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PROBLEM ON ESTIMATION OF THE CONTENT OF ^{131}I IN MILK IN THE "IODINE" PERIOD OF THE CHERNOBYL ACCIDENT

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Measurements of the beta-activity of milk, serving as the main source of information on the radioactive contamination of the environment by the iodine isotope ^{131}I, carried out on a DP-100 radiometer in the early post-Chernobyl period (1986) in Belarus, have been mathematically simulated. The results obtained allow the conclusion that the indicated measurements should be analyzed again with consideration for all of the nuclides present in milk.

Introduction. Radioactive iodine isotopes, mainly ^{131}I, were the main sources of the radioactive contamination of the environment in the first months after the accident at the Chernobyl atomic power plant ("iodine" period). At the time of the Chernobyl accident, cattle pastured in the majority of the Belarusian regions suffered to the greatest extent from the Chernobyl accident. The radionuclides fallen-out, in particular ^{131}I, began to enter the human organism through the food chain, predominantly with milk and milk products. At that time, in the Republic of Belarus, the control of the content of radioactive substances in agricultural products, especially in milk, was organized at the places of their production, processing, and realization.

The majority of measurements were carried out with the use of a DP-100 beta radiometer that makes it possible to determine the total beta-activity of a sample. Since the standard procedure of measurements on the indicated radiometer [1] had been developed for analysis of a definite mixture of radionuclides with a mean energy of 0.3 MeV, the processing of the data obtained with the use of a DP-100 radiometer in 1986 presented certain difficulties. To estimate the content of ^{131}I in a milk sample by its total beta activity, it is necessary to know the content of all the radioactive isotopes contained in this sample at the instant it was measured and the factors of scaling (CF) of the rate of counting (pulses/min) of these isotopes to their specific activity (Bq/kg). The scaling factors ((Bq/kg)/(pulses/min)) for the isotopes ^{131}I, ^{137}Cs, ^{134}Cs, ^{90}Sr, ^{144}Ce, and ^{106}Ru (we will call them the base isotopes) were experimentally determined at the Moscow Institute of Biophysics [2] with the use of standard solutions of these isotopes. These scaling factors were repeatedly verified in the process of measurements carried out in the early period after the accident and in the process of subsequent revisions of the data obtained.

However, the scaling factors of a number of radionuclides cannot be determined experimentally. For example, it is impossible to prepare a standard solution for the isotope ^{89}Sr. The scaling factor of this isotope was determined in [2] by interpolation of the mean energy of its beta spectrum and the scaling factors of other isotopes. However, this approach is not exact, which led to the appearance of a large error in determining the content of ^{131}I. Moreover, since the scaling factors were determined only for the above-indicated base nuclides, the contribution of other nuclides (^{133}I, ^{136}Cs, ^{103}Ru, ^{140}Ba, ^{141}Ce) to the total beta-activity of milk was not considered. A sophisticated treatment of errors in determining the scaling factors of radionuclides in the process of measurement of the beta activity of milk has been done in [3]. The errors in experimentally determining the scaling factors for the base isotopes were, respectively, 21, 39, 22, 25, 24, and 74% [3]. The errors in the data on the radioactive contamination of

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TABLE 1. Coefficients of Interpolation of the Response of a DP-100 Radiometer to a Monoenergetic Source of Electrons

Geometry of measurements	A_1	A_2	E_0	a	χ^2/DF*)
1	–0.00071±0.00014	0.14226±0.01165	2.046±0.200	1.666±0.064	1.054
2	–0.00047±0.00008	0.14373±0.02800	3.297±0.68	1.568±0.073	1.26
3	–0.00034±0.00006	0.09565±0.02530	3.6091±0.01	1.519±0.086	0.612

*) χ^2/DF is the statistical criterion χ^2 for determining the error normalized to the number of degrees of freedom DF.

food products substantially influence the estimation of the dose of irradiation of people. In this connection, the minimization of the errors in determining the content of ^{131}I in milk is of importance for correct estimation of the accident of the Chernobyl consequences.

