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Influence of the external and internal radioactive contamination of the body and the clothes on the results of the thyroidal ^{131}I measurements conducted in Belarus after the Chernobyl accident. Part 1: Estimation of the external and internal radioactive contamination

Vladimir Drozdovitch,
Division of Cancer Epidemiology and Genetics, National Cancer Institute, NIH, DHHS, 9609 Medical Center Drive, Room 7E548 MSC 9778, Bethesda, MD 20892-9778, USA

Valeri Khrouch,
State Research Center - Burnasyan Federal Medical Biophysical Center of Federal Medical Biological Agency, Moscow, Russia

Sergey Shinkarev,
State Research Center - Burnasyan Federal Medical Biophysical Center of Federal Medical Biological Agency, Moscow, Russia

Yuri Gavrilin,
State Research Center - Burnasyan Federal Medical Biophysical Center of Federal Medical Biological Agency, Moscow, Russia

Victor Minenko,
Institute for Nuclear Problems, Belarusian State University, Minsk, Belarus

Arkady Khrutchinsky,
Institute for Nuclear Problems, Belarusian State University, Minsk, Belarus

Semion Kutsen,
Institute for Nuclear Problems, Belarusian State University, Minsk, Belarus

Yuri Konstantinov,
Institute of Radiation Hygiene, Saint Petersburg, Russia

Tatiana Kukhta,
United Institute of Informatics Problems, Minsk, Belarus

André Bouville,
U.S. National Cancer Institute (retired), Bethesda, MD, USA

Nickolas Luckyanov, and
U.S. National Cancer Institute (retired), Bethesda, MD, USA

Paul Voillequé

Phone: +1-240-276-7399, Fax: +1-240-276-7840, drozdovv@mail.nih.gov.

MJP, Risk Assessment, Inc., Denver, CO, USA

Abstract

The estimation of the thyroid doses received in Belarus after the Chernobyl accident is based on the analysis of exposure-rate measurements performed with radiation detectors placed against the necks of about 130,000 residents. The purpose of these measurements was to estimate the ^{131}I activity contents of the thyroids of the subjects. However, because the radiation detectors were not equipped with collimators and because the subjects usually wore contaminated clothes, among other factors, the radiation signal included, in addition to the gamma rays emitted during the decay of the ^{131}I activity present in the thyroid, contributions from external contamination of the skin and clothes and internal contamination of organs other than the thyroid by various radionuclides. The assessment of the contributions of the external and internal contamination of the body to the radiation signal is divided into two parts: (1) the estimation of the radionuclide activities deposited on, and incorporated in, various parts of the body, and (2) the responses of the radiation detectors to the gamma rays emitted by the radionuclides deposited on, and incorporated in, various parts of the body. The first part, which is presented in this paper, includes a variety of exposure scenarios, models, and calculations for 17 of the most abundant gamma-emitting radionuclides contributing to the thyroid detector signal, while the second part is presented in a companion paper. The results presented in the two papers were combined to calculate the contributions of the external and internal contamination of the body to the radiation signal, and, in turn, the ^{131}I activities in the thyroids of all subjects of an epidemiologic study of thyroid cancer and other thyroid diseases among 11,732 Belarusian-American cohort members who were exposed in childhood and adolescence.

Keywords

Chernobyl; thyroid measurement; ^{131}I activity

Introduction

During the first few weeks following the Chernobyl accident, which took place in Ukraine on 26 April 1986, radiation detectors were placed against the necks of about 130,000 residents of Belarus to derive the ^{131}I activity contents of their thyroids from exposure-rate measurements (Gavrilin et al. 1999). The results of these measurements (hereafter called ‘direct thyroid measurements’) are the foundation used to estimate the thyroid doses received in Belarus after the Chernobyl accident. The U.S. National Cancer Institute is conducting in Belarus and Ukraine a long-term cohort study of thyroid cancer among 25,000 persons exposed in childhood to radioactive fallout from the Chernobyl accident (Stezhko et al. 2004). The reconstruction of the individual doses for the cohort members is based on the ^{131}I activities in their thyroid glands that were derived from the measured exposure rates against the necks of the study subjects (Drozdovitch et al. 2013a).

