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Activity concentrations of ^{131}I and other radionuclides in cow's milk in Belarus during the first month following the Chernobyl accident

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Abstract

The accident at the Chernobyl nuclear power plant (NPP) in Ukraine on April 26, 1986 led to a considerable release of radioactive material resulting in environmental contamination over vast areas of Belarus, Ukraine and western Russian Federation. The major health effect of the Chernobyl accident was an increase in thyroid cancer incidence in people exposed as children and adolescents, so much attention was paid to the thyroid doses resulting from intakes of ^{131}I. Because cow's milk consumption was the main source of ^{131}I intake by people, it was important to measure the ^{131}I activity concentrations in cow's milk to calculate, or to validate, the thyroid doses to the exposed population. Almost 11,000 measurements of total beta-activity in cow's milk were performed using a DP-100 device during the first month after the Chernobyl accident in the most contaminated regions of Belarus. Using an ecological model and calibration coefficients for the DP-100 device the activity concentration of ^{131}I in cow's milk was derived as well as the activity concentrations of the other radiologically important radionuclides, namely ^{134}Cs, ^{137}Cs, ^{89}Sr and ^{90}Sr. The activity concentrations of other radionuclides, such as ^{90}Y, ^{132}Te, ^{132}I, ^{133}I, ^{136}Cs, ^{140}Ba, ^{140}La, ^{141}Ce and ^{144}Ce, in cow's milk were also estimated and were shown to be of minor importance. The concentrations of ^{95}Zr, ^{95}Nb, ^{103}Ru and ^{106}Ru in cow's milk were negligible. The data obtained in this study were validated by comparing derived ^{131}I and ^{137}Cs concentrations in cow's milk with gamma-spectrometry measurements performed in milk

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Declaration of competing interest
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produced in the same location close to the same date. The results of this study were essential to assess and validate the radiation doses received by the subjects of epidemiological studies related to the health consequences of the Chernobyl accident.

Keywords

Chernobyl; Cow's milk; Total beta-activity; ^{131}I; ^{134}Cs; ^{137}Cs; ^{89}Sr; ^{90}Sr

1. Introduction

The Chernobyl accident on April 26, 1986 resulted in massive releases of radioactive materials into the atmosphere and contamination of large areas in Belarus, Ukraine and western Russia with significant levels of isotopes of radioiodine, radiocesium and other radionuclides. Radiation epidemiology studies (e.g., Cardis et al., 2005; Tronko et al., 2017; Zablotska et al., 2011) found that an increase of incidence of thyroid cancer in persons exposed as children and adolescents was one of the major health effects of the Chernobyl accident among members of the public. Thyroid doses after the Chernobyl accident arose largely from intakes of ^{131}I, a radionuclide with a physical half-life of 8.02 days that accumulates in the thyroid. For most individuals, the ^{131}I intake was largely due to the consumption of fresh cow's milk, inhalation of contaminated air played a minor role (Drozdovitch et al., 2013a). To evaluate the radiation-related risk of thyroid cancer in epidemiological studies, considerable efforts were made to reconstruct individual thyroid doses received by the people included in these studies. Thyroid doses and associated uncertainties were calculated using models (Gavrilin et al., 2004; Drozdovitch et al., 2010) with use of the measurement of exposure rate against the neck (called 'direct thyroid measurement') when available (Drozdovitch et al., 2013a; Likhtarov et al., 2014). To reduce uncertainties in thyroid doses, it is essential to use measurements of ^{131}I activity in environmental samples, including soil, grass and cow's milk.

Shortly after the accident, a wide-scale screening of radionuclide contamination of locally produced cow's milk was undertaken in Belarus. Measurements of total beta-activity were organized by the Sanitary and Hygiene Centers of the Ministry of Health and the Veterinary Laboratories of the State Agro-Industrial Committee of the USSR. Most of the measurements were made with a beta-radiometer device, called DP-100, equipped with a Geiger-Mueller (GM) detector.

The data on total beta-activity in cow's milk in Belarus were used to derive ^{131}I activity concentrations and to calculate the thyroid doses from ^{131}I intake (Savkin et al., 2004; Drozdovitch et al., 2006). The first analysis, performed by Savkin et al. (2004), included 18,494 measurements of total beta-activity in cow's milk that were collected from the archives of sanitary and veterinary laboratories of 17 raions in Gomel Oblast. About 40% of measurements were below the minimum detectable level of the DP-100 device and could not be used in the analysis. For the other milk samples, an empirical approach was developed to assess the activity concentration of the ^{131}I, ^{137}Cs, ^{134}Cs, ^{90}Sr, ^{144}Ce, and ^{106}Ru mixture and to derive the concentration of ^{131}I from the total beta-activity measurement.

Drozdovitch et al. (2006) used a radioecological model to derive the time-dependent activity concentrations of ^{131}I from the total beta-activity measurements of cow's milk made in Brest Oblast of Belarus. The results were used to reconstruct the ^{131}I activity concentrations in milk for territories with different ^{137}Cs deposition and to estimate thyroid doses from the consumption of ^{131}I-contaminated cow's milk by the population of Brest Oblast. A similar analysis of total beta-activity measurements in cow's milk was also carried out in Russia by Panchenko (1999) and Zvonova et al. (2004).

This paper presents the results of a new study to derive the activity concentrations of ^{131}I and other radionuclides from total beta-activity measurements in cow's milk performed in Gomel and Mogilev Oblasts of Belarus shortly after the Chernobyl accident. We used (i) the complete database of the total beta-activity and gamma-spectrometry measurements in cow's milk that were performed in the most contaminated Gomel and Mogilev Oblasts, and (ii) calibration coefficients for a broader list of radionuclides, including ^{89}Sr, ^{90}Sr, ^{90}Y, ^{95}Zr, ^{95}Nb, ^{103}Ru, ^{106}Ru, ^{131}I, ^{132}Te, ^{132}I, ^{133}I, ^{134}Cs,^{136}Cs, ^{137}Cs, ^{140}Ba, ^{140}La, ^{141}Ce, and ^{144}Ce (radionuclides are listed according to increasing mass number, not according to their contribution to cow's milk concentration), which were calculated using a Monte Carlo method of numerical simulation of electron and gamma-ray transport in the DP-100 device.

2. Materials and methods

2.1. Total beta-activity measurements in cow's milk

The total beta-activity and gamma-spectrometry measurements in cow's milk samples in Belarus were collected into a database of meteorological and radiation measurements (BRCRCEM database) (Drozdovitch et al., 2013b). A total of 24,388 measurements of total beta-activity in cow's milk done between 29 April and June 30, 1986 in Gomel and Mogilev Oblasts are available in the database. Of these data, the 10,631 measurements that were performed up to May 31, 1986 were the most appropriate for the estimation of ^{131}I concentrations. Unfortunately, only 122 gamma-spectrometry measurements of cow's milk were done from 5 May to May 31, 1986. Fig. 1 shows the locations where cow's milk samples were taken from 29 April to May 31, 1986 in Gomel and Mogilev Oblasts.

2.2. Description of the beta-radiometer DP-100

The total beta-activity in milk was measured with a beta-radiometer DP-100 equipped with a GM counter named MST-17. The milk samples were placed in an aluminum dish located in a plastic glass support (MA, 1968). Both the plastic glass support with the sample and the gas end-window GM counter were placed into a lead cylindrical shielding. Table 1 gives the size of the dish and the distance between the detector and the dish for the three standard geometries that were used to measure the total beta-activity in the milk samples. Fig. 2 shows a schema of the DP-100 and of the geometry of measurement. The cathode of the counter is a thin layer (~1 μm) of metal covering the inner wall of the GM tube, while the anode is a metallic wire along the axis of the tube. The diameter of the GM counter is 2 cm, while its length is 2.5 cm.

Beta particles enter the GM counter through the end-window, which is a plane metallic ring with a thin layer (5 mg cm^{-2}) of mica. It was assumed that any beta-particle traversing the space between the anode and the cathode would be counted with a probability equal to 1. The probability of counting photons is less than that for beta-particles. Photons contribute to the counts recorded by the device due to their interaction with the shielding, milk sample, and components of the counter that produces Compton scattering, photoelectrons and electron-positron pairs. GM-type counter is sensitive to all secondary electrons from these processes. Therefore, modeling of the process of measurement with the DP-100 device should consider all types of radiation from the radioactive decay of radionuclides in a milk sample. Beta-particles that are emitted from the sample lose some energy when they interact with surrounding materials, and some beta particles will be lost on the way to the sensitive volume of the GM counter. Monte-Carlo simulations of the transport of radiation emitted from the milk sample were performed to calculate the number of electrons entering the sensitive volume of the GM counter and, consequently, the DP-100 device response (Khrutchinsky et al., 2009).

