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An application of *in vivo* whole body counting technique for studying strontium metabolism and internal dose reconstruction for the Techa River population

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Abstract. Nuclear techniques for direct assessment of internally deposited radionuclides are essential for monitoring and dosimetry of members of the public exposed due to radiation accidents. Selection of the methods for detection of internal contamination is determined by nuclear-physical characteristics of the deposits. For the population living in settlements located along the Techa River contaminated in 1950s by liquid radioactive wastes from the plutonium production complex Mayak (Southern Urals, Russia) the main dose-forming radionuclide was ⁹⁰Sr. It is a bone-seeking radionuclide that incorporates in the skeleton and remains there for many years so it can be detected for long periods after the intake occurred. Measurements of pure beta-emitting ⁹⁰Sr are possible through detection of bremsstrahlung from the ⁹⁰Sr/⁹⁰Y beta rays in the low energy range (30-160 keV) using phoswich detectors. This suggested the development of a unique whole body counter in 1974 for monitoring of the ⁹⁰Sr- and ¹³⁷Cs-body burden in the Techa River population with the use of phoswich detectors. Long-term observations with the WBC covering more than 38,000 measurements on over 20,000 people have been made. This has created a unique database for studying strontium and calcium metabolism and for assessment of the internal dose for residents of the Techa River settlements due to ingestion of 90Sr. This paper describes the main results obtained for the Techa River population essential for bone metabolism and dosimetry, epidemiological studies, and radiation protection.

1. Introduction

Internal exposure of the population occurs as a result of drinking water, eating food and breathing air, containing radioactive substances released to or naturally occurring in the environment. Global environmental contamination by radioactive products resulted from nuclear weapons testing in 1945–1980. Local contamination by radioactive substances results from nuclear plants and installations or other human activities involving radioactive sources. Population exposure in the Urals (Russia) in addition to global fallout from nuclear weapons testing resulted from the operation of the Mayak Production Association, which was the first Russian site for the production and separation of weapons grade plutonium. One of the most essential population exposures occurred through releases of liquid radioactive effluent into the rather small Techa River in 1949-1956. The river was the main source of drinking water for residents of riverside communities; therefore, this resulted in chronic internal and also external exposure of about 30,000 persons. The major intake of radionuclides by inhabitants of the area occurred in 1950–1951, of which ⁹⁰Sr mainly contributed to the internal dose. ⁹⁰Sr is known to be a bone-seeking radionuclide that incorporates in the skeleton and retains there for many years

irradiating radiosensitive cells of the red bone marrow and those adjacent to bone surfaces. This may, in turn, induce leukaemia and bone sarcomas. On average, the residents of the upper- and mid-Techa regions ingested about 3,000 kBq of ⁹⁰Sr with river water, which was more than two orders magnitude higher than the worldwide level of radionuclide intake due to global fallout from nuclear weapon tests [2]. The "Extended Techa River Cohort" (ETRC) has been studied for several decades by scientists from the Urals Research Centre for Radiation Medicine (URCRM) [3, 4]. A special database was established for follow-up of the exposed population. This database contains a roster of the exposed persons, their residential history, and results of medical and dosimetric examinations. One of the primary features of the long-term dosimetric study is a unique set of measurements on ⁹⁰Sr in the human body, including measurements of this radionuclide in bones, teeth and the whole body for a period of more than 45 years [5, 6].

