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MONTE CARLO MODELING OF BETA-RADIOMETER DEVICE USED TO MEASURE MILK CONTAMINATED AS A RESULT OF THE CHERNOBYL ACCIDENT

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Abstract

This paper presents results of Monte Carlo modeling of the beta-radiometer device with Geiger-Mueller detector used in Belarus and Russia to measure the radioactive contamination of milk after the Chernobyl accident. This type of detector, which is not energy selective, measured the total beta-activity of the radionuclide mix. A mathematical model of the beta-radiometer device, namely DP-100, was developed, and the calibration factors for the different radionuclides that might contribute to the milk contamination were calculated. The estimated calibration factors for ^{131}I, ^{137}Cs, ^{134}Cs, ^{90}Sr, ^{144}Ce, and ^{106}Ru reasonably agree with calibration factors determined experimentally. The calculated calibration factors for ^{132}Te, ^{132}I, ^{133}I, ^{136}Cs, ^{103}Ru, ^{140}Ba, ^{140}La, and ^{141}Ce had not been previously determined experimentally. The obtained results allow to derive the activity of specific radionuclides, in particular ^{131}I, from the results of the total beta-activity measurements in milk. Results of this study are important for the purposes of retrospective dosimetry that uses measurements of radioactivity in environmental samples performed with beta-radiometer devices.

Keywords

Geiger-Mueller; radionuclide; calibration factor; Chernobyl

INTRODUCTION

The accident at the Chernobyl nuclear power plant resulted in widespread radioactive contamination, particularly of the territories of Belarus, the Russian Federation, and Ukraine.

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For the majority of persons living in these territories, most of the radiation exposure to the thyroid was from iodine isotopes, especially ^{131}I. The U.S. National Cancer Institute is conducting a long-term cohort study in Belarus of thyroid cancer among the persons exposed to radiation in childhood following the Chernobyl accident. The reconstruction of the individual doses for the cohort members is based on the measurements of the ^{131}I contents in their thyroid glands, which were performed during the few first weeks after the accident. As the consumption of locally produced milk was the main source of ^{131}I intake by people, the determination of ^{131}I concentration in milk can be used to confirm the validity of the thyroid doses. The measurements of total beta-activity in milk provide the opportunity to derive ^{131}I concentration.

Sanitary and Hygiene Centers of the USSR Ministry of Health performed measurements of total beta-activity in milk in different regions of Belarus to control the level of radioactive contamination in the locally produced foodstuffs following the Chernobyl accident. The majority of the measurements of radioactivity in cow's milk were made with a beta-radiometer device, namely DP-100, equipped with a Geiger-Mueller (GM) detector. This type of detector, which is not energy selective, recorded counts due to the activity of the radionuclide mix. In the initial processing of the results that was carried out in 1986, a so-called "official" calibration factor was used to derive the total beta-activity of the sample from the count rate recorded by the device. The "official" calibration factor corresponds to beta rays of 0.3 MeV (Lyarsky, 1965) and is independent of the time of the measurement. To estimate the activity of specific radionuclides, in particular ^{131}I, from the results of the total beta-activity measurements in milk in a more rigorous manner, it is necessary: (1) to evaluate the time-dependent composition of the radionuclide mix in milk; and (2) to calculate the calibration factors for these radionuclides.

Drozdovitch et al. (2006) estimated the time-dependent fraction of each radionuclide in the total activity in milk by using radioecological model with parameters adapted to the local conditions. Relative contribution of each radionuclide to the count rate of the device was calculated by dividing that fraction of the activity by the calibration factor for the considered radionuclide and, then, dividing it by the total calculated count rate. Finally, to estimate the activity of a given radionuclide in milk, the actual count rate recorded by the device during the measurements in 1986 was multiplied by the relative contribution of that radionuclide to the count rate of the device and by its calibration factor.