It should be stressed that direct thyroid measurements were performed in Belarus in the conditions of a large-scale nuclear reactor accident using devices that were not designed to measure radioactivity in humans, mainly the DP-5 dose-rate meter and the SRP-68–01

survey meter. These devices did not measure ^{131}I activity in the thyroid gland, as it could be measured by gamma spectrometry, but exposure rate at the surface of the neck at the level of the thyroid gland. Therefore, because the radiation detectors were not equipped with collimators and because the subjects usually wore contaminated clothes, among other factors, the radiation signal included, in addition to the gamma rays emitted during the decay of the ^{131}I present in the thyroid, several components that had to be taken into consideration:

- The room background, which includes the natural background radiation and the radioactive contamination due to the Chernobyl accident of the room where the subject was measured. According to the measurement protocol, the room background should have been measured systematically. Unfortunately, in some cases, the room background was either not recorded or not measured. Estimation of the values of room background for various situations was described in (Drozdovitch et al. 2013a) and is not discussed in this paper.
- The external contamination of the body (skin and hair) and of the clothes of the subjects, which was usually the main component of the radiation signal, excluding that due to the thyroidal ^{131}I activity. The external contamination of the body and clothes depends on many factors including the indoor and outdoor concentrations of a range of radionuclides as a function of time after the accident, along with the age, personal habits, and residential history of the subject.
- The internal contamination of the body, which resulted mainly from the intakes by inhalation and ingestion of the radionuclides released during the Chernobyl accident, and, for a small part, from the uptake by blood of iodine isotopes deposited on the skin. The processes involved in the internal contamination of the body are extensively discussed in several scientific publications (e.g., NCRP 1980; Andersson et al. 2001; Cross et al. 1992; Cygan et al. 1992; Faw 1992; Gavrilin et al. 1997). The magnitude of the internal contamination depends on the residential history and dietary habits of the subject, as well as on the time elapsed since the accident.

Figure 1 presents the scheme of the thyroid dose assessment based on direct thyroid measurements. The assessment of the contributions of the external and internal contamination of the body to the radiation signal is divided into two parts: (1) the estimation of the radionuclide activities deposited externally on various parts of the body, and, internally, in various organs of the body, and (2) the responses of the radiation detectors to the gamma rays emitted by the radionuclides deposited externally on and incorporated in various parts of the body. The first part is presented in this paper, while the second part is presented in a companion paper (Kutsen et al. 2019, this issue). The purpose of this detailed assessment of the contributions of the external and internal contamination of the body to the radiation signal is to improve the individual thyroid dose estimates of all subjects of an epidemiologic study of thyroid cancer and other thyroid diseases among 11,732 BelAm screening cohort members who were exposed in childhood and adolescence (Cahoon et al. 2017, Ostroumova et al. 2013, Zablotska et al. 2011).

Materials and methods

The general characteristics of the study and the data and methods used to estimate the radionuclide activities deposited externally on various parts of the body and deposited internally in various organs and tissues will be described in turn in the following sections.

General characteristics of the study

The direct thyroid measurements were performed at any time, T, between the day of the accident (26 April 1986) and 30 June 1986, when the ^{131}I (half-life of 8.02 d) activity in the thyroid became too small to yield a signal that could be distinguished from background. As a result, the models used to estimate the external and internal contamination of the body had to provide the capability to calculate the radionuclide activities during that entire time period. In this article, the simplifying assumption is made that the study subjects did not change their residence, lifestyle, or dietary habits during the approximately two months when the direct thyroid measurements were made. Data about these behavioral aspects are available for each subject of the epidemiological study and could be used for more detailed assessments for particular individuals.