Mathematical Model of a DP-100 Radiometer. The DP-100 beta radiometer is designed for measuring the beta activity of a substance. It consists of a lead shield, an MST-17 end-window Geiger counter serving as a detector, and an electronic unit. The procedure of simulation of the instrumental measurement of an ionizing radiation on a DP-100 radiometer with the use of the Monte Carlo method involves a detailed geometric description of all the physically significant regions (source, detector, environment) with consideration of the required characteristics of the substance (its density, the content of chemical elements and/or isotopes). The electronic unit is not essential for the transfer of radiation from the source to the detector; therefore, it is not included in the geometry of the model. The MST-17 Geiger counter was simulated with account for its actual dimensions.

The beta-activity of a milk sample was measured in the three geometries [4] (Table 1) characterized by the distance L between the surface of the liquid and the surface of the beta-counter, the inner diameter d of a cuvette, and its height h: 1) L = 10 mm, d = 26 mm, h = 10 mm; 2) L = 10 mm, d = 40 mm, h = 10 mm; 3) L = 20 mm, d = 40 mm, h = 10 mm. The measurement procedure involved two successive measurements. Initially, the background (the empty cuvette) and than the cuvette filled with liquid to the brim were measured. The total specific activity A of the sample was estimated by the simple formula

$$A = \mathrm{CF}\,(P_\mathrm{m} - P_\mathrm{b})\,. \tag{1}$$

The scaling factors determined in [1] for the measurement geometries 1, 2, and 3 are equal, respectively, to 0.48, 0.48, and 1.0. The total beta activity of milk, estimated in 1986 by formula (1) with the use of the officially recommended scaling factors $\mathrm{CF}_{\mathrm{off}}$, was repeatedly refined [3, 5–7] for the purpose of reestimation of the iodine content in milk by the results of measurements of the specific activity of ^{131}I in it.

It should be noted that, in the case where the beta-activity of a substance is measured on a DP-100 radiometer, there exist two sources of electrons: the first source (main) is formed by the beta-particles arising as a result of the nuclear decay, and the second source is formed by the Compton electrons appearing as a result of the scattering of the gamma-radiation, emitted in the process of nuclear decay, in the medium. The simulation was carried out on the assumption that the Geiger counter operates in the counting regime and any electron, beta-particle, or Compton electron, entering the sensitive volume of the counter, activates it with a probability of 100%. Even though the sensitive region of the counter adjacent to the cathode occupies a small part of the counter volume, only it, in fact, is a detector. According to the certificate of the counter, the length of its sensitive region is 25 mm, while the total length of the counter is 70 mm. The source of electrons (as well as photons if gamma-radiation appears as a result of the nuclear decay) was distributed homogeneously over the volume of the cuvette, and the direction of escape of particles was always isotropic. In the process of simulation, we followed the whole path of electrons (photons), from the point of their escape to the lead shield absorbing them or to the other elements of the device or the entrance to the active volume of the counter, where they activate the counter.

As a result of simulation of the detection of electrons by a DP-100 radiometer, we obtained the response function (or, simply, the response) η of the device. For a monoenergetic source with an energy E, this function has the form

$$\eta(E) = \frac{N_c}{N_h(E)}. \quad (2)$$

From relations (1) and (2) follows the relation between the scaling factor CF(E) and the response $\eta(E)$ to the monoenergetic source with an energy E:

$$CF(E) = 1/(60M\eta(E)), \quad (3)$$

where 60 is the time-dimension factor, sec/min. The relation between the scaling factor CF and the response η of the device to a source with a complex spectral composition has an analogous form. If $p(E_i)$ is the relative intensity of particles with an energy E_i in the spectrum of a nuclide (yield of the line E_i), the response of the device to the nuclide represents the corresponding sum of partial responses

$$\eta = \sum_i p(E_i)\,\eta(E_i), \quad (4)$$

and the scaling factor for it is determined, in accordance with (3) and (4), from the relation

$$1/CF = 60M\eta = \sum_i p(E_i)/CF(E_i). \quad (5)$$

Formulas (3)–(5) define the contribution of the beta-particles, arising as a result of the nuclear decay, and the Compton electrons to the response of the device and its scaling factor. For the nuclides representing the sources of beta-radiation, the above-indicated sums obtained with respect to the spectral lines should be considered as the integrals with respect to the beta-spectrum of the nuclides.