Contamination from the Chernobyl fallout of cow's milk with ^{131}I, ^{133}I, ^{134}Cs, ^{136}Cs, ^{137}Cs, ^{89}Sr, ^{90}Sr, ^{103}Ru, ^{106}Ru, ^{140}Ba, ^{141}Ce, and ^{144}Ce was considered in a paper by Drozdovitch et al. (2006). However, calibration factors were available only for ^{131}I, ^{134}Cs, ^{137}Cs, ^{90}Sr, ^{106}Ru, and ^{144}Ce. They were evaluated experimentally by Savkin et al. (1992) based on measurements of standard solutions of these radionuclides. Savkin et al. (1992) also estimated the calibration factor for ^{89}Sr by interpolation of the calibration factors for other radionuclides with similar beta spectra and energies. However, the calibration factors for other potentially important radionuclides (^{132}Te, ^{132}I, ^{133}I, ^{136}Cs, ^{103}Ru, ^{140}Ba, ^{140}La, and ^{141}Ce) were not determined in those studies.

The purpose of this work was to estimate the calibration factors for all radionuclides that contaminated milk after the Chernobyl accident using a mathematical model of the beta-radiometer DP-100. Accounting for all radionuclides presented in contaminated milk may greatly improve the results of estimation of ^{131}I activity derived from total beta-activity measured in milk shortly after the Chernobyl accident.

METHODS

Description of the beta-radiometer DP-100

The total beta-activity in milk was measured with the beta-radiometer DP-100 by placing the milk sample in an aluminum dish located in a plastic glass support. Both the plastic glass support with the sample and the gas end-window GM counter, namely MST-17, were placed in a lead cylindrical shield. Figure 1 shows a scheme of the DP-100 and of the geometry of measurement. Table 1 gives the size of the dish and the distance between the detector and the dish for three standard geometries that were used to measure the total beta-activity in the milk samples.

Beta particles enter the GM counter through a window in a plane metallic base that is covered with a thin layer (5 mg cm^{-2}) of mica. Cathode of the counter is a thin layer (~ 1 μm) of metal covered the GM tube inside; anode is a metal wire placed as axis of the tube. Density of the gas inside the GM counter is around 120 g m^{-3}. The diameter of the GM counter is 2 cm, while its length is 2.5 cm.

Any beta-particle traversing the space between the anode and the cathode is counted with a probability equal to 1. Due to the construction of the GM counter, the probability of registration of gamma-rays is three to four orders of magnitude less than that of beta-particles. However, gamma-radiation contributes to counting due to interaction of gamma-radiation with matter in the shield, milk sample, and components of the counter, which produces Compton scattering, photoelectrons and electron-positron pairs. The secondary electrons from these processes are also registered by the counter. Therefore, modeling of the process of measurement with the DP-100 device should take into account both beta and gamma chains of decay of radionuclides in the milk sample. It should be noted that emitted beta-particles (both primary and secondary) lose some energy when they interact with matter, and, therefore, only a fraction of the beta particles will reach the sensitive volume of the GM counter. This fraction needs to be calculated in order to estimate the device response. This requires modeling of the transport of the beta- and gamma-rays emitted by the source through the materials surrounding the detector.

The MCNP4 software (Briesmeister, 1994) was used to model the DP-100 device, the processes of the electron and photon transport and interaction with matter, and registration by the GM counter.

Mathematical model of the DP-100 device

A detailed mathematical model of the DP-100 device with the GM counter was developed. All parts of the device, namely, the source of particles, the detector, and the surrounding of the detector, were considered in the Monte Carlo modeling of the process of instrumental measurements. Density and chemical composition of the constructing materials were also taken into account.

