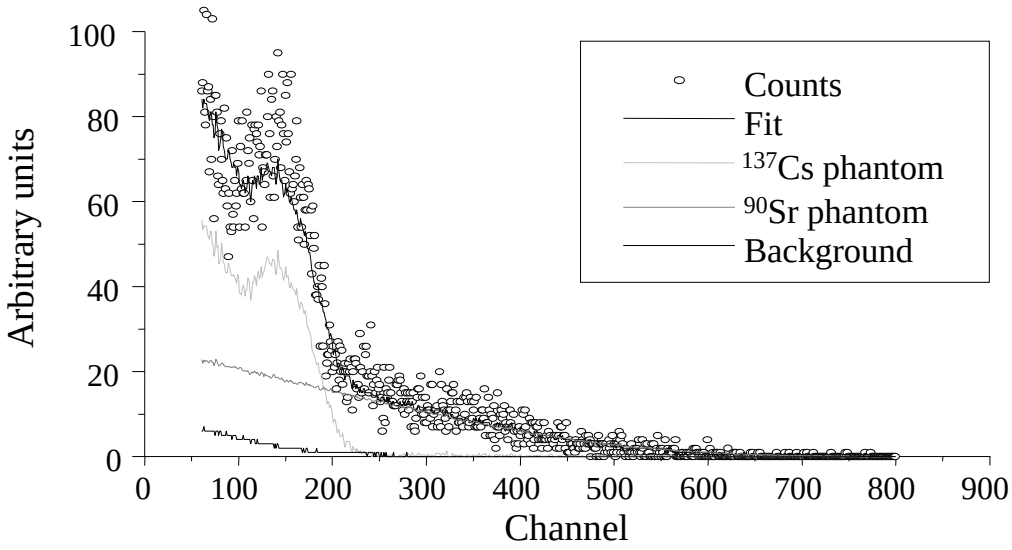
C

Fig. 1. Schematic and photographs of the mobile gamma/beta spectrometry system (c).

Fig. 2. Photograph of the mobile laboratory (bus) deployed in the field. The electric generator is in the foreground (covered).



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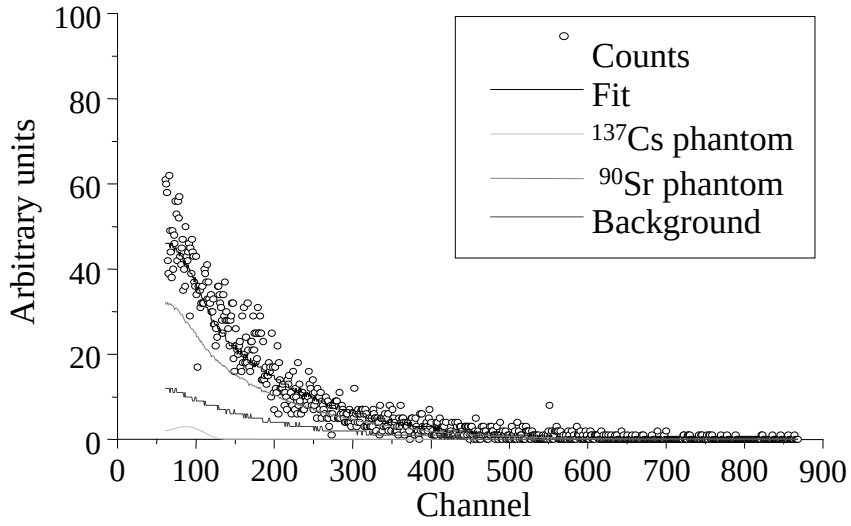
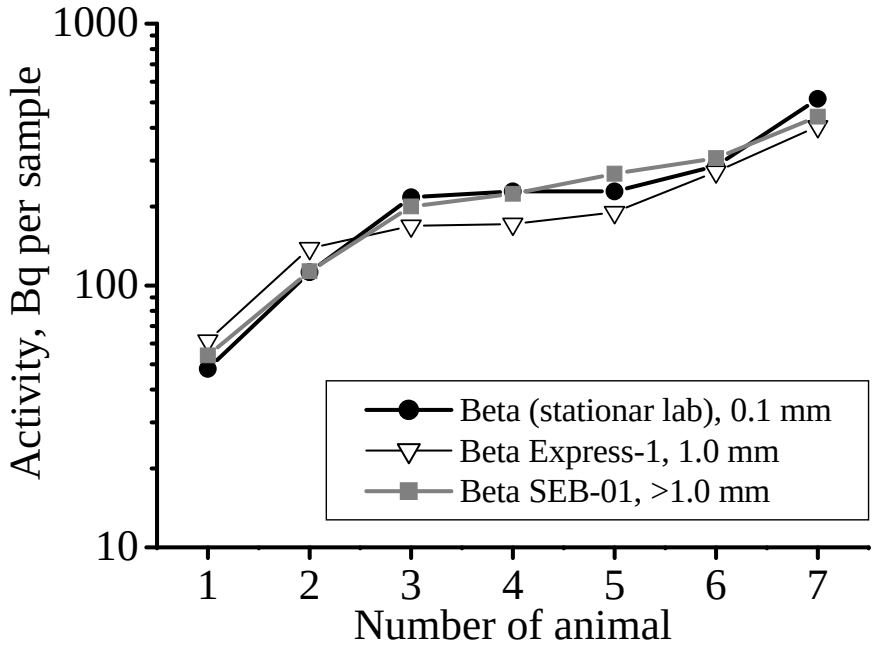