A family of phantoms, representing the newborn, children aged 1 y, 5 y, 10 y, 15 y, and adults, was used to calculate the external contamination of the body and clothes (Kutsen et al. 2019, this issue). Such a phantom is presented in Fig. 2. The gamma-emitting radionuclides that were considered in the estimation of the external contamination of the body and clothes are those that were most abundant in the atmospheric releases of the Chernobyl accident: ^{95}Zr, ^{95}Nb, ^{99}Mo, ^{103}Ru, ^{106}Ru, ^{132}Te, ^{131}I, ^{132}I, ^{133}I, ^{134}Cs, ^{136}Cs, ^{137}Cs, ^{140}Ba, ^{140}La, ^{141}Ce, ^{144}Ce, and ^{239}Np (radionuclides are listed according to increasing mass number, not according to their importance). Regarding internal contamination, the activities in lungs were estimated for each of the radionuclides listed above. In addition, the activities of ^{131}I in the thyroid and of ^{134}Cs, ^{136}Cs, and ^{137}Cs in the whole body were estimated. Table 1 provides the most important characteristics of the radionuclides that were considered in the present study.

Model used to estimate the external contamination of body surfaces and clothes

The main features related to the radioactive contamination of the body surfaces and clothes are presented in Fig. 3. Those features include: (1) the radionuclide concentrations in outdoor air; (2) the transfer of the outdoor contamination to indoor air; (3) the deposition of the radionuclides on the ground and other surfaces, both indoors and outdoors; (4) the resuspension of the deposited radioactive material; and (5) the transfer of the deposited radioactive material to the body surfaces and clothes, both indoors and outdoors. The starting point of the model is the outdoor air concentration, $DC_{137,out}(t)$, of radionuclide i at the location of interest at time t after the accident. The outcome of the model is the deposition density, $Q_{i,j}(t)$, of radionuclide i on region j of the body (see Fig. 2), which may or may not be covered with clothes, at time t after the accident and, in particular, at time T of the direct thyroid measurement.

Estimation of the radionuclide concentrations in outdoor air—The time-integrated outdoor air concentrations of ^{137}Cs during day t (from 00:00 to 24:00), $DC_{137,out}(t)$, in the settlement under consideration were derived from measurements of the total ^{137}Cs ground deposition density, Q_{137}, onto the lawn, which was selected as the environmental surface of reference (Andersson et al. 2002; Fogh et al. 1999). The values of Q_{137} (Bq m^{-2}) are available for all contaminated settlements of Belarus (Table 2). The time-integrated concentrations $DC_{137,out}(t)$, expressed in Bq d m^{-3}, were calculated using the following equation:

$$DC_{137,out}(t) = Q_{137} \cdot f_{137}(t) / \left(v_{137,lawn,dry} + v_{137,lawn,wet} \cdot R(t)\right), \quad (1)$$

where $f_{137}(t)$ is the fraction of ^{137}Cs that deposited during day t onto the lawn of the settlement (unitless). The values of the fractions $f_{137}(t)$ were estimated according to daily ^{137}Cs fallout, which were calculated for all Belarusian settlements using the model of atmospheric transport LEDI (Talerko 2005); v_{137} is the deposition velocity of ^{137}Cs (m d^{-1}) with *dry* identifying the deposition under dry conditions (no precipitation) and *wet* identifying the deposition of activity incorporated into drops of water, this process being independent of, and additive to, dry deposition. On the basis of a literature review (Chamberlain 1991; Müller and Pröhl 1993; Rosner et al. 1990; Underwood 2001), the deposition velocity for ^{137}Cs was considered to be constant and equal to 100 m d^{-1} under dry conditions and 290 m d^{-1} per mm of rain during day t under condition of wet deposition. Information presented in Table 2 on dates, time of beginning, duration and amount of precipitation, $R(t)$, collected in Belarus (Drozdovitch et al. 2013b) was used in the calculation of deposition; $R(t)$ is the amount of precipitation during day t (mm)