Two types of electrons were considered to be registered by the detector: (a) primary electrons emitted by the source (major contribution to the device count rate) and (b) secondary electrons resulted from the interaction of the emitted photons with the surrounding materials (minor contribution). The MCNP4 software (Briesmeister, 1994) was used to simulate the electrons and photons transport as well as generation of secondary electrons due to photoelectric effect, Compton scattering, and pair production. It was assumed that any electron was counted when it reached the sensitive volume of the detector. The source of electrons and photons was considered to be uniformly distributed in the dish with milk sample, and isotropic direction of particle's emission from that source was also assumed. Track of electron (photon) was modeled until registration in the sensitive area of the detector or absorption by the lead shield or other parts of the device. Absorption in the milk sample was also taken into account.

Device response and calibration factor for mono-energetic electrons

The response of the DP-100 device to a mono-energetic source of electrons is defined as

$$\eta(E_i)=\frac{N_c}{N_h(E_i)}, \tag{1}$$

where

$\eta(E_i)$ is the response of the device to electron with energy E_i (unitless), N_c is the measured count rate (s^{-1}), and N_h (E_i) is the total number of particles with energy E_i emitted per unit time (s^{-1}).

The calibration factor for a monoenergetic source is defined as the activity of the sample per unit count rate and is determined as

$$CF(E_i)=1/(M\cdot k\cdot \eta(E_i)), \tag{2}$$

where $CF(E_i)$ is the calibration factor for the source of energy E_i (Bq g^{-1} per count min^{-1}), M is the mass of sample (g), and k = 60 is the number of seconds in a minute (s min^{-1}).

Calibration factor for radionuclides

Response of the DP-100 device to a source with a complex energy spectrum is defined as

$$\eta=\sum_i p(E_i)\cdot \eta(E_i), \tag{3}$$

where η is the response of the DP-100 device for the source with a complex energy spectrum (dimensionless) and $p(E_i)$ is the yield of particles with energy E_i (dimensionless).

The calibration factor for the DP-100 device for a source with a complex energy spectrum is defined as

$$CF=1/[M\cdot k\cdot \sum_i p(E_i)\cdot \eta(E_i)]. \tag{4}$$

For a pure beta emitter, the sum in equation (4) can be replaced with the integral of the beta-spectrum of the nuclide, including its progeny (Eckerman et al., 1994):

$$CF=1/[M\cdot k\cdot \int_{E_{\min}}^{E_{\max}} p(E)\cdot \eta(E)dE], \tag{5}$$

where $E_{\min}$ is the minimal energy of electron that can pass through window of the GM counter (MeV), $E_{\max}$ is the maximal energy of beta-spectrum (MeV), $p(E)$ is the beta-spectrum taken from ICRP Publication 38 (ICRP, 2003).

The same calculations were made for the gamma emission of the radionuclides to take into account secondary electrons produced by interaction of gamma-radiation with matter. Device response for each gamma-emitting radionuclide was calculated with the MCNP4 software

taking into account the production of secondary electrons by interaction of the emitted photons with the surrounding matter and registration of the secondary electrons by the detector.

RESULTS AND DISCUSSION

Response of the DP-100 device to monoenergetic electrons

Response of the DP-100 device was calculated for electrons and photons of the energies in the range from 0.1 to 3 MeV using Monte Carlo simulations of particle transport from the source to the detector and interaction of electrons and photons with matter. Tables 2 and 3 give estimated energy-dependent device response to primary and secondary electrons, respectively.

As can be seen from Tables 2 and 3, the response of the DP-100 device to electron of a given energy that are products of radioactive decay is more than one order of magnitude higher than that to the secondary electrons produced by photon of the same energy. The energy-dependent device response given in Table 2 for primary electrons was fitted by the following curve:

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

where - A_1, A_2, and a are dimensionless parameters (Table 4), E is the energy of the emitted electron (MeV) and E_0 is a fitted parameter value (MeV).

Figure 2 shows the estimated response of the DP-100 device for different measurement geometries and fitted function described by Equation 6. Errors in estimated response vary from (10–13)% for energies below 0.2 MeV to (3–5)% for energies above 1 MeV.