Since the early 1950s, different methods have been used at the URCRM for the measurement of 90 Sr-body burdens. The first involved radiometric measurements of bioassay and autopsy samples. From 1951 to 1960 ashes samples were measured using a gas-flow counter calibrated with a 90 Sr- 90 Y standard [7]. The average- 90 Sr concentration in the skeleton was calculated on the basis of measuring several samples of trabecular and compact bone. Since 1958, a radiochemical method (based on coprecipitation of 90 Sr with nitrates) has been used [7]. *Post mortem* measurements could not involve many people for the monitoring program and, since 1974, *in vivo* measurements of 90 Sr-body burden were also conducted with the unique URCRM whole body counter (WBC) named SICH-9.1 [6]. The main goal of this WBC was to allow *in vivo* measurements of 90 Sr for the members of the ETRC. Long-term observation of the exposed population with the WBC has created a unique database to study strontium metabolism during periods long after the major intake occurred and bone metabolism by considering strontium as a tracer for the metabolism of bone. The results of these studies also serve as a basis for the development of metabolic models of strontium in humans and the estimation of doses due to ingested 90 Sr for the Techa River population.

2. Description of the SICH-9.1 whole-body counter

This unique measurement system was designed by a group of scientists headed by Prof. Yu. Belle, specifically for the exposure situation on the Techa River [6, 8]. Its principal features are illustrated in figure 1. Quantification of ⁹⁰Sr is achieved by measuring the bremsstrahlung from the ⁹⁰Y-beta rays with a phoswich detector; for this purpose a scanning-bed geometry enclosed in a special shielded room is used. The shielded room is made of cast-iron rings with a wall thickness of 200 mm. The inner surface has linings of lead (5 mm), cadmium (1 mm), and copper (2 mm). Four phoswich detectors fixed in the central vertical plane are used. These detectors consist of two layers: a thin CsI crystal and a thick NaI crystal (figure 1). During measurement a supine person is moved through the detector array over a scanning length of 2 m. The motion is controlled by signals from the analyzer's real-time clock. The standard routine counting time is 20 min.

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Gamma-emitting radionuclides, such as ⁴⁰K and ¹³⁷Cs, also produce a "Compton continuum" of scattered radiation that includes a significant amount of low energy photons. Therefore, it is necessary to measure also ⁴⁰K and ¹³⁷Cs, so that their contribution to the low-energy bremsstrahlung region can be subtracted. Analyses of ¹³⁷Cs and ⁴⁰K are accomplished at the same time with the same detector by measurement of their photopeaks within the energy ranges of 620–740 keV and 1400–1580 keV, respectively. The spectrum of bremsstrahlung radiation registered in the low-energy region of 30–160 keV is obtained for individuals after subtraction of the contribution of ⁴⁰K and ¹³⁷Cs scattered radiation in this energy region.

Initial calibration of the system with respect to bremsstrahlung was performed in 1973 by use of two natural adult human skeletons with known activity of ⁹⁰Sr uniformly distributed in its skeletal mass, paraffin simulating soft tissues and dry paper simulating lungs. The calibration coefficient for ⁹⁰Sr was obtained [6]. Water-filled phantoms made of plastic tanks simulating a human body of different height and weight were used for calibration of ¹³⁷Cs and ⁴⁰K. Calibration coefficients based on a person's weight and height were derived [6]. Repeated calibration with an anthropomorphic

phantom of an adult person (FST-06T) with tissue substitutes adequately simulating the attenuation properties of human tissues with uniformly distributed ⁹⁰Sr in the skeleton, was performed in 1998 [9]. A solid whole-body phantom set (UP-02T set) made of polyethylene blocks and rod radionuclide sources of ¹³⁷Cs and ⁴⁰K inserted into them was used for calibration for these radionuclides. This phantom set simulates the body characteristics of children and adults of different weights [10]. In general, the calibration factors obtained in 1973 and in 1998 agreed well—the more important ones were within a few percent and all factors agreed within 16%. This confirmed the reliability of the multiple WBC measurements.