2.3. Processing of results of total beta-activity measurements

We processed the results of the 10,631 measurements of total beta-activity in cow's milk that were done between the end of April and May 31, 1986. The following information was recorded in the database in counts min^{-1}: the measured DP-100 device count rate of the milk sample, N_{dev}; the background count rate, N_{bg}; and the net count rate due to the activity in the milk sample, $N_{net} = N_{dev} - N_{bg}$. More than half of the results of the total beta-activity in cow's milk recorded in the database had values of the net count rate of the sample, N_{net}, close to zero because of the relatively high background count rate of the DP-100 device, which ranged from 7 to 73 counts min^{-1} depending on the background at the measurement site. According to the database records, the measured total beta-activities in 7697 of the 10,631 milk samples had net count rates greater than the critical level (CL), which was the counting rate from a sample containing the minimum significant activity (IAEA, 2007). From these 7697 results, only 5447 provided estimates of total beta-activity in milk exceeding the lower limit of detection (LLD) (see Appendix A). For the remaining 5184 results, the net count rates, N_{net}, were lower than CL, and the value of N_{net} was replaced with a value 'less-than level', L_{l-t}. The description of how the results of total beta-activity measurements that were close or less than CL were processed is given in Appendix B.

To derive the concentration of a specific radionuclide (e.g. ^{131}I) in cow's milk from the results of total beta-activity measurements, it was necessary: (1) to evaluate the time-dependent composition of radionuclides in milk from the Chernobyl fallout; (2) to know the calibration coefficients for all these radionuclides; and (3) to extract from the overall count rate of the DP-100 device the contribution of the specific radionuclide under consideration (e.g. ^{131}I). Fig. 3 presents the schema of calculation of the activity concentration of a specific radionuclide in cow's milk from the total beta-activity measurements. The steps of calculation are discussed in the following sections.

2.4. Radionuclide composition in milk

To estimate the radionuclide composition in cow's milk, the fraction of the total concentration due to each important radionuclide was calculated. The time-dependent fractional concentration of radionuclide *i* in cow's milk, $F_i(t)$, was defined as:

$$F_i(t) = Q_i(t) \Big/ \sum_i Q_i(t) \quad (1)$$

where $Q_i(t)$ is the concentration of radionuclide *i* in milk at time *t* (kBq kg^{-1}).

A detailed description of how the time-dependent fractional concentrations of radionuclides in cow's milk, $F_i(t)$, were calculated is given elsewhere (Drozdovitch et al., 2006). In brief, the variation with time of the concentration in cow's milk due to the consumption of radionuclide *i* in pasture grass was expressed as (Müller and Pröhl, 1993):

$$Q_i(t) = \sigma_i \cdot \frac{f_i}{Y} \cdot I_g \cdot TF_i \cdot \int_0^t e^{-\left(\lambda_i^r + \lambda_i^w\right) \cdot \tau} \cdot \sum_{j=1}^{J_i} a_{i,j} \cdot \lambda_{i,j}^m \cdot e^{-\left(\lambda_i^r + \lambda_{i,j}^m\right) \cdot (t-\tau)} d\tau, \quad (2)$$

where σ_i is the ground deposition density of radionuclide *i* (kBq m^{-2}); f_i is the interception factor of radionuclide *i* by grass (unitless); *Y* is the yield of pasture grass at the time of deposition (kg m^{-2}); I_g is the daily intake of grass by cows (kg d^{-1}); TF_i is the cow's intake-to-milk transfer coefficient (d kg^{-1}); λ_i^r is the radioactive decay constant of radionuclide *i* (d^{-1}); λ_i^w is the rate of loss of radionuclide activity by grass due to weathering and growth dilution (d^{-1}); J_i is the number of components *j* of the function describing the retention of radionuclide *i* in the cow's body; $a_{i,j}$ is the fraction of activity associated with component *j*; $\lambda_{i,j}^m$ is the biological transfer rate associated with component *j* (d^{-1}); *t* is time counted since deposition of fallout on the ground (d).

The ground deposition density of radionuclide *i*, σ_i, was derived from the ground deposition density of ^{137}Cs as:

$$\sigma_i = \sigma_{Cs137} \cdot R_i, \quad (3)$$

where σ_{Cs137} is the ground deposition density of ^{137}Cs, which was measured in each settlement in Belarus (kBq m^{-2}) (SCHRB, 2006); R_i is the ratio of ground deposition densities of radionuclide *i* and of ^{137}Cs (unitless).

In the same manner, the value of the interception factor by grass of radionuclide *i*, f_i, was derived from the corresponding value for ^{137}Cs as follows:

$$f_i = f_{Cs137} \cdot R_{f,i}, \quad (4)$$

where f_{Cs137} is the interception factor of ^{137}Cs by grass (unitless); $R_{f,i}$ is the ratio of interception factors for radionuclide *i* and for ^{137}Cs (unitless).

From eqns (1)–(4), the time-dependent fraction of the total activity concentration in cow's milk due to radionuclide *i* can be written in the following form:

$$F_i(t) = \frac{R_i \cdot R_{f,i} \cdot TF_i \cdot \int_0^t e^{-\left(\lambda_i^r + \lambda_i^w\right) \cdot \tau} \cdot \sum_{j=1}^{J_i} a_{i,j} \cdot \lambda_{i,j}^m \cdot e^{-\left(\lambda_i^r + \lambda_{i,j}^m\right) \cdot (t-\tau)} d\tau}{\sum_i R_i \cdot R_{f,i} \cdot TF_i \cdot \int_0^t e^{-\left(\lambda_i^r + \lambda_i^w\right) \cdot \tau} \cdot \sum_{j=1}^{J_i} a_{i,j} \cdot \lambda_{i,j}^m \cdot e^{-\left(\lambda_i^r + \lambda_{ij}^m\right) \cdot (t-\tau)} d\tau} \quad (5)$$

For the purposes of this study, the territory of Gomel and Mogilev Oblasts (Fig. 4) was sub-divided into three regions considering dates and type (dry or wet) of deposition:

- Southern spot, including the contaminated raions of the southern part of Gomel Oblast, namely Bragin, Khoiniki, Elsk, Mozyr, Narovlya, Kalinkovichi, Lelchitsy, Loev, and Rechitsa raions, where predominantly dry deposition occurred on 26–27 April 1986 and the pasture grazing season started before the deposition;
- Northeastern spot, including: (i) the highly contaminated raions of the northern part of Gomel Oblast, namely Buda-Koshelevo, Dobrush, and Vetka raions (pasture grazing season started before the deposition), Chechersk and Korma raions (pasture grazing season started on April 30, 1986), and (ii) the southern part of Mogilev Oblast, including Bykhov, Klichev, Chaussy, Kostukovichy, Krasnopolie, and Slavgorod raions (pasture grazing season started on May 1, 1986), and Belynichi, Cherikov, Krichev and Mogilev raions (pasture grazing season started on May 3, 1986) where predominantly wet deposition occurred on 28–29 April 1986 (see Fig. 4); and
- Central spot, including the reminder of Gomel Oblast where predominantly dry deposition occurred on 26–27 April 1986 and the pasture grazing season started before the deposition.

Such sub-division was important in the determination of the values of R_i and $R_{f,i}$ for these three regions:

- The ratios of ground deposition densities of radionuclide i and of ^{137}Cs, R_i, were taken from Minenko et al. (2006) for all radionuclides except ^{89}Sr and ^{90}Sr. For ^{90}Sr, the ratio of ground deposition density to ^{137}Cs was estimated from activity measurements in soil performed after the accident (SCHRB, 2006). For ^{89}Sr, a ratio to ^{90}Sr of 5.4 was used (Balonov 1993).
- The values of the interception factor by grass, $R_{f,i}$, depended on the radionuclide that was considered and on the type of deposition (dry or wet). Based on measurements of radionuclides in soil and grass done in Belarus (Ulanovsky et al., 2004; Drozdovitch et al., 2006), the ratio of interception factors for radioiodine isotopes and for ^{137}Cs for dry deposition was $R_{f,i} = 2$ and for wet deposition $R_{f,i} = 1$. For all other radionuclides the interception factors were assumed to be equal to those for ^{137}Cs. These values are, in general, similar to values obtained for Western European areas distant from the Chernobyl reactor (IAEA, 2010).

- According to Drozdovitch et al. (2006) the rate of loss of activity by grass due to weathering and growth dilution was $\lambda_I^w = 0.0667\left(\mathrm{d}^{-1}\right)$ and $\lambda_{Cs}^w = 0.047\left(\mathrm{d}^{-1}\right)$ for iodine and caesium, respectively. Values of the cow's intake-to-milk transfer coefficient, TF_i, were taken from (IAEA, 2009). Values of biological half-lives and their contribution fractions for radionuclides in cow's milk were taken from Müller and Pröhl (1993). These values are generally in good agreement with those found in the relevant Russian literature, as recently reviewed by Fesenko et al. (2015).

Parameter values used to estimate the radionuclide composition in cow's milk produced in the considered region are summarized in Table 2.

The following processes were also considered in the calculations of the radionuclide composition in cow's milk:

- Milk samples were measured, on average, one day after milking because of the time needed for milk collection from either private household or collective farms, cooling of milk, and transportation to milk plants. Therefore, radioactive decay and accumulation of daughter radionuclides was considered for pairs of ^{90}Sr and ^{90}Y as well as ^{140}Ba and ^{140}La.
- Short-lived daughters, such as ^{137m}Ba, ^{103m}Rh, ^{106}Rh and ^{144}Pr, were in radioactive equilibrium with parent's radionuclides ^{137}Cs, ^{103}Ru, ^{106}Ru and ^{144}Ce, respectively, in milk samples one day after milking.
- Correction of activity concentration of ^{131}I, ^{132}Te, ^{132}I, ^{133}I and ^{136}Cs in cow's milk due to radioactive decay of one day between milking and the time of measurement.