Calibration factors for radionuclides

Calibration factors for the DP-100 device were calculated using Equations 5 and 6 for the following nuclides: ^{131}I, ^{132}Te, ^{132}I, ^{133}I, ^{134}Cs, ^{136}Cs, ^{137}Cs, ^{89}Sr, ^{90}Sr, ^{103}Ru, ^{106}Ru, ^{140}Ba, ^{140}La, ^{141}Ce, and ^{144}Ce (Table 5). The calibration factors for ^{137}Cs, ^{90}Sr, ^{106}Ru, and ^{144}Ce include contribution from short-lived progenies ^{137m}Ba, ^{90}Y, ^{106}Rh, and ^{144}Pr, respectively. It should be noted that the secondary electrons contributed significantly to the value of the calibration factor for radionuclides with low energy of electrons, but with high energy of photons, for example ^{136}Cs.

The radionuclide-specific calibration factors calculated in this study were compared with those evaluated experimentally by Savkin et al. (1992) (Table 6). Radionuclide-specific errors in experimentally evaluated calibration factor were also taken from the paper by Savkin et al. (1992). As can be seen from Table 6, the two sets of values (calculated and experimental) are rather close, mainly within the uncertainties of the experimental calibration factor. The biggest difference between the calculated and experimental calibration factors was found for ^{89}Sr. It is noteworthy that the calibration factor for this radionuclide given by Savkin et al. (1992) was actually estimated by interpolation on average energy of beta-spectrum and experimental calibration factors for other radionuclides. That interpolation did not take into account the fact that low-energy electrons have small contribution to the device response (Table 2). Therefore, the calibration factor for ^{89}Sr calculated in this paper can be regarded as more accurate.

CONCLUSIONS

Results of the total beta-activity measurements in cow's milk are very useful for the reconstruction of thyroid doses from ^{131}I intake with cow's milk in areas affected by the

Chernobyl accident. These measurements were performed with radiometer DP-100 with the GM counter, which is not energy selective. As a result, the total beta-activity of the radionuclide mix was measured.

Several studies were conducted to estimate radionuclide-specific activities from the results of total beta-activity measurements (Panchenko, 1999; Savkin et al., 2004; Zvonova et al., 2004; Drozdovitch et al., 2006). However, these studies considered only the following nuclides: ^{131}I, ^{137}Cs, ^{134}Cs, ^{90}Sr, ^{144}Ce, and ^{106}Ru (i.e., radionuclides for which calibration factors were known at that time from experiment).

In this work, a mathematical model of the DP-100 device was used to estimate the calibration factors for a broader list of radionuclides that might contribute to the milk contamination, including ^{132}Te, ^{132}I, ^{133}I, ^{136}Cs, ^{103}Ru, ^{140}Ba, ^{140}La, and ^{141}Ce., The DP-100 calibration factors for these radionuclides had never been estimated. The calibration factors calculated for the short-lived radionuclides are especially important for the measurements performed shortly after the Chernobyl accident. Results of this study make it possible to derive the activity of ^{131}I from the total beta-activity measurements more accuracy.