The Response of a DP-100 Radiometer to Monoenergetic Electrons. The response of a DP-100 radiometer to a concrete isotope was determined using the MCNP program of simulation of the stochastic transfer of radiometer radioactive radiation in different media [8] on the basis of the beta-spectrum of this isotope, obtained by generation of electrons in the program, or by calculation of the responses of the radiometer to monoenergetic sources and subsequent convolution of these responses with the beta-spectrum of the indicated isotope (analogous procedures are performed for gamma-radiation). The response of the DP-100 radiometer, calculated as an energy function for the three above-indicated geometries (Table 1), is described adequately by the logistic function

$$\eta(E) = A_2 + \frac{A_1 - A_2}{1 + (E/E_0)^a}. \quad (6)$$

For the supposed macroscopic process of photon and electron transfer in the medium, it may be assumed that the initial energy distribution of electrons in the source corresponds to the beta-spectrum of a nuclide in vacuum (the so-called "vacuum" beta-spectra). The nuclide spectra of beta-emitters in vacuum were taken from [9, 10]. The efficiency of the counting (or its reciprocal equal to the CF) can be easily determined by convolution of the interpolation curves (6) for the corresponding geometry and the beta-spectrum of the decay $p(E)$ of the nuclides considered, including the daughter nuclides [9, 10]:

$$1/CF = 60M \int_{E_{min}}^{E_{max}} p(E)\,\eta(E)\,dE. \quad (7)$$

Comparison of the Calculation and Experimental Data. The scaling factors calculated for the three above-indicated geometries were compared with the experimental data of [3] (Tables 2 and 3). The confidence limits were obtained in accordance with [3] on the assumption that the absolute error in determining the scaling factors comprises 02–0.3. Within the limits of this error, the results of simulation agree with the experimental results. The one exception

TABLE 2. Theoretical Scaling Factors Determined for the DP-100 Radiometer by the Monte Carlo Method

Geometry of measurements	Scaling factor CF						
	^{131}I	^{134}Cs	^{137}Cs	^{89}Sr	^{90}Sr	^{144}Ce	^{106}Ru
1	1.72	1.6	1.25	0.26	0.26	0.19	0.16
2	1.096	1.02	0.73	0.14	0.14	0.089	0.096
3	1.8	1.6	0.93	0.23	0.24	0.18	0.16

TABLE 3. Comparison of the Theoretical (T) and Experimental (E) Values of the Scaling Factors Determined for the DP-100 Radiometer According to [3]

Isotope	Scaling factor CF for a measurement geometry						Error in the experimental estimation of CF, %
	1		2		3		
	T|E	(T–E)/E, %	T|E	(T–E)/E, %	T|E	(T–E)/E, %	
^{131}I	1.72|1.9	–9.5	1.096|1.2	–8.7	1.8|1.8	0	21
^{137}Cs	1.25|1.3	–3.8	0.73|0.65	12.3	0.93|0.94	–1.1	39
^{134}Cs	1.6|1.4	14.3	1.02|0.84	21.4	1.6|1.2	33.3	22
^{90}Sr	0.26|0.36	–27.8	0.14|0.20	–30	0.24|0.3	–20	25
^{144}Ce	0.19|0.15	26.7	0.089|0.083	7.2	0.18|0.13	38.5	24
^{106}Ru	0.16|0.14	14.3	0.096|0.11	–12.7	0.16|0.15	6.7	74
^{89}Sr	0.26|0.67	–61.1	0.14|0.39	–64.1	0.23|0.58	–60	—

is represented by ^{89}Sr, for which the calculation result is explicitly beyond the confidence limit. Savkin of [3] notes that the value of CF for ^{89}Sr was obtained by interpolation of the scaling factors of other radionuclides with the use of the exponential of the mean energy of the beta-spectrum. In this case, the scatter of the initial points in the interpolation in the neighborhood of one and the same energy could reach 200%. We believe that this interpolation is not exactly correct. According to the results of simulation (6), the contribution of an electron to the response of the DP-100 radiometer increases with increase in the electron energy. Therefore, the low-energy electrons make a small contribution to this response; however, this fact was not taken into account in [3] in deciding on the argument of the interpolation function. Undoubtedly, in this case, a more suitable argument for the interpolation would be a variable obtained by averaging of the energy with a weight equal to the product of the spectrum at a given point and the maximum depth of the layer in a sample from which electrons can still escape and reach the detector. However, this argument cannot be determined experimentally. For this reason, more correct values of CF can be obtained by simulation with the use of available fairly exact

a) vacuum beta-decay spectra obtained using the Fermi theory taking in account the interactions in the finite state;

b) cross sections of interaction of electrons and gamma-quanta with a substance;

c) mathematical models of the source, the detector, and their surroundings.