Fig. 5 shows the relative contributions of individual radionuclides to the total activity concentration in cow's milk produced in the Southern spot. Radionuclides that contributed more than 1% to the total activity concentration in milk during the first few weeks after deposition were ^{131}I, ^{137}Cs, ^{134}Cs, ^{89}Sr, ^{136}Cs, ^{140}Ba, ^{133}I and ^{90}Sr. During the first month after deposition, ^{131}I was the main contributor to the total activity concentration in milk. Later, ^{137}Cs and ^{134}Cs became gradually more important than ^{131}I.

Fig. 6 shows the relative contributions of individual radionuclides to the total activity concentration in cow's milk from the Northeastern spot. The deposition of non-volatile radionuclides was depleted in this spot in comparison with Southern spot because of the greater distance from the Chernobyl NPP. Therefore, ^{131}I was the main contributor to the total activity concentration in milk during the first 3 weeks after the deposition. Later, ^{137}Cs and ^{134}Cs were more important than ^{131}I.

The contribution of ^{95}Zr, ^{95}Nb, ^{103}Ru and ^{106}Ru to the activity concentration of cow's milk produced in different areas in Gomel and Mogilev Oblasts was less than 0.01% during the first month after the deposition. Therefore, these radionuclides were not considered further in this paper.

2.5. Calibration coefficients for the DP-100 device

The response of the DP-100 device to a source of each type of radiation from radionuclide i was defined as:

$$\eta_i(\beta, \gamma, IC, X - ray) = \frac{N_c}{N_h}, \quad (6)$$

where η_i is the response of the device to the radiation of radionuclide i (counts per source particle), N_c is the measured count rate (counts min^{-1}), and N_h is the total number of particles emitted per minute.

The following types of radiation were considered in this study to calculate the calibration coefficient: beta-radiation, gamma-rays, internal conversion (IC) electrons, and, in some cases, X-rays. The calibration coefficient for radionuclide i was calculated as:

$$CF_i = \frac{1}{60 \cdot M \cdot (\eta_i(\beta) + p_i(\gamma) \cdot \eta_i(\gamma) + p_i(IC) \cdot \eta_i(IC) + p_i(X) \cdot \eta_i(X))} \quad (7)$$

where 60 is the number of seconds in a minute (s min^{-1}); M is the mass of the sample (g); $\eta_i(\beta)$, $\eta_i(\gamma)$, $\eta_i(IC)$, and $\eta_i(X)$ are the respective detection efficiencies calculated for beta-radiation, gamma-rays, IC electrons and X-rays from radionuclide i in cow's milk, (unitless); $p_i(\gamma)$, $p_i(IC)$, and $p_i(X)$ are the yields of gamma-rays, IC electrons and X-rays for radionuclide i, respectively (unitless). The contribution of X-rays and low-energy electrons to the calibration coefficient was negligible as these types of radiation were not able to penetrate through the end-window to reach the sensitive volume of the GM counter.

Detailed description of the calculations of the calibration coefficients for the DP-100 device can be found elsewhere (Khrutchinsky et al., 2009). The calibration coefficients, CF_i, of the DP-100 device estimated for the selected radionuclides are shown in Table 3. The calibration coefficients for ^{137}Cs and ^{144}Ce included the contributions from their short-lived progenies ^{137m}Ba and ^{144}Pr, respectively. In contrast, the calibration coefficients for other radionuclides that have progenies (^{90}Sr, ^{132}Te and ^{140}Ba) did not include the contribution from their progeny. The third geometry of measurements of milk samples by DP-100 device was used in the majority of measurements done in Belarus shortly after the Chernobyl accident.

The values of CF_i that were calculated in this study are slightly different from those estimated by Khrutchinsky et al. (2009). The difference was caused by the consideration in this study of:

- An improved one-step method of calculation of the DP-100 response, according to which the spectra of all types of radiation emitted by the radionuclide under consideration were simulated, while Khrutchinsky et al. (2009) used a two-step approach: (1) calculation of energy-dependent response of the DP-100 device to monoenergetic sources in the range from 100 keV to 3000 keV for beta and gamma radiations, and (2) convolution of these responses according to the beta radiation and gamma ray spectra of the radionuclide under consideration;

- Realistic geometry of measurement and inclusion of aluminum covering inside of the lead shielding;
- Realistic physical and chemical composition of milk: water (87.7%), lactose (4.8%), fat (3.7%), protein (3.2%), and minerals (0.6%) with a volume density of 1.028 kg L^{-1}, while the previous study of Khrutchinsky et al. (2009) considered milk as water with a volume density of 1.0 kg L^{-1};
- The response of the DP-100 device to internal conversion electrons.

2.6. Estimation of the time-dependent relative contributions of specific radionuclides in milk to the response of the DP-100 device

The time-dependent relative contribution to the DP-100 device response of the fractional activity of radionuclide *i* in cow's milk, $DR_i(t)$, was calculated as:

$$DR_i(t) = {P_i(t)}\Big/{\sum_i P_t(t)} \tag{8}$$

where

$$P_i(t) = F_i(t)/CF_i, \tag{9}$$

where $P_i(t)$ is the DP-100 device response to the fractional activity of radionuclide *i* in cow's milk (counts $\min^{-1}$ per kBq kg^{-1}).

Figs. 7 and 8 show the variation with time of the relative contribution to the DP-100 device response of the fractional activities of the radionuclides in cow's milk produced in the Southern spot and Northeastern spot, respectively. For the Southern spot, ^{131}I was the main contributor to the DP-100 device response during the first four weeks after deposition. Later, ^{137}Cs, ^{134}Cs and ^{89}Sr defined the response of the DP-100 device. For the Northeastern spot, ^{131}I was the predominant contributor to the DP-100 response during the first 2.5 weeks after deposition. Later, ^{137}Cs and ^{134}Cs became more important than ^{131}I.

The contribution of all other radionuclides considered in the study to the DP-100 response was less than a few per cent, except for ^{133}I during the first few days and ^{132}I during the first two weeks after the deposition. Therefore, only five radionuclides, namely ^{131}I, ^{134}Cs, ^{137}Cs, ^{89}Sr and ^{90}Sr, that were estimated to be the major contributors to the DP-100 device response during the first month after the accident were considered further in this paper. Although the intake of ^{131}I with locally produced cow's milk was the primary exposure pathway of radiation exposure of the thyroid gland, the contamination of milk with radiologically important isotopes of Cs and Sr also occurred to a non-negligible extent. The approach developed here can be useful for the assessment of radiation doses for epidemiological studies with other health endpoints, e.g. leukemia or breast cancer, in populations exposed to fallout from the Chernobyl accident.

2.7. Estimation of the activity concentration of specific radionuclides in milk

The activity concentration of radionuclide *i* in cow's milk was calculated as:

$$Q_i^{meas}(t) = N_{net} \cdot DR_i(t) \cdot CF_i = N_{i,net} \cdot CF_i \,. \tag{10}$$

where $Q_i^{meas}(t)$ is the activity concentration of radionuclide i in cow's milk measured on day t (kBq kg^{-1}); $N_{i,net}$ is the net count rate of the DP-100 device due to the activity of radionuclide i in cow's milk (counts min^{-1}).

2.8. Geographical pattern of cow's milk contamination with ^{131}I, ^{134}Cs, ^{137}Cs, ^{89}Sr and ^{90}Sr

The geographical pattern of the radionuclide activity concentrations in cow's milk in Gomel and Mogilev Oblasts on May 10, 1986 was reconstructed using settlement-average values of activity concentration in milk obtained from the total beta-activity measurements (eqn (10)), the time dependency given in eqn (2), and the parameter values given in Table 2. A regular (uniform rectangular) grid was used for modeling. Ordinary kriging interpolation was applied to reconstruct values at those points of the grid where numerical information was absent (Oliver, 1990). Maps of radioactive contamination of cow's milk were constructed using the MapInfo geoinformation system (PBS, 2010).

3. Results and discussion

3.1. Concentrations of ^{131}I, ^{134}Cs, ^{137}Cs, ^{89}Sr and ^{90}Sr in cow's milk derived from total beta activity measurements

Table 4 provides the raion-average concentrations of ^{131}I, ^{134}Cs, ^{137}Cs, ^{89}Sr and ^{90}Sr in cow's milk, normalized to May 10, 1986, that were derived from the results of total beta-activity measurements. The highest concentrations were found in the most contaminated raions of the Southern spot, namely Bragin, Khoiniki and Narovlya.

Figs. 9 and 10 show the geographical pattern of the ^{131}I and ^{137}Cs concentrations in cow's milk in Gomel and Mogilev Oblasts, respectively, normalized to May 10, 1986. The concentration of ^{131}I in cow's milk on May 10, 1986 was more than 100 kBq kg^{-1} in some settlements in Southern spot, while the concentration of ^{137}Cs exceeded 10 kBq kg^{-1} in the same area and in the Northeastern spot. Because of the limited number of measurements, it was not possible to reliably reconstruct the radionuclide concentrations in cow's milk for some contaminated raions of Mogilev Oblast, even with the use of the kriging interpolation approach.