Acknowledgements

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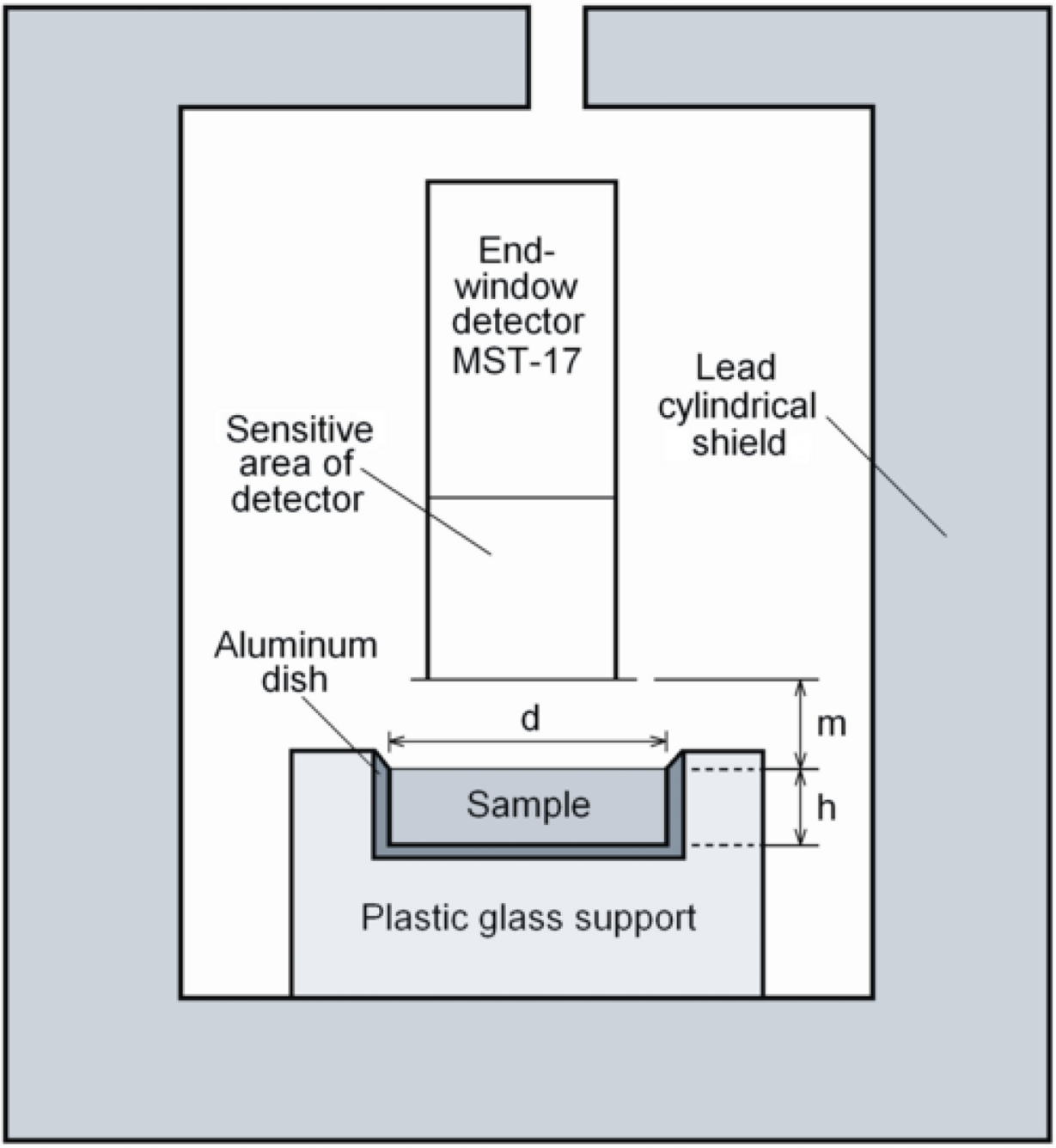

Figure 1.
DP-100 device and geometry of measurement performed by the device. The values of *d*, *h*, and *m* for the three standard geometries are given in Table 3.