The time-integrated outdoor air concentrations of a radionuclide i other than ^{137}Cs during day t, $DC_{i,out}(t)$, were estimated as:

$$DC_{i,out}(t) = DC_{137,out}(t) \cdot r_{i,137}(t) = Q_{137} \cdot f_{137}(t) \cdot r_{i,137}(t) / (v_{137,lawn,dry} + v_{137,lawn,wet} \cdot R(t)), \quad (2)$$

where $r_{i,137}(t)$ is the ratio of the time-integrated concentrations of radionuclide i and of ^{137}Cs in ground level air of the settlement during day t (unitless). In the absence of measured ratios, $r_{i,137}(t)$, of the concentrations in ground level air of radionuclide i and ^{137}Cs a crude estimation was made, using the assumption that they are equal to the ratios of the activities released at the time of the accident (t=0), modified to take account of the radioactive decay during the time the radioactive cloud travelled to the settlement into consideration:

$$r_{i,137}(t) = r_{0i,137} \cdot e^{-\left(\lambda_i - \lambda_{137}\right) \cdot t}, \quad (3)$$

where $r_{0i,137}(t)$ is the ratio of the released activities of radionuclide i and of ^{137}Cs, which was obtained according to a method that is described in a later section "Estimation of the deposition of radionuclide i on a surface j of the body or clothes indoors and outdoors at day t" (unitless); and λ_i and λ_{137} are the radioactive decay constants of radionuclide i and of ^{137}Cs, respectively (d^{-1}).

Applying this procedure assumes that all radionuclides, except for the radioisotopes of iodine, have deposition velocities that are proportional or equal to those of ^{137}Cs, both under dry and wet weather conditions (Underwood 2001). Also, Eq. (3) does not apply in the case of decay chains, for example ^{140}Ba-^{140}La or ^{95}Zr-^{95}Nb; for those decay chains the following equation was used:

$$r_{i,137}(t) = r_{0i,137} \cdot e^{-(\lambda_i - \lambda_{137}) \cdot t} + r_{0ip,137} \cdot f_i \cdot \frac{\lambda_i}{\lambda_i - \lambda_{ip}} \cdot \left(e^{-(\lambda_{ip} - \lambda_{137}) \cdot t} - e^{-(\lambda_i - \lambda_{137}) \cdot t} \right), \quad (4)$$

where $r_{0i,137}(t)$ is the ratio of the released activities of the precursor of radionuclide i and of ^{137}Cs (unitless); λ_{ip} is the radioactive decay constant of the precursor of radionuclide i (d^{-1}); f_i is the branching ratio, generally equal to 1, from the precursor to the decay product (unitless).

Equation (2) could not be directly applied to the radioisotopes of iodine, which were in three physico-chemical forms: aerosol-associated, reactive gaseous (such as molecular iodine), and nonreactive gaseous (such as methyl iodide). The characteristics of the three forms were considered to estimate the ratios of these radioisotopes to ^{137}Cs in ground level air and in fallout. According to literature data (Andersson et al. 2004; Müller and Pröhl 1993; Roed 1990; Underwood 2001):

- The reactive gaseous form was assumed to be the main contributor to dry deposition;
- The ^{131}I aerosol-associated form was assumed to be the main contributor to wet deposition;
- The nonreactive form of iodine had a negligible significance in the contamination of the ground by radioisotopes of iodine. However, the nonreactive gaseous component present in the atmosphere was very significant with respect to the inhalation intake of these radioisotopes both outdoors and indoors.

In the application of Eq. (2) for these radioisotopes, each of the three forms was considered. Measured values indicate that the distribution of the physico-chemical forms of iodine in the Chernobyl radioactive cloud changed with time, distance, and weather conditions (Nedveckaite et al. 2004). Based on literature reviews (Nedveckaite et al. 2004; UNSCEAR 1998), the typical fractions of the three forms of iodine radioisotopes in air were taken to be:

0.25 for aerosol-associated iodine; 0.35 for reactive gaseous iodine; and 0.40 for nonreactive gaseous iodine.