3.2. Calibration coefficient of the DP-100 device

The total beta-activities of the milk samples were estimated in 1986 from the results of the measurements using the following equation:

$$Q_{1986} = CF_{off} \cdot \left(N_{dev} - N_{bg}\right) = CF_{off} \cdot N_{net}, \tag{11}$$

where Q_{1986} is the total beta activity concentration of the milk sample calculated in 1986 (kBq kg^{-1}); CF_{off} is the calibration coefficient for the mixture of radionuclides in the milk sample indicated in the official guideline for each geometry of measurement (Table 1) (kBq kg^{-1} per count min^{-1}); N_{dev} is the measured count rate of the milk sample (counts min^{-1}); N_{bg} is the background count rate (counts min^{-1}); N_{net} is the net count rate due to the activity of the milk sample (counts min^{-1}).

According to (Lyarsky, 1965; MA, 1968), the official value of the calibration coefficient, CF_{off}, was considered to be time-independent and to correspond to less than 1-y old fission products with a mean energy of beta particles of 0.3 MeV. However, the composition of radionuclides of Chernobyl origin in cow's milk changed rapidly with time after the accident because of the presence of short-lived radionuclides (see Figs. 5 and 6). Therefore, the 'real' values of the calibration coefficient of the DP-100 devices also depended on the time after deposition. Fig. 11 compares the time-dependent calibration coefficient of the DP-100 device estimated in this study for Southern, Central and Northeastern spots with the official calibration coefficient, CF_{off} = 1.0 kBq kg^{-1} per count min^{-1}. Using the official calibration coefficient (see eqn (11)) tended to overestimate by up to about 20% the measured total beta-activity, except for 3–18 days after the deposition in Southern and Central spots.

3.3. Validation of the results

As mentioned above, the BRCRCEM database contains 122 gamma-spectrometry measurements of cow's milk done between May 5 and May 31, 1986 in Gomel and Mogilev Oblasts. To validate the results obtained in this study, we compared the ^{131}I and ^{137}Cs activity concentrations in cow's milk derived from the total beta-activity measurements with those obtained from gamma-spectrometry measurements done around the same date in cow's milk produced in the same location. We found 69 pairs of samples for which a comparison could be carried out.

The mean of the ratios of ^{131}I concentration in milk derived from total beta-activity measurement to those measured by gamma-spectrometry was 1.3 ± 0.9, the median was 0.9. Fig. 12 compares the ^{131}I concentrations in milk derived from total beta-activity measurements with those measured by gamma-spectrometry. The two sets of values agreed for 93% of the pairs within a factor of 3 (shown by dashed lines) with a correlation coefficient r = 0.94. The mean of the ratios of ^{137}Cs concentrations in milk derived from total beta-activity measurements to those measured by gamma-spectrometry was 1.2 ± 1.2, the median was 0.8; the two sets of values agreed for 77% of the pairs within a factor of 3 with a correlation coefficient r = 0.84 (not shown).

There are limitations in this validation exercise. Some samples measured by the DP-100 device represented milk from collective farms while milk samples for gamma-spectrometry measurements were obtained from privately owned cows. Although there was no substantial difference (around 25–30%) in radioactivity measured in milk samples from privately owned cows and from collective farm produced in the same settlement (Savkin et al., 2004), milk from collective farms was typically a mixture of milk produced in different settlements, which belonged to the same collective farm and located up to 10 km apart. Despite these limitations, the validation exercise showed that the model, which was developed in this study to derive concentrations of ^{131}I and other radionuclides from the results of total beta-activity measurements of cow's milk, yielded results with a reasonable degree of reliability.

4. Conclusions

The Chernobyl accident on April 26, 1986 resulted in massive releases of radioactive materials into the atmosphere and contamination of large areas in Belarus, Ukraine and western Russia with significant levels of isotopes of radioiodine, radiocesium and other radionuclides. An increase of incidence of thyroid cancer in persons exposed as children and adolescents, largely due to the consumption of fresh cow's milk contaminated with ^{131}I, was one of the major health effects of the Chernobyl accident among members of the public. To evaluate the radiation-related risk of thyroid cancer in epidemiological studies, considerable efforts were made to reconstruct individual thyroid doses received by the people included in these studies. Shortly after the accident, a wide-scale survey of total-beta activity concentrations in locally produced cow's milk was performed in the most contaminated Oblasts of Belarus (Gomel and Mogilev) by means of a radiometry device.

In this study, we developed an approach to derive the activity concentration in cow's milk of ^{131}I and other radiologically important radionuclides, namely ^{134}Cs, ^{137}Cs, ^{89}Sr and ^{90}Sr, from the measurements of total beta-activity performed between April 29 and May 31, 1986. The radionuclide concentrations were estimated from the total beta-activity measurements on the basis of (1) estimations of the time-dependent radionuclide composition in milk using a radioecological model and (2) calculations of the calibration coefficients of the radiometry device for the radionuclides under consideration using Monte Carlo simulation of electron and gamma-ray transport from the milk source to the radiometry device.

Maps were prepared to show the geographical pattern of the ^{131}I and ^{137}Cs concentrations in cow's milk in Gomel and Mogilev Oblasts, normalized to May 10, 1986. At that date, the concentrations of ^{131}I and ^{137}Cs in cow's milk exceeded 100 and 10 kBq kg^{-1}, respectively, in only a few settlements. The concentrations of other radionuclides, such as ^{90}Y, ^{95}Zr, ^{95}Nb, ^{103}Ru, ^{106}Ru, ^{132}Te, ^{132}I, ^{133}I, ^{136}Cs, ^{140}Ba, ^{140}La, ^{141}Ce and ^{144}Ce, in cow's milk were 2–3 orders of magnitude smaller than those of ^{131}I.

The model was validated through a comparison of ^{131}I and ^{137}Cs activity concentrations in cow's milk derived from the total beta-activity measurements with gamma-spectrometry measurements of cow's milk produced in the same location (area) around the same date. The model enabled derivation of concentrations of radionuclides from the total beta-activity measurements of cow's milk with a reasonable degree of reliability. The developed methodology is important for the assessment of radiation doses for epidemiological studies in populations exposed to fallout from the Chernobyl accident.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

Acknowledgements

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Agency Agreement between the National Institute of Allergy and Infectious Diseases (NIAID, USA) and the NCI, NIAID agreement # DCC-OD-12-900.

Appendix A.: Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jenvrad.2020.106264.

Appendix B.: Processing the results of total beta-activity measurements that were close to, or less than, the critical level

The DP-100 device has a relatively high own background level. Therefore, for the majority of the measurements of milk samples the net count rates, N_{net}, were lower than critical level, CL, which is calculated according to (ISO, 2000) as:

$$CL = k \cdot \sqrt{N_{bg}/T_{dev} + N_{bg}/T_{bg}}, \tag{A.1}$$

where k is the quantile of the standardized normal distribution for the predetermined probability α of avoiding the error of the first kind (error of the first kind is the acceptance of background fluctuations as a useful signal with probability α); T_{dev} = 5 min and T_{bg} = 10 min are the times of measurement of milk sample and background, respectively.

The critical level means the counting rate from a sample containing the minimum significant activity, MSA, that can be reliably distinguished from background with a certain level of confidence (IAEA, 2007). In this study, the probability α of the error of the first kind was selected to be 0.05 and k = 1.65.

The lower limit of detection, LLD, is calculated according to (ISO, 2000) as:

$$LLD = CL + k \cdot \sqrt{N_{dev}/T_{dev} + N_{bg}/T_{bg}} \tag{A.2}$$

The product of the LLD and the calibration coefficient determines a minimum detectable activity, MDA. The net count rate more than LLD ensures detection of the total beta-activity in the sample with relative statistical deviation of the DP-100 device at given confidence probability.

For some measurements, net count rates equal to zero, or to less than zero, were recorded. For these measurements, the total count rate of the measured sample is not statistically different from the background count rate at a given confidence level. For these measurements, the net count rate, N_{net}, was replaced with value of 'less-than level', L_{l-t}, calculated according to (Lochamy, 1981) using the following equation:

$$L_{l-t} = \left(N_{dev} - N_{bg}\right) + k \cdot \sqrt{N_{dev}/T_{dev} + N_{bg}/T_{bg}} \tag{A.3}$$

Measurements of milk samples on May 16, 1986 collected at village Gridni (Gomel Oblast, ground deposition density of ^{137}Cs of 830 kBq m^{-2}) can serve as a typical example of using 'less-than level' concept. The database contains several measurements of background (T_{bg} =

10 min) at that date, the average background count rate of the DP-100 device was Nbg = 28 (counts min^{-1}). The measured count rate of the milk sample (T_{dev} = 5 min) was also N_{dev} = 28 (counts min^{-1}). The net count rate of milk sample was equal to N_{net} = 0 (counts min^{-1}).