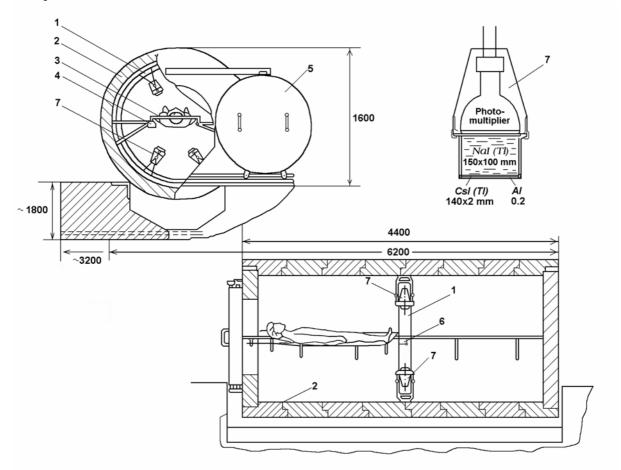


Figure 1. A schematic sectional view of the SICH-9.1 whole body counter: 1 - ring for mounting the detectors; 2 - shielding, 200-mm cast iron and linings of lead (5 mm), cadmium (1 mm) and copper (2 mm); 3 - movable bed; 4 - brackets for mounting the rails; 5 - door; 6 - motors; and 7 - phoswich detectors (all measurements in mm).

Figure 2 illustrates the spectra of the measured radionuclides in adult phantoms in the low energy range from a CsI(Tl) crystal (left panel) and in the high energy range from a NaI(Tl) crystal (right panel). The bremsstrahlung spectrum of 90 Sr/ 90 Y in low energy range in adult anthropomorphic phantom FST-06T [9], shown in figure 2, is given per 10 kBq of 90 Sr in the phantom. The spectra of gamma-emitting radionuclides, 40 K and 137 Cs, in adult F4 phantom of UP-02T set [10], shown in figure 2, are given per 1 kBq of 137 Cs and activity of 40 K corresponding to 120 g of natural K respectively.

The results of measuring people with the WBC are the following: ¹³⁷Cs activity in the whole body (Bq) obtained by measuring its photopeak in the energy range 620–740 keV; Potassium in the whole

body (g) obtained by measuring the photopeak of its natural isotope 40 K in the energy range 1400–1580 keV; and count of bremsstrahlung radiation in the low-energy region 30–160 keV after subtracting the contribution of 40 K and 137 Cs scattered radiation into this energy region, and normalized according to the calibration coefficient for 90 Sr in the phantom (Bq).

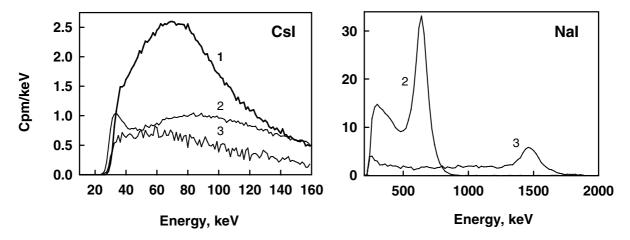


Figure 2. Bremsstrahlung spectrum of 90 Sr/ 90 Y in FST-06T phantom (1) and the spectra of scattered radiation of 137 Cs (2) and of 40 K (3) in UP-02T-F4 phantom in the low energy range (left) and the photopeak of 137 Cs (2) and of 40 K (3) in the high energy range (right). Spectrum of 90 Sr/ 90 Y is scaled per 10 kBq of 90 Sr in the phantom and the spectra of 137 Cs and 40 K are scaled per 1 kBq of 137 Cs and 120 g of natural K respectively.

More than 38,000 measurements have been carried out on more than 20,000 people over a period of 25 years. All measurements were made by the WBC group under head of V. Kozheurov [6].

The measurements of ⁹⁰Sr in humans obtained over several decades using the SICH-9.1 WBC have been analyzed recently in order to evaluate factors influencing ⁹⁰Sr-body burden measurements and to reevaluate the uncertainties associated with these data and the sensitivity of the whole body counting technique using the corrected and updated data base on ⁹⁰Sr in humans [11]. Statistical analysis has been applied to repeated WBC ⁹⁰Sr-body burden measurements obtained within a very short interval for people exposed to different levels of ⁹⁰Sr intake on the Techa River in order to estimate reproducibility errors. The detection limits for the WBC have been derived using Bayes' rule applied to *a posteriori* data. The minimal detectable activity that can be measured with the whole body counting technique is 2 kBq and the uncertainty of the single measurements of ⁹⁰Sr in body is 1.6 kBq [11]. This analysis has led to a more accurate evaluation of ⁹⁰Sr-body burden.