The satisfactory agreement between the theoretical and experimental results obtained for the nuclides for which the scaling factors were reliably determined in experiments (^{131}I, ^{137}Cs) supports the correctness of the mathematical model proposed. The best agreement between the experimental and theoretical data was obtained for ^{137}Cs. This is explained, first of all, by the fact that the activity of this isotope can be reliably measured by the gamma-spectrometry method.

It should be noted that the Compton electrons, which are due to the gamma-radiation of the gamma-emitting nuclides (all the nuclides presented in Table 3, except ^{90}Sr), contribute to the scaling factor; this contribution can reach 23–27%.

Estimation of the Error in Measuring the Content of ^{131}I in Milk by a DP-100 Radiometer That Is Due to the Error in Determining the Scaling Factor for ^{89}Sr. The initial information for analysis of the measurement data on the content of ^{131}I in milk is the total beta activity of milk samples measured on a DP-100 in April–June 1986. It is precisely this value that was written in the working registers. To determine the iodine activity in milk by these data, it is necessary to obtain additional information. For this purpose, two analytical methods can be used —

the additive method and the multiplicative one — the names of which correspond to the approaches used in them for estimation of the contribution of individual nuclides to the total activity of a sample.

It is assumed in the additive method that the main contribution to the beta-activity of milk in the "iodine" period was made by the known nuclides ^{131}I, ^{134}Cs, ^{137}Cs, ^{89}Sr, ^{90}Sr, ^{144}Ce, and ^{106}Ru. If the content of each nuclide in a sample a_i (i = 1–7), determined relative to the content of another nuclide by any reasonings, is known, it is easily shown that the specific activity of the isotope ^{131}I in milk is equal to

$$A\,(^{131}\mathrm{I}) = \eta^{-1}\,(^{131}\mathrm{I})\left(\frac{A_{\mathrm{hist}}}{M\cdot 60\mathrm{CF}_{\mathrm{off}}} - A_0\sum_{i=2}^{7} a_i\eta_i\right), \tag{8}$$

where A_0 is the specific activity of the nuclide to which the activity of all the other nuclides is normalized and $\mathrm{CF}_{\mathrm{off}}$ is the official scaling factor determined in accordance with [1, 11, 12].

The relative error in determining the activity of ^{131}I, which is due to the error in determining the scaling factor of any nuclide i, can be obtained by the known rules from (8):

$$\delta = \frac{\Delta A\,(^{131}\mathrm{I})}{A\,(^{131}\mathrm{I})}.$$

After simple rearrangements, we obtain

$$\delta = \frac{\mathrm{CF}\,(^{131}\mathrm{I})\,\dfrac{a_i}{\mathrm{CF}_i}\,\dfrac{\Delta\mathrm{CF}_i}{\mathrm{CF}_i}}{a\,(^{131}\mathrm{I})} = \frac{\mathrm{CF}\,(^{131}\mathrm{I})}{\mathrm{CF}_i}\,\frac{a_i}{a\,(^{131}\mathrm{I})}\,\frac{\Delta\mathrm{CF}_i}{\mathrm{CF}_i}. \tag{9}$$

Here, the relative error δ is expressed only in terms of the relative quantities. All these quantities, except $R_i = \dfrac{a_i}{a(^{131}\mathrm{I})}$, can be calculated by the Monte Carlo method. It should be noted that the quantity R_i for the ith nuclide in a sample is a function of time t. In this case, its estimation depends on the radiological model used for determining the entry of radionuclides into the human organism by the food chain. As an example, we will consider the R_i values obtained by the data of [6], where the measurement data on the beta-activity of milk produced in the Brest region were analyzed. The desired values of R_i were obtained by normalization of the relative content of the nuclides [6] ^{133}I, ^{134}Cs, ^{137}Cs, ^{136}Cs, ^{89}Sr, ^{90}Sr, ^{103}Ru, ^{106}Ru, ^{140}Ba, ^{141}Ce, and ^{144}Ce to the content of ^{131}I. Substituting the values of R_i determined for ^{89}Sr into formula (9), we obtain the error caused by the error in determining the scaling factor of ^{89}Sr; this error changes with time from 15% within 10 days after the radioactive fallout to 100% within 48–50 days after this fallout.