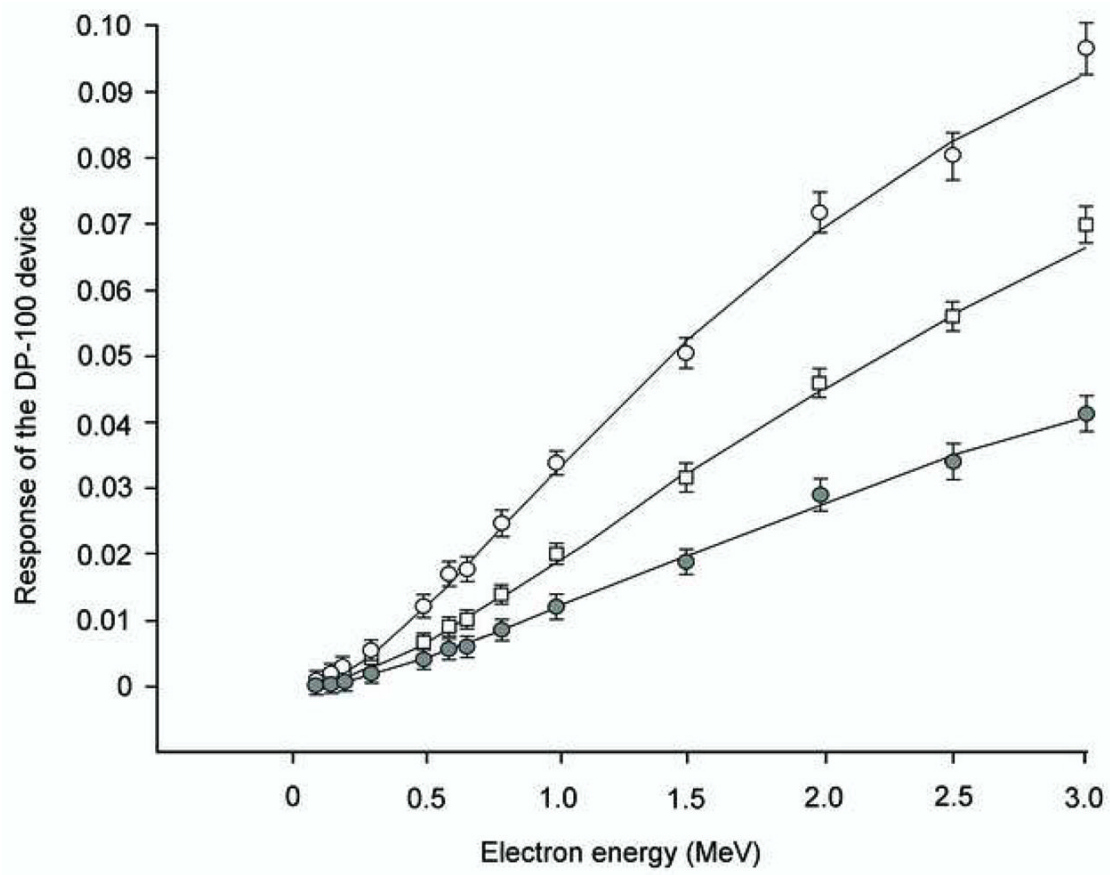

Figure 2.
Energy-dependent response of the DP-100 device for standard geometries #1 (open circles); #2 (open squares); and #3 (filled circles). The solid curves represent the Equation 6 fits.

Table 1

Standard geometries used to measure the total beta-activity in milk samples

Parameter	Measurement geometry		
	#1	#2	#3
Inner diameter of dish with milk sample, *d* (cm)	2.6	4	4
Depth of dish, *h* (cm)	1	1	1
Distance between the detector and the dish, *m* (cm)	1	1	2

Table 2

Energy-dependent response of the DP-100 device to electrons

Electron energy (MeV)	Device response		
	Geometry #1	Geometry #2	Geometry #3
0.1	0.00025	0.0001	0.00007
0.15	0.0011	0.0008	0.0004
0.2	0.0021	0.0012	0.0008
0.3	0.0053	0.0028	0.0021
0.5	0.012	0.0067	0.0041
0.6	0.017	0.0089	0.0057
0.7	0.018	0.0097	0.0058
0.8	0.025	0.014	0.0087
1.0	0.033	0.020	0.012
1.5	0.049	0.031	0.019
2.0	0.069	0.046	0.029
2.5	0.080	0.056	0.034
3.0	0.096	0.070	0.041

Table 3

Energy-dependent response of the DP-100 device to secondary electrons produced by photons