3. Studying of strontium metabolism in humans based on WBC data

This large unique database on ⁹⁰Sr content in humans allows the investigation of strontium metabolism in human body over long periods after the intake occurred.

3.1. Age- and gender-dependent features in strontium retention

Analysis of the WBC data was performed for permanent residents of Muslyumovo village who lived there at least during the period of major ⁹⁰Sr intake. Muslyumovo is a non-evacuated village located in the middle Techa region. Almost all residents of the village used river water as a source of drinking water, therefore, it should be expected that the intake of ⁹⁰Sr among those persons was relatively homogenous. WBC data obtained for these people 30 years after the intake are shown in figure 3.

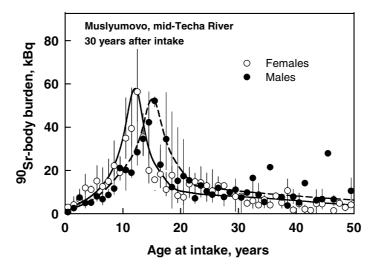


Figure 3. Median values and quartile ranges of 90 Sr-body burden for women (n=365) and men (n=305) obtained with WBC measurements for permanent residents of Muslyumovo for the time period of 30 years after intake. Lines show the predictions in 90 Sr-body burden obtained with the age-and gender-biokinetic model [12] for women (solid line) and men (dashed line) and will be described in subsection 3.3.

It is seen in figure 3, that ⁹⁰Sr retention in the human skeleton is dependent on the age and gender of the person and these dependencies remain strongly pronounced even 25–30 years after the onset of intake. The peak in ⁹⁰Sr content is observed for people who were adolescents at the time of major intake of the radionuclide. For them, the period of ⁹⁰Sr intake coincided with increased skeletal growth during pubertal development, which resulted in an increased accumulation of ⁹⁰Sr due to its similarity with the physical and chemical properties of calcium ion. These data are in agreement with experimental measurements of calcium accretion rates using Dual Energy X-ray Absorptiometry (DEXA) showing a substantial increase in early puberty [13, 14]. Taking into account that, for the Techa River residents, the average age of menarche for girls in the 1950s was 15.3 years, a sharp increase in strontium retention is observed before menarche [15]. It is also seen in figure 3, that there is a delay in peak ⁹⁰Sr retention of 2-3 years in boys compared to girls because boys have a later onset of puberty. Also, as the pubertal growth spurt in boys lasts for four years rather than three years for girls [16], a wider peak in ⁹⁰Sr content in boys is observed.

3.2. Age- and gender-dependent features in strontium elimination

Many persons exposed at the Techa River have been examined with the WBC for two or more times over long periods of time. These unique results allow studying the process of strontium elimination from the skeleton, which occurs at the late period after the intake. Previous analysis of repeated measurements has shown that strontium elimination from a human body at the late period (25-45 years) after major intake is described by exponential function [6]. Therefore, by fitting an exponential function to repeated WBC measurements obtained for the same person during a long period of time, the individual rate of ⁹⁰Sr diminution (due to radioactive decay and biological elimination) has been estimated. Figure 4 shows typical examples of the assessment of individual strontium-elimination rates (hereinafter the term "elimination rate" is used to indicate the process of *biological* elimination) for two persons selected from the ETRC; they are 1) a man (IC 11830) born in 1939, who was examined with the WBC during 1977–1995, and 2) a woman (IC 63811) born in 1940, who was measured with the WBC over 20-y period from 1977 to 1997. Figure 4 shows good agreement

between the fitted exponential functions and the repeated WBC measurements obtained for these persons.