The critical level for this measurement was calculated using eqn (A.1) to be CL = 4.78 (counts min^{-1}). The measured net count rate of the milk sample, N_{net}, was less than the critical level, CL. In such instances, we assumed that the measured count rate of the DP-100 device from milk sample, N_{dev}, is not statistically different from the background count rate of the device, N_{bg}, with probability α = 0.05. To estimate the response of the DP-100 device to activity concentration of the radionuclides mix in the milk sample, we used the 'less-than level' concept. The 'less-than level' was calculated using eqn (A.3) to be L_{l-t} = 4.78 (counts min^{-1}). This value was assigned in the database to be net count rate, N_{net}, and was used in this study to estimate activity concentration of ^{131}I and other radionuclides in the cow's milk samples.

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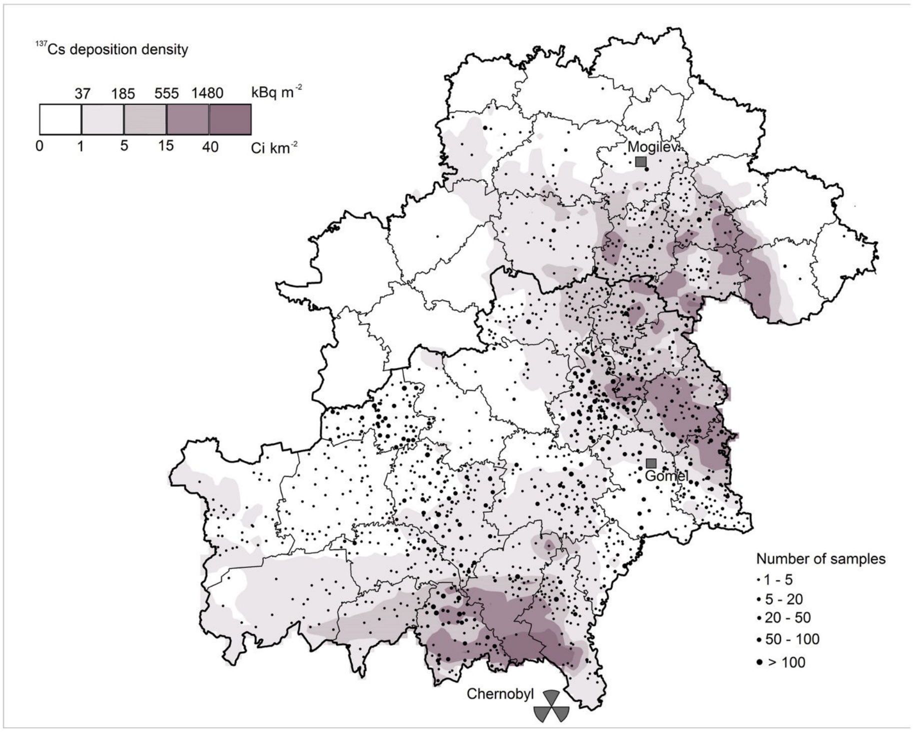

Fig. 1.

Geographical pattern of locations where cow's milk samples were taken between 29 April and May 31, 1986 in the most contaminated Gomel and Mogilev Oblasts in Belarus.

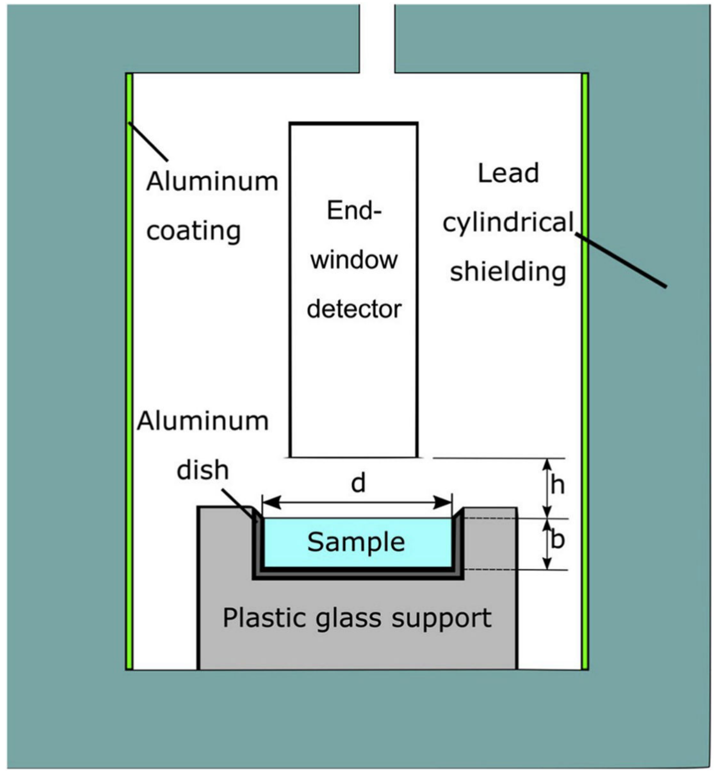

Fig. 2.
DP-100 device and geometry of measurement performed by the device. The values of *d*, *h*, and b for the three standard geometries are given in Table 1.

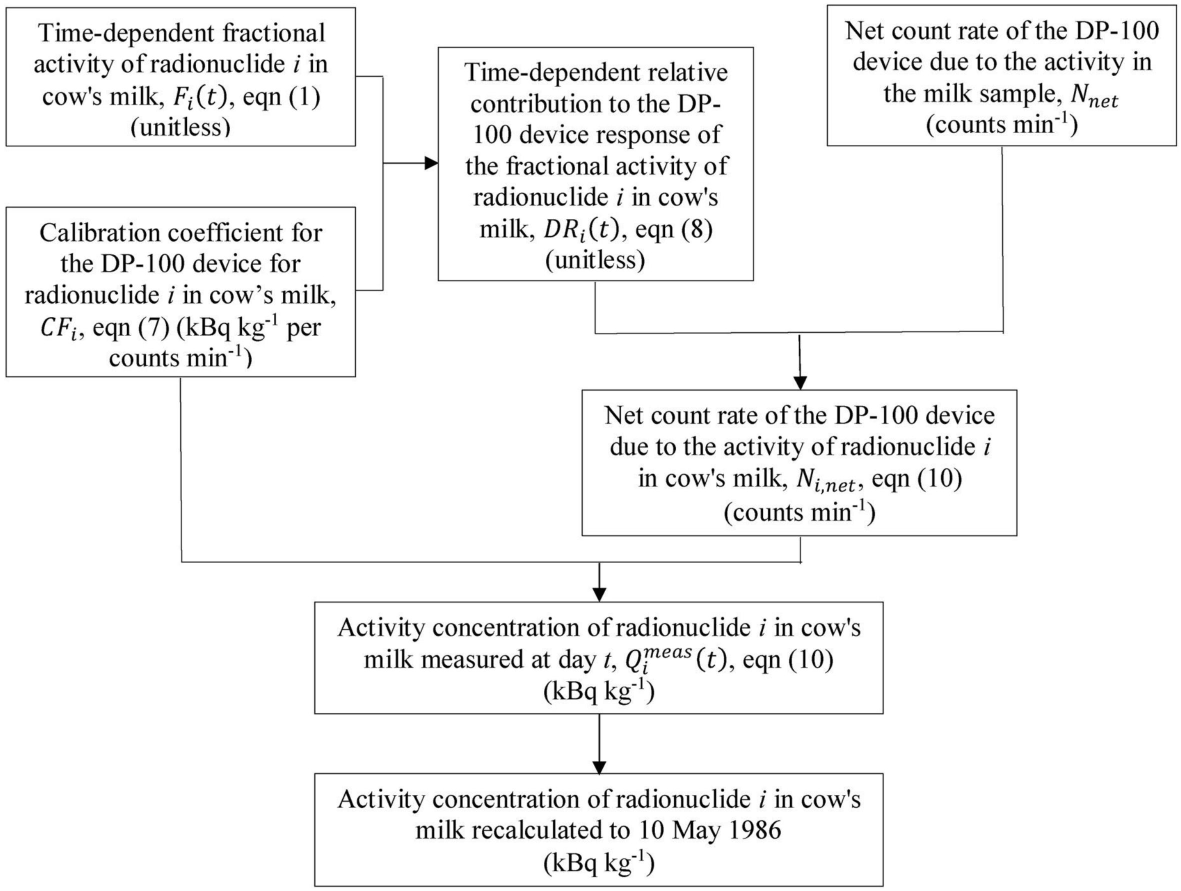

Fig. 3.

Schema of calculation of the activity concentration of a specific radionuclide in cow's milk from the results of total beta-activity measurements.