The appearance of such large errors is explained by the fact that, in the additive method of estimating the contributions of nuclides to the total activity of a milk sample, the absolute values of these contributions are considered (see formula (8)). Because of the small half-decay period of ^{131}I, its activity decreases rapidly with time and, before long, becomes equal to the activity of the nuclide for which the scaling factor was determined with a large error. For ^{89}Sr, this situation is realized within 48–50 days after the accident.

The multiplicative method of calculating the activity of ^{131}I in milk is much more stable in the sense of errors in determining the scaling factors. This method is based on the estimation of the relative content $F_i(t)$ of all the nuclides, including ^{131}I, in a milk sample:

$$F_i\,(t) = \frac{A_i\,(t)}{A_{\mathrm{total}}\,(t)}.$$

The relative content of the nuclides $F_i(t)$ is proportional to the response of the DP-100 radiometer to the radiation of the ith radionuclide in the sample. In this approach, the activity of iodine in milk is determined by the formula [6]

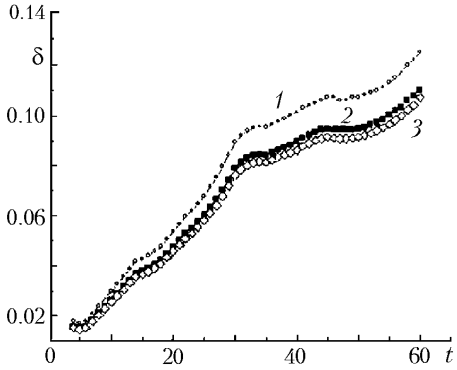

Fig. 1. The dependence of the relative error in determining the content of ^{131}I in milk δ caused by the error in determining the scaling factor for ^{89}Sr in milk, obtained for the Brest region with the use of the data presented in [8]: 1–3) numbers of measurement geometries.

$$A\,(^{131}\mathrm{I}) = \frac{A_{\mathrm{hist}}}{\mathrm{CF}_{\mathrm{off}}}\,\mathrm{DR}_{^{131}\mathrm{I}}\,(t)\,\mathrm{CF}_{^{131}\mathrm{I}}\,, \tag{10}$$

where the quantity $\mathrm{DR}_i(t)$ for the ith nuclide is determined from the expression

$$\mathrm{DR}_i\,(t) = \frac{F_i\,(t)/\mathrm{CF}_i}{\sum\limits_{k=1}^{N} F_k\,(t)/\mathrm{CF}_k} \tag{11}$$

(N is the total number of nuclides in milk). It is easily shown that an error in determining the scaling factor for the ith nuclide leads to a relative error in determining the iodine activity in milk:

$$\delta = \mathrm{DR}_i\,(t)\,\frac{\Delta\mathrm{CF}_i}{\mathrm{CF}_i}\,. \tag{12}$$

The values of $\mathrm{DR}_i(t)$ for the above-indicated nuclides, determined for the Brest region, are presented in [6]. The error in determining the scaling factor $\Delta\mathrm{CF}(^{89}\mathrm{Sr})$ can be estimated by the difference between the value of $\mathrm{CF}_{\mathrm{exp}}(^{89}\mathrm{Sr})$ taken from [3], which was used in the analysis, and the theoretical value of this quantity taken from Table 2. As a result, we obtained that $\Delta\mathrm{CF}(^{89}\mathrm{Sr})/\mathrm{CF}_{\mathrm{theor}}(^{89}\mathrm{Sr})$ is equal to 1.54, 1.79, and 1.52 for geometries 1, 2, and 3, respectively.