Photon energy	Device response		
(MeV)	Geometry #1	Geometry #2	Geometry #3
0.1	0.00004	0.000035	0.00002
0.15	0.00006	0.00004	0.00003
0.2	0.00008	0.00008	0.00005
0.3	0.00022	0.00018	0.00011
0.5	0.0007	0.0007	0.0003
0.6	0.0008	0.0007	0.00048
0.7	0.0011	0.0009	0.00049
0.8	0.0014	0.0011	0.00076
1.0	0.0017	0.0014	0.00083
1.5	0.0027	0.0022	0.0013
2.0	0.0033	0.0028	0.0017
2.5	0.0035	0.0029	0.0018
3.0	0.0036	0.0031	0.0020

Table 4

Parameters of the fits of the DP-100 response dependence with Equation 6

Parameter	Geometry #1	Geometry #2	Geometry #3
A_1	−0.00071 ± 0.00014	−0.00047 ± 0.00008	−0.00034 ± 0.00006
A_2	0.1423 ± 0.0117	0.1437 ± 0.0280	0.0957 ± 0.0253
E_0	2.05 ± 0.20	3.30 ± 0.68	3.61 ± 1.01
a	1.666 ± 0.064	1.568 ± 0.073	1.519 ± 0.086

Table 5

Estimated calibration factors for the DP-100 device, CF_i

Radionuclide	Main electron energy[a] (MeV)	Calibration factor (Bq g^{-1} per count min^{-1})		
		Geometry #1	Geometry #2	Geometry #3
^{131}I	0.33	1.5	0.94	1.5
^{132}Te	0.15	16.1	8.6	13.4
^{132}I	0.67	0.29	0.18	0.29
^{133}I	0.67	0.4	0.25	0.4
^{134}Cs	0.36	0.91	0.48	0.73
^{136}Cs	0.20	1.0	0.5	0.75
^{137}Cs[b]	0.31	1.25	0.73	0.93
^{89}Sr	0.94	0.26	0.14	0.23
^{90}Sr[b]	1.40	0.26	0.14	0.24
^{103}Ru	0.143	4.1	2.2	3.4
^{106}Ru[b]	1.8	0.16	0.096	0.16
^{140}Ba	0.56	0.73	0.46	0.73
^{140}La	0.77	0.27	0.17	0.28
^{141}Ce	0.26	2.5	1.6	2.4
^{144}Ce[b]	1.66	0.19	0.089	0.17

[a] Energy of electrons that make the biggest contribution to the response of the DP-100 device.

[b] Calibration factor for that radionuclide includes a contribution from the short-lived progeny.

Table 6

Comparison of the calibration factors for the DP-100 device calculated in this study (C) and determined experimentally by Savkin et al. (1992; 2004) (E)

Radionuclide	Calibration factor (Bq g^{-1} per count min^{-1})						Errors in experimental *CF* (%)
	Geometry #1		Geometry #2		Geometry #3		
	C | E	(C–E)/E (%)	C | E	(C–E)/E (%)	C | E	(C-E)/E (%)	
^{131}I	1.5 | 1.9	−21	0.94 | 1.2	−22	1.5 | 1.8	−17	21
^{134}Cs	0.91 | 1.4	−35	0.48 | 0.84	−43	0.73 | 1.2	−39	22
^{137}Cs	1.25 | 1.3	−4	0.73 | 0.65	12	0.93 | 0.94	−1	39
^{89}Sr	0.26 | 0.67	−61	0.14 | 0.39	−64	0.23 | 0.58	−60	30
^{90}Sr	0.26 | 0.36	−28	0.14 | 0.20	−30	0.24 | 0.30	−20	25
^{106}Ru	0.16 | 0.14	14	0.096 | 0.11	−13	0.16 | 0.15	7	74
^{144}Ce	0.19 | 0.15	27	0.089 | 0.083	7	0.17 | 0.13	31	24