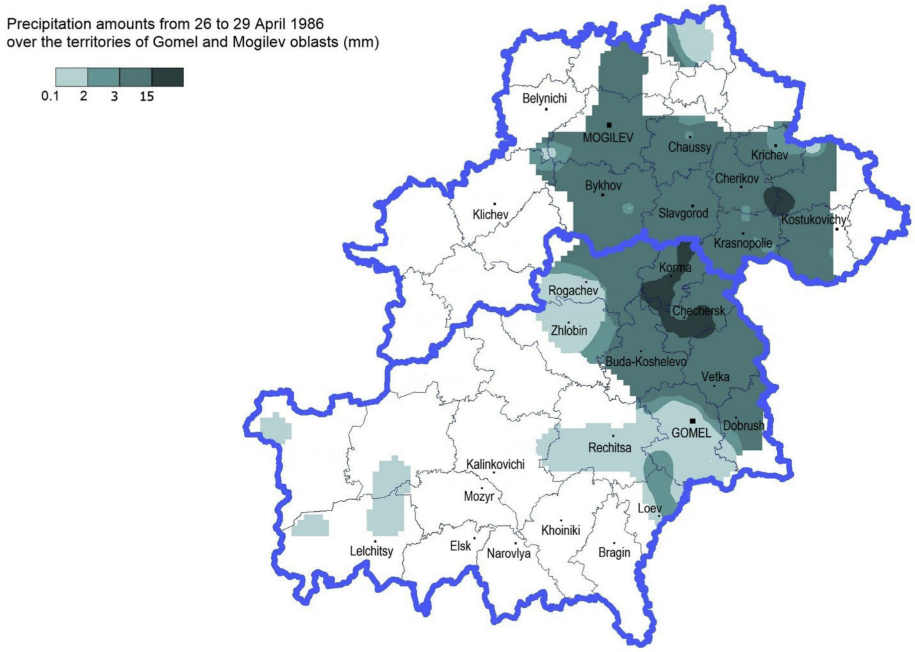

Fig. 4.

Precipitation amounts from April 26 to April 29, 1986 over the territory of Gomel and Mogilev Oblasts of Belarus (adapted from Drozdovitch et al., 2013b).

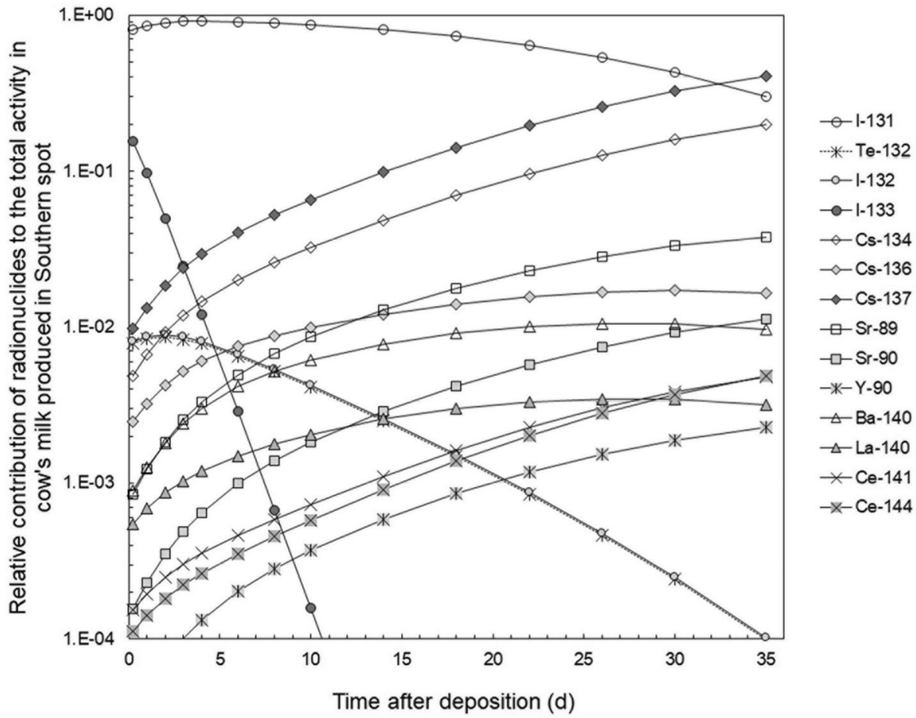

Fig. 5.

Relative contributions of individual radionuclides to the activity concentration in cow's milk calculated for Southern spot.

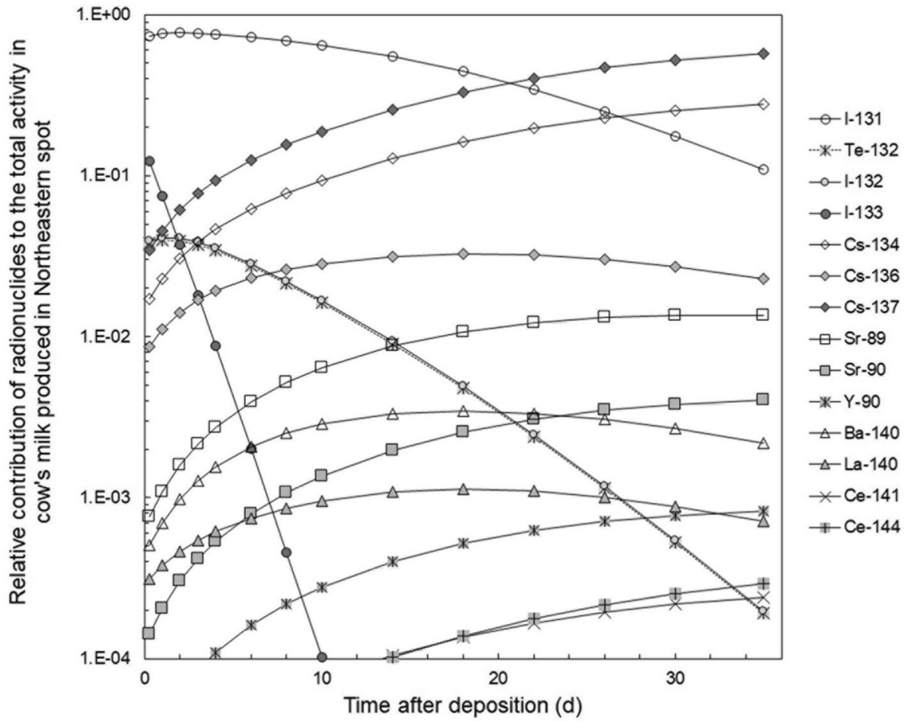

Fig. 6.

Relative contributions of individual radionuclide to the activity concentration in cow's milk calculated for Northeastern spot.

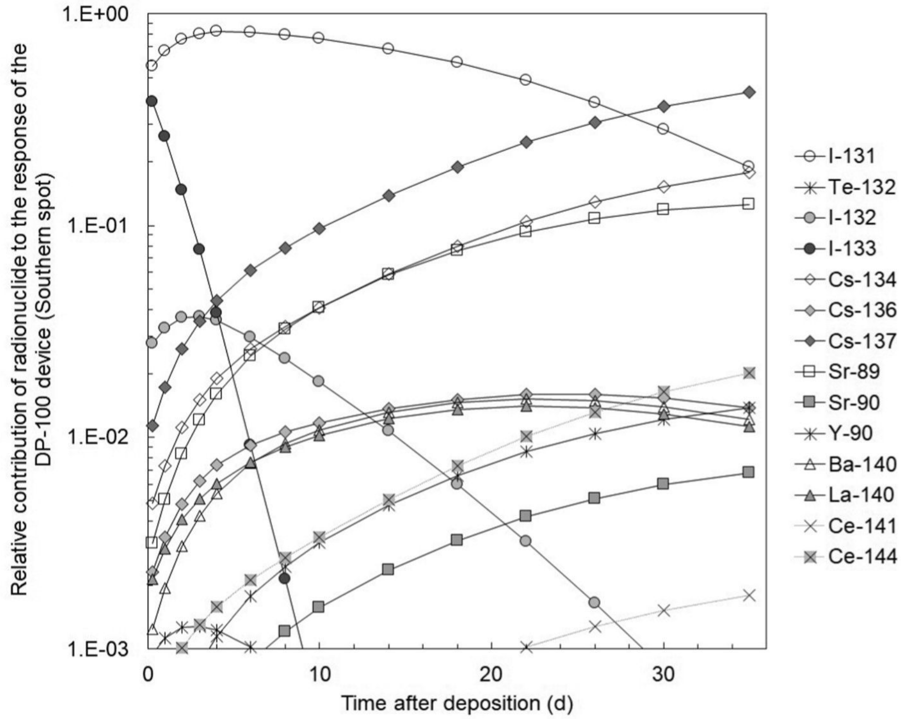

Fig. 7.

Relative contributions of radionuclide activity concentrations in cow's milk to the response of the DP-100 device calculated for Southern spot.