The change in the relative error in determining the content of ^{131}I in milk obtained in the Brest region, caused by the error in determining the scaling factor for ^{89}Sr, with time is shown in Fig. 1. This error increases with time; however, for a physically significant period (2 months), it does not exceed 12%.

Conclusions. A procedure of simulation of the process of measurement of the total beta-activity of a milk sample with the use of a DP-100 radiometer has been developed. This procedure allows one to determine the response of this radiometer to different nuclides and their combinations in a milk sample in the process of measurement of its beta-activity. The responses calculated by us for the three measurement geometries allow one to determine the scaling factors for any isotope with a known beta spectrum.

It is shown that the contribution of the Compton electrons arising due to the gamma-radiation emitted in the process of decay of the nuclides to the total beta-activity of a milk sample measured by the indicated radiometer comprises 25–30%.

In calculating the activity of individual nuclides by total beta-activity of a sample, preference should be given to the theoretical values of the scaling factors determined for these nuclides. This is supported by the fact that the

scaling factors calculated for the isotopes ^{137}Cs and ^{131}I, whose activity was reliably controlled by the gamma-chain of the nuclear decay, coincide with a high degree of accuracy with the corresponding experimental data.

The multiplicative analysis of the contribution of individual nuclides to the total beta-activity of a sample is more preferential than the additive analysis because it gives a smaller error in determining the content of ^{131}I in the sample and is much more stable in the sense of errors in determining the scaling factors for individual nuclides. For example, even though the errors in determining the content of ^{131}I in milk produced in the Brest region caused by the errors in determining the scaling factor for ^{89}Sr increase with time, they do not exceed 12% for a physically significant period (two months).

Since the estimation of the content of ^{131}I in milk by the results of measurements of its total beta-activity in 1986 is based on the assumption that only the nuclides ^{131}I, ^{137}Cs, ^{134}Cs, ^{89}Sr, ^{90}Sr, ^{144}Ce, and ^{106}Ru, for which the scaling factors were determined experimentally, are contained in milk, at present the results of these measurements should be analyzed again with consideration for all the nuclides contained in milk. Along with the above-listed nuclides, probably the nuclides ^{133}I, ^{140}Ba, and ^{140}La, for which the scaling coefficients were not determined experimentally, should be taken into account.

NOTATION

A, total specific activity, Bq/g; A_{hist}, total beta-activity of a sample calculated in 1986 with the use of a scaling factor CF_{off}, Bq/g; A_i, specific activity of the ith isotope in milk, Bq/g; A_{total}, total activity of all the isotopes contained in milk, Bq/g; A_1, A_2, a, dimensionless interpolation coefficients (Table 1); CF, scaling factor, Bq/g (pulse/min); CF_{off}, officially recommended scaling factor; d, inner diameter of the cuvette, mm; DF, number of degrees of freedom used in the interpolation of the response of the DP-100 radiometer by the analytical formula (6); DR_i, relative contribution of the activity of the ith nuclide contained in a milk sample to the total response of the detector; E, energy of electrons, MeV; E_0, interpolation coefficient, MeV; E_i, energy of the ith source of ionizing radiation, keV; E_{min}, minimum energy of electrons capable of passing through the entrance window of a Geiger counter, MeV; E_{max}, maximum energy of the beta-spectrum of a nuclide, MeV; $F_i(t)$, relative content of the ith nuclide in the sample being studied; h, height of the cuvette, mm; L, distance between the surface of the liquid and the surface of the beta-counter, mm; M, mass of the sample, g; N_c, number of counts of the counter, pulse; $N_h(E_i)$, total number of particles with an energy E_i emitted from the source, pulse; P_m, rate of counting of a sample, pulse/min; P_b, rate of counting of the background, pulse/min; $p(E_i)$, relative intensity of the particles with an energy E_i; t, time elapsed from the Chernobyl accident, days; δ, relative error in determining the content of ^{131}I in milk caused by the error in determining the scaling factor for the ith nuclide; η, response function of the device; η_i, response of the radiometer to the radiation of the ith nuclide; χ^2, statistical criterion for determining an error. Subscripts: total, summary; off, official; 0, initial; m, mean; i, serial number of a radionuclide; min, minimum; max, maximum; c, calculational; b, background; hist, historical; h, events taking place in the history.

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