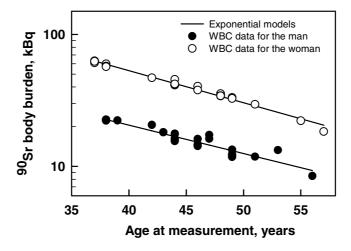


Figure 4. Examples of assessment of individual strontium-elimination rates for the man (IC11830, born in 1939, individual elimination rate is estimated as 2.57% per year) and for the woman (IC63811, born in 1940, individual elimination rate is estimated as 3.32% per year)

The analysis was performed for persons who were measured twelve or more times over a period of more than 5 years. For most of them the rate of strontium elimination independent of age has been estimated [17].

To study the dependencies in the strontium-elimination rate according to age and gender, data from all individuals in the study were grouped into six age categories and analyzed. Figure 5 presents the comparison of the age-dependent changes in strontium-elimination rates for males and females.

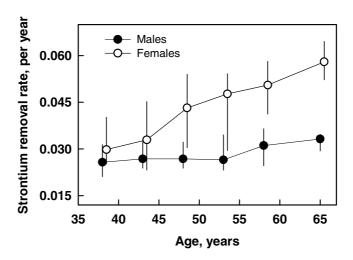


Figure 5. Age-dependent change in median values of strontium-elimination rate for men and women at long periods after the intake occurred. The bars indicate values of quartile ranges.

It is seen in figure 5, that in women there is a sharp increase in the rate of strontium elimination after the age of 45 years. This is explained by the fact that at such long periods after the major 90 Sr intake (which occurred 25-45 years before WBC measurements commenced) the only way of

strontium elimination from the human skeleton is bone resorption [17]. The results of bone metabolism studies using other methods have shown that in pre-menopausal and especially in post-menopausal women the rate of bone resorption, resulting into removal of minerals from bone, increases due to hormonal changes [18, 19]. Taking into account that the average age of menopause in studied population is 50 years [15] a sharp increase in strontium-elimination rate is observed in the same age period (45–55 years, figure 5). The same, but less pronounced patterns, are observed in men after the age of 55 years. These results are in agreement with other studies of bone resorption in men [20]. Distinct differences in age-dependent rates are observed between men and women that continuously increase after the age of 45 years, and for persons aged 60 years the value of the strontium-elimination rate for women is higher than that for men by nearly a factor of 2.

3.3. Development of the biokinetic model for strontium with allowances made for age and gender of a person

Observed features of strontium metabolism have created a basis for the development of mathematical models using WBC data for the Techa River population [12, 21]. Recently, a biokinetic model for strontium in the human body was created [which is hereafter referred to as Techa Biokinetic Model (TBM)] with model parameters specifically evaluated for the Techa River population. This model describes strontium metabolism in the whole body with the use of a system of linear differential equations of the first order. The age- and gender dependent parameters have been obtained using the unique data set on 90 Sr-body burdens for Techa River residents as well as data on global 90 Sr in human bones obtained in the UK and the USA, data on whole body mineral content, and published data on bone metabolism. The TBM was validated using different sets of data on strontium retention in humans and was shown to satisfactorily describe strontium retention for different kinds of intake [12]. Figure 3 shows predictions of the TBM model compared with measured 90 Sr-body burdens for male and female residents of Muslyumovo settlement 30 years after the intake commenced.

As seen from figure 3, the TBM model satisfactorily describes age- and gender-features in strontium retention and can be applied for the correct dose assessment necessary for epidemiological studies of the Techa River population.

4. Conclusions

This paper presents the results of long-term application of the whole body counting technique for *in vivo* measurements of ⁹⁰Sr in residents of the Techa Riverside settlements exposed as a result of radioactive effluents into the Techa River. WBC measurements acquired during long-term monitoring have created a unique database, which allow the study of features in strontium and bone metabolism and development of mathematical models describing strontium retention in human body.

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