Transfer of the contamination from outdoor air to indoor air—The time-integrated radionuclide concentration $DC_{i,in}(t)$ in indoor air was derived from its concentration outdoors according to:

$$DC_{i,in}(t) = \int_0^t FC_i(x) \cdot \lambda_{exch}(x) \cdot DC_{i,out}(x) \cdot e^{-\left(\lambda_i + \lambda_{exch}(x) + \lambda_{i,d}(x)\right) \cdot (t-x)} dx, \quad (5)$$

where $FC_i(x) \leq 1$ is the filtration factor at time x (unitless); λ_{exch} is the rate of air exchange in the building (d^{-1}); $\lambda_{i,d}$ is the rate of removal of radionuclide i from indoor air to deposition onto all surfaces inside the building (d^{-1}).

In general, all parameters in Eq. (5) are functions of time. For example, opening or closing windows in the room could change the regime of air exchange and the filtration factor, $FC_i(x)$. Flows of air in the room and the effective rate of radionuclide deposition can also be time dependent. However, because of the warm weather at that time and absence of information from the local authorities, residents kept windows open, and, therefore, all parameter values of Eq. (5) were taken to be constant during a given day t. Assuming that $exp[-(\lambda_i + \lambda_{exch} + \lambda_{i,d}) \times \Delta t] << 1$ ($\Delta t = 1$ d) and also $\lambda_i << (\lambda_{exch} + \lambda_{i,d})$, the time-integrated outdoor and indoor air concentrations during day t, $DC_{i,out}(t)$ and $DC_{i,in}(t)$ (Bq d m^{-3}) are related as follows (Roed and Cannell 1987):

$$\frac{DC_{i,in}(t)}{DC_{i,out}(t)} = FC_i(t) \cdot \frac{\lambda_{exch}}{\lambda_{exch} + \lambda_{i,d}}, \quad (6)$$

The filtration factor, $FC_i(t)$, is about 1, if there are unobstructed ways of entry of outdoor air into the building (Andersson et al. 2002, 2004; Liu and Nazaroff 2001; Roed and Cannell 1987). This was true for most of the Belarusian settlements in the late April – May of 1986.

The rate of depletion of radionuclide i due to deposition indoors, $\lambda_{i,d}$, depends on its effective deposition velocity onto horizontal and vertical surfaces in the room, $v_{i,in}$, (m d^{-1}) and on the ratio of the area of all surfaces (A) (m^2) to the volume of the room (V) (m^3) (Roed and Cannell 1987):

$$\lambda_{i,d} = v_{i,in} \cdot \frac{A}{V} \quad (7)$$

Estimates of deposition velocities, relative to those of ^{137}Cs are presented in Table 3. The radionuclides were classified into four groups (Fogh et al. 1999):

- A. Coarse particles: ^{95}Zr, $^{95m,\ 95}$Nb,^{140}Ba, ^{140}La, $^{141,\ 144}$Ce, ^{239}Np;

- B. Fine particles: ^{99}Mo, $^{103, 106}$Ru, particulate $^{131, 132,133}$I, ^{132}Te, and $^{134, 136, 137}$Cs;
- C. Reactive gaseous $^{103, 106}$Ru,$^{131, 133}$I;
- D. Nonreactive gaseous $^{131, 133}$I.

Because of its short half-life of 2.3 h, ^{132}I, as the decay product of the relatively long-lived ^{132}Te, was considered to be only in particulate form.