Fig. 4. Experimental beta-gamma spectrum (dots), adjustable and calibration spectra (solid line) for the detector with a 0.1 mm thick plastic scintillator.



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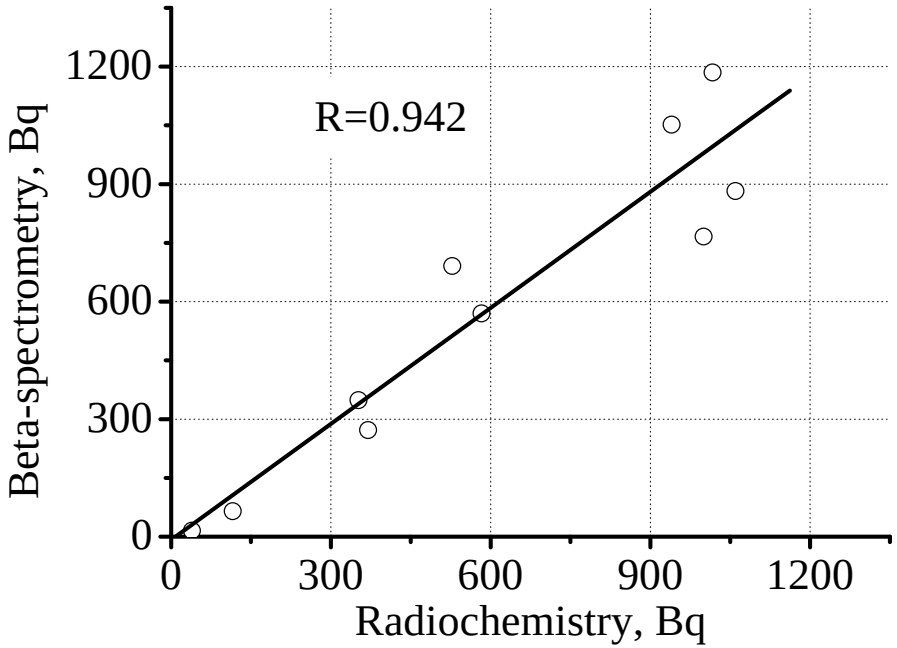


Fig. 6. ^{90}Sr activity measurements in mice (n = 10) obtained by beta spectrometry and radiochemical analysis, Bq per sample.