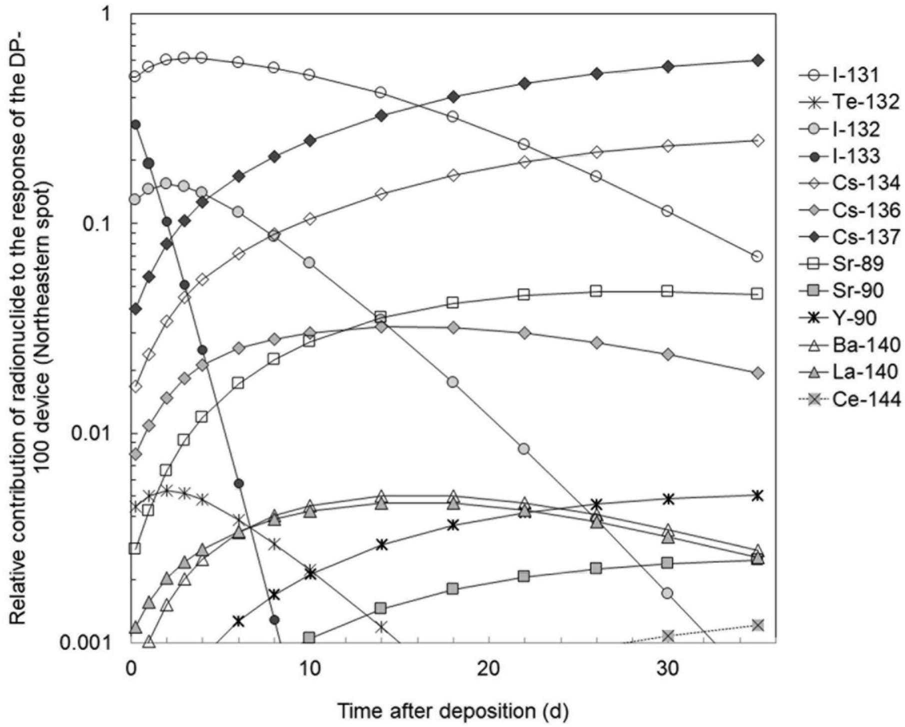

Fig. 8.

Relative contributions of radionuclide activity concentrations in cow's milk to the response of the DP-100 device calculated for Northeastern spot.

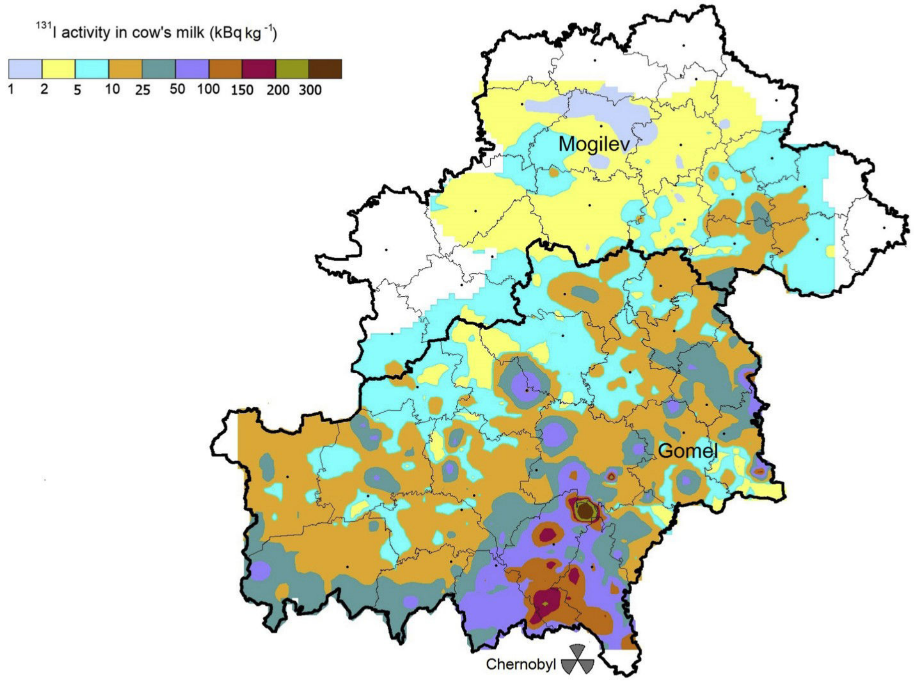

Fig. 9.

Geographical pattern of ^{131}I activity concentration in cow's milk derived from total beta-activity measurements done in Gomel and Mogilev Oblasts in Belarus (normalized to May 10, 1986).

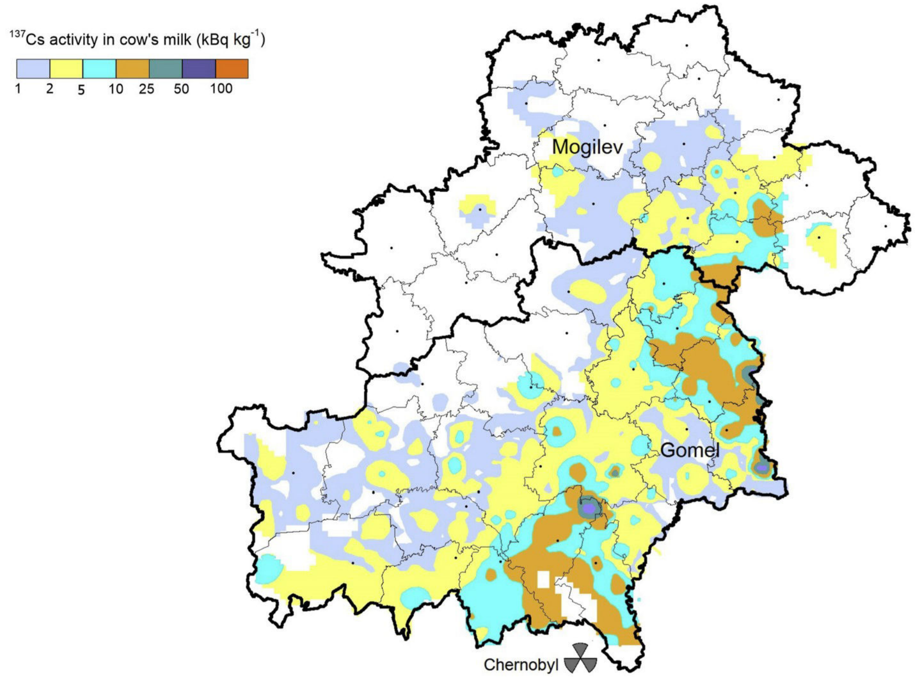

Fig. 10.

Geographical pattern of ^{137}Cs activity concentration in cow's milk derived from total beta-activity measurements done in Gomel and Mogilev Oblasts in Belarus (normalized to May 10, 1986).

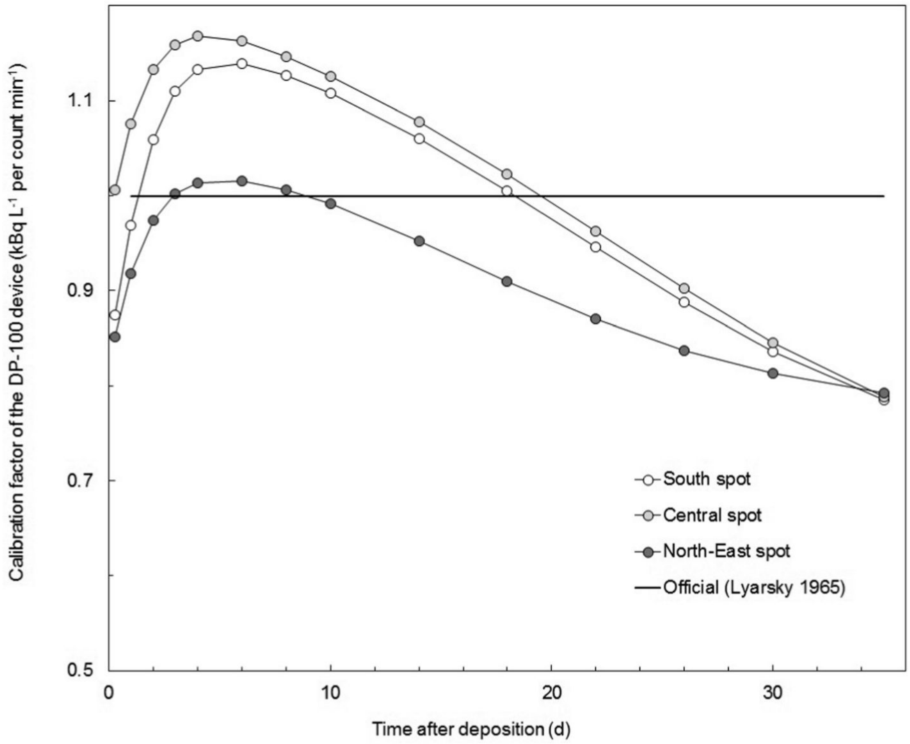

Fig. 11.

Comparison of time-dependence of calibration coefficient of the DP-100 device for third geometry of measurement for Southern, Central and Northeastern spots with official value, CF_{off}.