Considering the air exchange rate in houses to be 20 d^{-1} (Roed and Cannell 1987), a typical value of 2 m^{-1} for A/V, and the values of $v_{i,n}$ for indoor surfaces that are presented in Table 3, the ratios of the time-integrated radionuclide concentration in indoor air and of the time-integrated radionuclide concentration in ground level outdoor air were estimated from Eqs. (6) and (7) to be:

- 0.20 for group A, obtained from $\lambda_{i,d} = 80$ d^{-1}, using Eq. (7);
- 0.56 for group B, obtained from $\lambda_{i,d} = 16$ d^{-1};
- 0.14 for reactive gaseous iodine and 0.40 for reactive gaseous ruthenium (group C), obtained from $\lambda_{i,d} = 120$ d^{-1} and 30 d^{-1}, respectively; and
- 0.93 for nonreactive gaseous iodine (group D), obtained from $\lambda_{i,d} = 1.6$ d^{-1}.

Deposition of airborne radionuclides on the ground and other surfaces

Deposition during a given day, t: Taking into account Eqs. (5)–(7), the deposition density $dQ_{i,j}(t)$ (Bq m^{-2}) of radionuclide i on surface j of the body or clothes indoors and outdoors during day t can be written as:

$$dQ_{i,j}(t) = DC_{i,out}(t) \cdot \left\{ (1 - f_{in}) \cdot \left[v_{i,j,out,dry} + v_{i,j,wet} \cdot R(t) \right] + FC_i \cdot \frac{\lambda_{exch}}{\lambda_{exch} + \lambda_{i,d}} \cdot f_{in} \right. \quad (8)$$
$$\left. \cdot \left[f_{sleep} \cdot v_{i,j,sleep} + \left(1 - f_{sleep}\right) \cdot v_{i,j,in} \right] \right\},$$

where i identifies the radionuclide in particulate form, which is the case for all, except for radioisotopes of iodine, which were in three forms (see above); j identifies the contaminated body region. Three classes of body regions were considered depending on degree to which the body region was covered with clothes (see Fig. 2): (1) the open regions, in contact with the contaminated air (##1–6, 15, 16, 18, 19); (2) the semi-open regions, located near open regions, with some degree of contact with the contaminated air (##7, 8, 16, 17); and (3) the regions of skin shielded by clothes from direct contact with ambient contaminated air (##9–14); *out* and *in* stand for outdoors and indoors, respectively. The fraction of day, f_{in}, when the subject resided inside any building was taken to be 0.833 (20 out of 24 h); $f_{sleep} = 0.5$ is the fraction of sleeping time (10 out of 20 hours spent inside building).

The contamination of the shoes was an important source of indoor contamination in some circumstances (see, for example (Sextro et al. 2002)). However, it is not considered in this

article because it did not contribute significantly to the reading of the detector, which was located against the thyroid.

Estimation of the deposition of radionuclide i on a surface j of the body or clothes indoors and outdoors at day t.: Using Eqs. (2) and (8), and ^{137}Cs (i=137) as the radionuclide of reference the deposition density $dQ_{i,j}(t)$ (Bq m^{-2}) on some surface j of the body or clothes indoors and outdoors during day t can be expressed as:

$$dQ_{i,j}(t) = Q_{137} \cdot f_{137}(t) \cdot r_{i,137}(t) \cdot \left\{ \left(1 - f_{in}\right) \cdot \frac{v_{i,j,out,dry}}{v_{137,lawn,dry}} \cdot \frac{1 + \frac{v_{i,j,wet} \cdot R(t)}{v_{i,j,out,dry}}}{1 + \frac{v_{137,wet} \cdot R(t)}{v_{137,lawn,dry}}} + FC_i \right. \quad (9)$$
$$\left. \cdot \frac{\lambda_{exch}}{\lambda_{exch} + \lambda_{i,d}} \cdot f_{in} \cdot \frac{v_{i,j,in}}{v_{137,lawn,dry}} \cdot \frac{f_{sleep} \cdot \frac{v_{i,j,sleep}}{v_{i.j.in}} + \left(1 - f_{sleep}\right)}{1 + \frac{v_{137,wet} \cdot R(t)}{v_{137,lawn,dry}}} \right\}$$