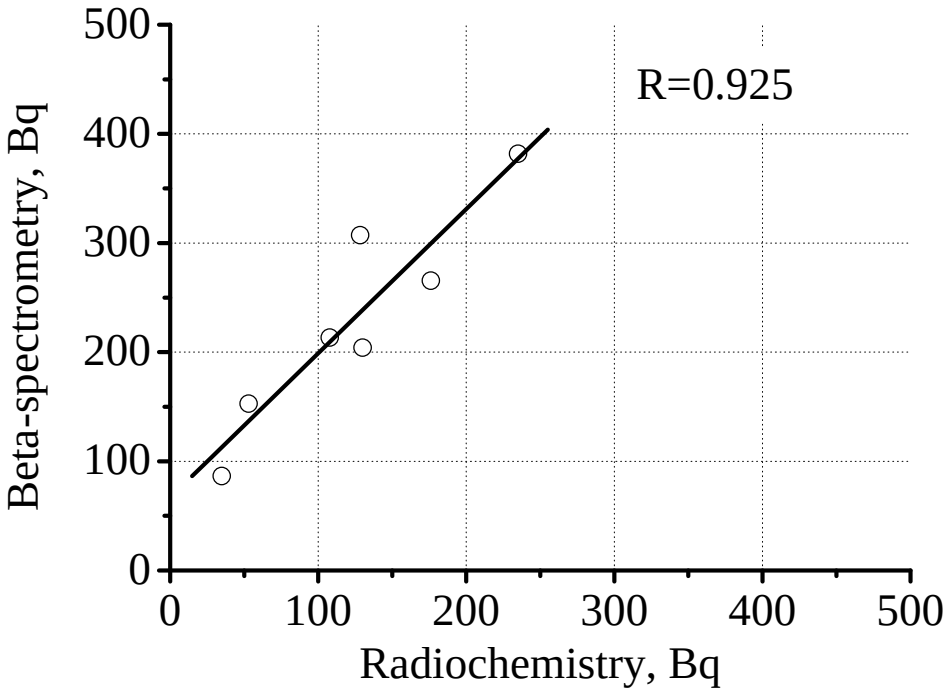
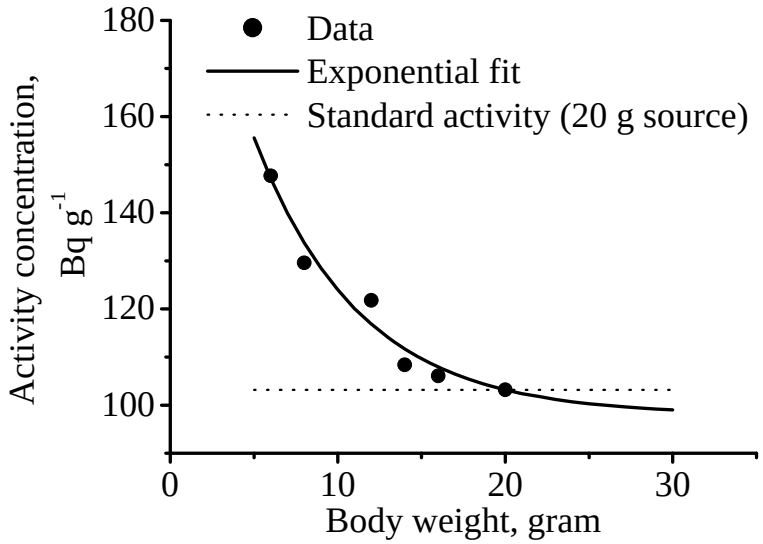


Fig. 7. Comparison of beta spectrometry and radiochemical measurements of the total ^{90}Sr content in birds (n = 7).



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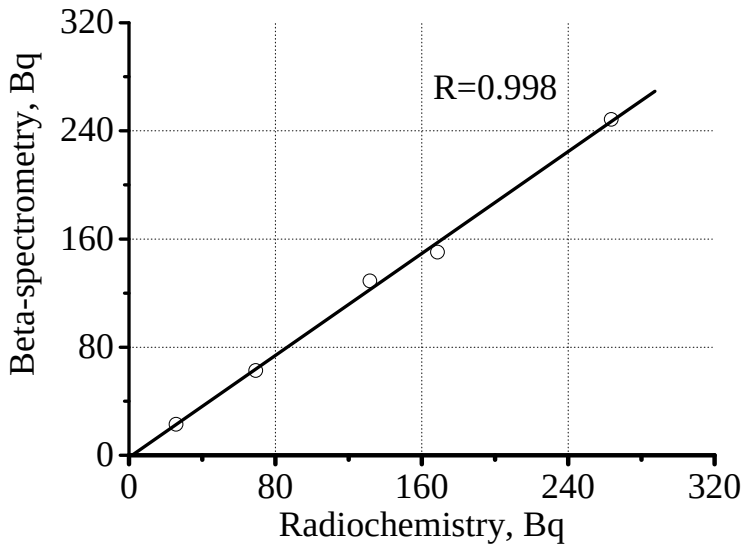

Fig. 9. Comparison of ^{90}Sr activity measurement results in plant samples (n = 5) obtained by beta spectrometry and radiochemical method, Bq per sample.