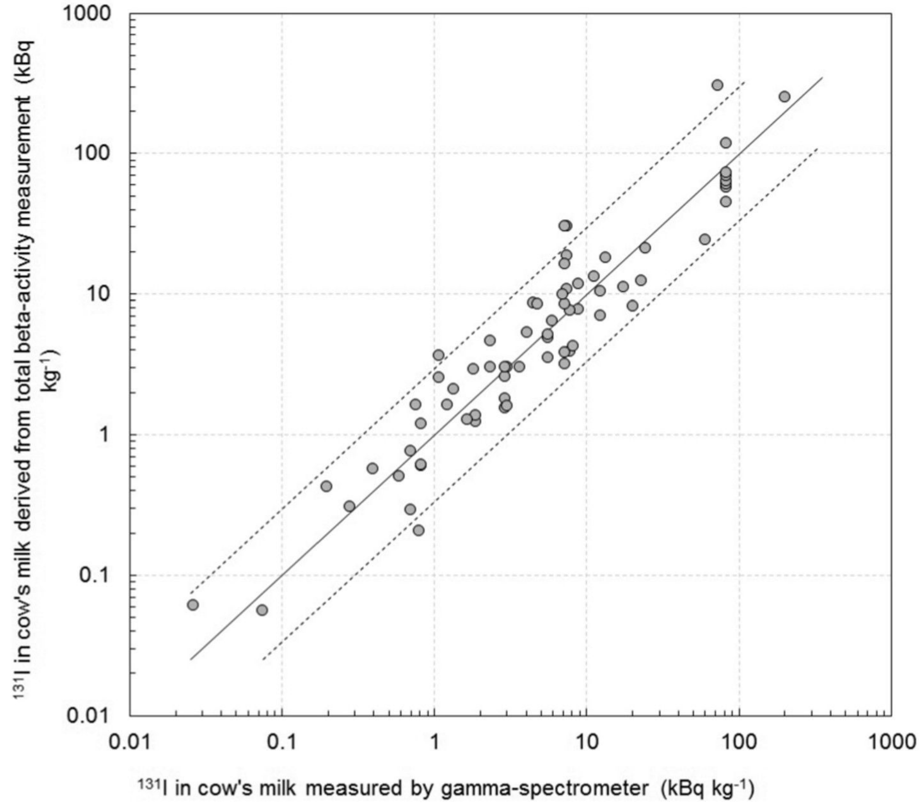

Fig. 12.

Comparison of ^{131}I activity concentrations in cow's milk derived from the total beta-activity measurements with those measured by gamma-spectrometry. Dashed lines indicate deviations from the central line by a factor of 3.

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Table 1

Standard geometries used to measure the total beta-activity in milk samples and values of official calibration coefficient for the DP-100 device, CF_{off}, used in 1986 according to (Lyarsky, 1965).

Parameter	Measurement geometry		
	#1	#2	#3
Distance between the detector and the dish, *h* (mm)	10	10	20
Depth of the dish, *b* (mm)	9	9	9
Inner diameter of the dish with milk sample, *d* (mm)	24	38	38
Diameter of GM counter, g (mm)	20	20	20
CF_{off}(μCi kg^{-1} per count min^{-1})	0.027	0.013	0.027
CF_{off}(kBq kg^{-1} per count min^{-1})	1.0	0.48	1.0

Table 2

Parameter values used to estimate the radionuclide composition in cow's milk.

	Southern spot		Central spot		Northeastern spot		Area-independent parameters						
	R_i^0	$R_{f,i}$	R_i^0	$R_{f,i}$	R_i^0	$R_{f,i}$	TF_i (d kg^{-1})	$\lambda_i^{ra}(\mathrm{d}^{-1})$	$\lambda_i^{w}(\mathrm{d}^{-1})$	$a_{i,1}$	$\lambda_{i,1}^{m}(\mathrm{d}^{-1})$	$a_{i,2}$	$\lambda_{i,2}^{m}(\mathrm{d}^{-1})$
^{131}I	16	2	21	2	8.3	1	5.4×10^{-3}	0.0864	0.067	1.0	0.99	–	–
^{132}Te	8.0	1	4.2	1	11	1	3.4×10^{-4}	0.216	0.047	1.0	0.69	–	–
^{133}I	7.5	2	5.0	2	3.4	1	5.4×10^{-3}	0.800	0.067	1.0	0.99	–	–
^{134}Cs	0.5	1	0.5	1	0.5	1	4.6×10^{-3}	9.21×10^{-4}	0.047	0.8	0.46	0.2	0.046
^{136}Cs	0.27	1	0.27	1	0.27	1	4.6×10^{-3}	0.0527	0.047	0.8	0.46	0.2	0.046
^{137}Cs	1.0	1	1.0	1	1.0	1	4.6×10^{-3}	6.3×10^{-5}	0.047	0.8	0.46	0.2	0.046
^{89}Sr	0.54	1	0.67	1	0.14	1	1.3×10^{-3}	0.0137	0.047	0.9	0.23	0.1	6.9×10^{-3}
^{90}Sr	0.10	1	0.12	1	0.026	1	1.3×10^{-3}	6.6×10^{-5}	0.047	0.9	0.23	0.1	6.9×10^{-3}
^{90}Y[b]	0.10	1	0.12	1	0.026	1	6.0×10^{-5}	0.260	0.047	0.9	0.23	0.1	6.9×10^{-3}
^{95}Zr	3.6	1	4.0	1	0.17	1	3.6×10^{-6}	0.0108	0.047	1	0.69	–	–
^{95}Nb	3.6	1	4.0	1	0.17	1	4.1×10^{-7}	0.0198	0.047	1	0.69	–	–
^{103}Ru	3.7	1	2.4	1	1.6	1	9.4×10^{-6}	0.0177	0.047	0.1	0.023	0.9	6.9×10^{-4}
^{106}Ru	0.93	1	0.85	1	0.42	1	9.4×10^{-6}	0.0019	0.047	0.1	0.023	0.9	6.9×10^{-4}
^{140}Ba	4.7	1	7.6	1	0.76	1	1.6×10^{-4}	0.0544	0.047	0.9	0.23	0.1	6.9×10^{-3}
^{140}La[c]	4.7	1	7.6	1	0.76	1	6.0×10^{-5}	0.413	0.047	0.9	0.23	0.1	6.9×10^{-3}
^{141}Ce	3.9	1	3.8	1	0.14	1	2.0×10^{-5}	0.0213	0.047	0.5	0.69	0.5	0.035
^{144}Ce	2.8	1	3.0	1	0.12	1	2.0×10^{-5}	0.0024	0.047	0.5	0.69	0.5	0.035

[a] ICRP (2008).

[b] Values of $a_{i,1}$, $\lambda_{i,1}^{m}$, $a_{i,2}$, $\lambda_{i,2}^{m}$ were assumed to be same as for ^{90}Sr.

[c] Values of $a_{i,1}$, $\lambda_{i,1}^{m}$, $a_{i,2}$, $\lambda_{i,2}^{m}$ were assumed to be same as for ^{140}Ba.

Table 3

Estimated calibration coefficients for the DP-100 device.

Radionuclide	Calibration coefficient (kBq kg^{-1} per count min^{-1}) for geometry of measurement		
	# 1	**# 2**	**# 3**
^{131}I	1.17	0.772	1.26
^{132}Te	6.64	4.36	7.23
^{132}I	0.251	0.156	0.257
^{133}I	0.336	0.219	0.355
^{134}Cs	0.883	0.534	0.876
^{136}Cs	1.03	0.574	0.933
^{137}Cs [a]	0.710	0.454	0.751
^{89}Sr	0.220	0.142	0.234
^{90}Sr	1.19	0.802	1.29
^{90}Y	0.124	0.0781	0.130
^{140}Ba	0.585	0.384	0.627
^{140}La	0.218	0.137	0.223
^{141}Ce	1.92	1.28	2.10
^{144}Ce [a]	0.184	0.114	0.190

[a] Calibration coefficient for that radionuclide includes the contribution from its short-lived progeny ^{137m}Ba and ^{144}Pr, respectively.

Table 4

Raion-average activity concentrations of ^{131}I, ^{134}Cs, ^{137}Cs, ^{89}Sr and ^{90}Sr in cow's milk recalculated to May 10, 1986 that were derived from total beta-activity measurement.

Oblast	Raion[a]	N of samples measured	Raion-average activity concentration (kBq kg^{-1})				
			^{131}I	^{134}Cs	^{137}Cs	^{89}Sr	^{90}Sr
Southern spot							
Gomel	Bragin	162	119	7.2	14	1.9	0.42
	Elsk	86	26	1.5	3.1	0.41	0.092
	Khoiniki	306	103	6.2	13	1.6	0.37
	Loev	293	20	1.2	2.4	0.32	0.071
	Narovlya	773	60	3.6	7.2	0.95	0.21
	Rechitsa	603	35	2.1	4.3	0.56	0.12
	Subtotal Southern spot[b]	4336	38	2.3	4.6	0.60	0.13
Central spot							
Gomel	Gomel	207	37	1.7	3.4	0.56	0.12
	Rogachev	357	19	0.86	1.7	0.28	0.061
	Zhlobin	68	8.5	0.39	0.79	0.13	0.028
	Subtotal Central spot[b]	1364	16	0.73	1.5	0.24	0.052
Northeastern spot							
Gomel	Buda-Koshelevo	2114	12	2.2	4.5	0.15	0.034
	Dobrush	669	17	3.1	6.3	0.21	0.047
	Vetka	439	27	5.0	10	0.35	0.076
Mogilev	Cherikov	149	9.6	1.8	3.6	0.29	0.064
	Krasnopolie	36	13	3.1	6.3	0.48	0.108
	Slavgorod	433	4.4	1.0	2.1	0.17	0.037
	Subtotal Northeastern spot[b]	4931	13	2.6	5.3	0.26	0.057
Gomel and Mogilev[b]		10,631	24	2.2	4.5	0.39	0.09

[a] Selected raions.

[b] Include all measurements done in this area.