Considering first the deposition of radionuclide i on lawn, Eq. (9) can be modified as follows:

$$dQ_{i,lawn}(t) = Q_{137} \cdot f_{137}(t) \cdot r_{0i,137} \cdot e^{-\left(\lambda_{0i} - \lambda_{0,137}\right) \cdot t} \cdot \frac{\frac{v_{i,lawn,dry}}{v_{137,lawn,dry}} + \frac{v_{i,lawn,wet} \cdot R(t)}{v_{137,lawn,dry}}}{1 + \frac{v_{137,lawn,wet} \cdot R(t)}{v_{137,lawn,dry}}} \quad (10)$$

Taking into account that there is a similar dependence of the deposition velocity of radionuclides in particulate form for dry deposition and for fallout with rain (Underwood 2001), it was assumed for all radionuclides, with the exception of radioisotopes of iodine, that:

$$\frac{v_{i,lawn,dry}}{v_{137,lawn,dry}} = \frac{v_{i,lawn,wet}}{v_{137,lawn,wet}} \quad (11)$$

Therefore, for all radionuclides except those of iodine in categories C and D, Eq. (10) can be simplified as:

$$dQ_{i,lawn}(t) = Q_{137} \cdot f_{137}(t) \cdot r_{0i,137} \cdot e^{-\left(\lambda_i - \lambda_{137}\right) \cdot t} \cdot \frac{v_{i,l,lawn,dry}}{v_{137,lawn,dry}} \quad (12)$$

Estimates of dry deposition velocities, relative to ^{137}Cs, $v_{i,lawn,dry}/v_{137,lawn,dry}$, were previously identified for the four categories of radionuclides (A, B, C, D) for a variety of indoor and outdoor surfaces, including urban lawn (Table 3). The deposition velocity

depends upon many parameters including, among others, wind speed, the size of the radionuclide particulates, the chemical reactivity and solubility in water of the radionuclide. International programs of comparison of parameter values for use in environmental models showed a very high range of parameter values selected by different authors (IAEA 1995, 1996, 2003a, b); however, the final results were much closer than the early ones, as the result of discussions among experts.

Values of the ratio $r_{0i,137}$ were derived from the analysis of a large number of samples of soil and grass collected in the contaminated areas of Belarus within a few months after the accident and analyzed by means of multi-channel spectrometers including semiconductor detectors. All available data related to the radionuclides under consideration (Table 2) were compiled in a database of the Republican Centre of Radiation Control and Environmental Monitoring (RCRCEM) (Drozdovitch et al. 2013b; Zhukova et al 2010). The results for a given settlement were taken into consideration if at least two independent samples were collected and measured before 30 June 1986, that is, about two months after the accident. Such criteria were applied here to have some reliable data on the decay-corrected ratio of ^{131}I and of ^{137}Cs in the settlement. In addition, the estimates of the deposition densities of radionuclides in relation to ^{137}Cs for the various regions of Belarus (Khrushchinskii et al. 2014; Minenko et al. 2006) also were taken into consideration.

The case of the cumulative deposition density of radionuclide *i* on the ground ("lawn") in a settlement at time T can be derived from Eq. (12), accounting for radioactive decay:

$$Q_{i,\,lawn}(T) = \sum_{t=1}^{T} dQ_{i,\,lawn}(t) \cdot e^{-\lambda_i \cdot t}. \quad (13)$$