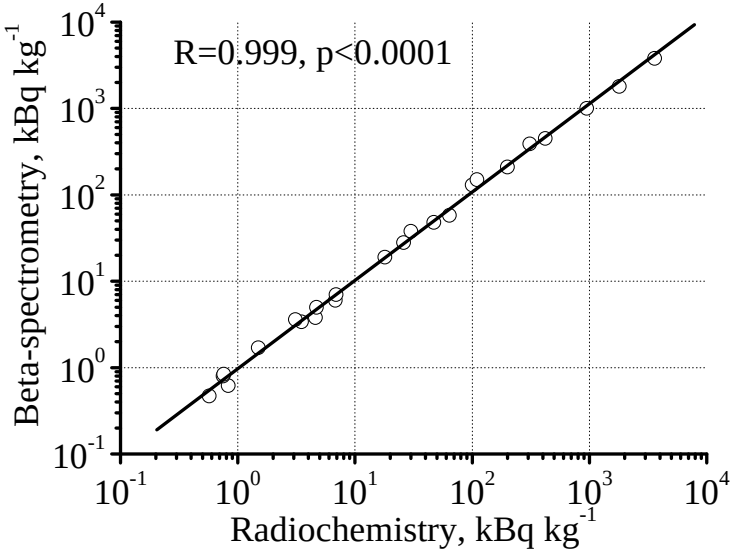

Fig. 10. Comparison of ^{90}Sr activity measurement results in soil samples (n = 24) obtained by beta spectrometry and radiochemical method, kBq kg^{-1}.

Table 1. Technical specifications of beta spectrometers.

Name of the Spectrometers	1. Beta Spectrometer	2. Beta Spectrometer	3. SEB-01	4. Express-1
Location of the spectrometer	Stationary Laboratory	Mobile Laboratory	Stationary Laboratory	Stationary Laboratory
Detector (manufacturer)	INR NASU[a]	INR NASU[a]	NPP Atom, *Prilad Komplex*	BDZhB-06P, INR NASU[a]
Thickness of the scintillator[b], mm	0.1	0.1	>1.0	1.0
Diameter of the scintillator, mm	60	60	73	65
Position of the detector in the spectrometer	Vertically, window facing down			
Type of passive shielding	Plastic box	Lead box, 5 cm	Lead box, 5 cm	Lead box, 5 cm
Multi-channel analyzer	ASA-100, CANBERRA	ASA-100, CANBERRA	ASA-100, CANBERRA	92X Spectrum Master, ORTEC
Software for the analyzer	Genie-2000, CANBERRA	Genie-2000, CANBERRA	Genie-2000, CANBERRA	ADCAM 100, ORTEC
Type of spectra processing	"Windows" method	Software application Beta+, INR NASU[a], Chernobyl Center IRL		

[a] Institute of Nuclear Research, National Academy of Sciences of Ukraine (INR NASU).
[b] All the scintillators are made of plastic.

Table 2. ^{90}Sr specific activity in soil samples measured by various methods.

Sampling location and depth, cm		Specific activity, Bq/kg		Deviation
		Radiochemistry	Spectrometry	
1 – 145	0 – 2	9.50×10^5	1.01×10^6	- 0.06
	2 – 4	3.10×10^5	3.88×10^5	- 0.20
	4 – 7	1.10×10^5	1.48×10^5	- 0.26
	7 – 10	4.70×10^4	4.75×10^4	- 0.01
	10 – 15	3.00×10^4	3.79×10^4	- 0.21
	15 – 20	6.90×10^3	7.02×10^3	- 0.02
	20 – 25	7.50×10^2	8.04×10^2	- 0.07
	25 – 30	7.60×10^2	8.36×10^2	- 0.09
2 – 145	0 – 2	1.80×10^6	1.80×10^6	0.00
	2 – 4	4.20×10^5	4.50×10^5	- 0.07
	4 – 7	1.00×10^5	1.33×10^5	- 0.25
	7 – 10	2.60×10^4	2.80×10^4	- 0.07
	10 – 15	4.70×10^3	4.96×10^3	- 0.05
	15 – 20	3.10×10^3	3.62×10^3	- 0.14
	20 – 25	3.50×10^3	3.41×10^3	0.03
	25 – 30	4.60×10^3	3.84×10^3	0.20
3 – 145	0 – 2	3.60×10^6	3.83×10^6	- 0.06
	2 – 4	2.00×10^5	2.14×10^5	- 0.07
	4 – 7	6.40×10^4	5.84×10^4	0.10
	7 – 10	1.80×10^4	1.85×10^4	- 0.03
	10 – 15	6.80×10^3	6.03×10^3	0.13
	15 – 20	1.50×10^3	1.68×10^3	- 0.11
	20 – 25	5.70×10^2	4.73×10^2	0.21
	25 – 30	8.30×10^2	6.16×10^2	0.35