Equations (11) and (12) cannot be used for the gaseous forms of radioiodine. Thus, Eq. (10) must be applied to estimate numerically the ^{131}I-to-^{137}Cs activity ratios in ground level air and in fallout. For example, in the town of Khoiniki where 15 soil samples were measured and recorded in the RCRCEM database, the average ratio of ^{131}I-to-^{137}Cs activity decay corrected to the time of the reactor accident was calculated to be 39 ± 10 (one standard deviation). As occurrence of precipitation was not recorded by the Khoiniki meteorological station from 26 April to 9 May 1986, only dry deposition needs to be considered. Based on the distribution of the three main physico-chemical forms of ^{131}I and of their associated dry deposition velocities presented in Table 3, the decay-corrected ratio of ^{131}I-to-^{137}Cs activities in ground level air was estimated to be 10.7. This is in good agreement with the ^{131}I-to-^{137}Cs activity ratio of 12.3 in the reactor at the time of the accident (UNSCEAR 2000), taking into account that the town of Khoiniki is located about 50 km away from the Chernobyl reactor and that the main fallout in that town occurred on 27–28 April 1986.

The estimated ratios of the deposition velocities of the four categories of radionuclides on various surfaces (open skin, hair, clothes, and indoor surfaces) and of the ^{137}Cs dry deposition velocity onto urban lawn are presented in Table 3. Because no information on the deposition velocities to partially or completely shielded regions of the body was available, values of 0.3 and 0.1 were subjectively estimated for the reduction factors for deposition on

semi-open skin and on skin shielded by the clothes, respectively. During the time of sleep, the sections of open skin were semi-open skin, excluding the face, and a reduction factor of 0.1 was used for the clothes at that time.

Cumulative deposition on body surfaces at the time of the direct thyroid measurement, T: The daily surface deposition densities described by Eq. (9) must be calculated for all days from 26 April onward to obtain the cumulative surface deposition density on day T of consideration, that is, when the direct thyroid measurement was performed. In doing so, the following processes contributing to the depletion of the deposited activities due to natural and of artificial cleaning must be accounted for:

- the continuous mechanical elimination from clothes and the body surfaces due to movements of a contaminated subject;
- the periodic laundry and change of the contaminated clothes;
- the daily wash of parts of skin such as face and hands several times per day, which can be reasonably considered as a continuous process;
- the periodic bathing of a subject;
- the continuous process of skin clearance by shedding the outermost skin layer (Fogh et al. 1999);
- the continuous percutaneous absorption of radioisotopes of iodine into blood.

Two components of clearance, fast and slow, were assumed in the model for each type of contaminant and body surfaces. For each day of deposition:

$$F_{fast} + F_{slow} = 1, \quad (14)$$

The cumulative deposition density on body surfaces at time T can be estimated according to:

$$Q_{i,j}(t) = \sum_{t=1}^{T} dQ_{i,j}(t) \cdot \left\{ F_{i,fast} \cdot ret_{i,j,fast}(T,t) + F_{i,slow} \cdot ret_{i,j,slow}(T,t) \right\}, \quad (15)$$

where retention functions $ref_{i,j,fast}(T,t)$ and $ref_{i,j,slow}(T,t)$ and are described by:

$$ret_{i,j,symb}(T,t) = \left[bath_{j,symb} \right]^{\left(N_{bath}(T) - N_{bath}(t) \right)} \cdot \frac{1 - e^{-\left(\lambda_i + \lambda_{i,j,symb} \right) \cdot \Delta t}}{\lambda_i + \lambda_{i,j,symb}} \cdot e^{-\left(\lambda_i + \lambda_{i,j,symb} \right) \cdot (T-t)}, \quad (16)$$

where "*symb*" is a designation for fast component, or for slow component, of clearance of the radionuclide; $bath_{j,symb}$ is a reduction washing factor, i.e. the ratio of the surface deposition densities after and before a bath or washing hands or face (unitless); N_{bath} is the

number of bathes or the number of times hands of face were washed between the time of the accident and day t or T; $\lambda_{i,j,symb}$ is the clearance rate of radionuclide i from surface j and component $symb$ of contamination (d^{-1}); $\Delta t = 1$ (d). Parameter values of the retention functions that were estimated on the basis of data provided in Andersson et al. (2002) and Fogh et al. (1999) are presented in Table 4.

The absorption from open and semi-open skin surfaces into blood was considered for gaseous radioiodine (categories C and D) deposition only. The age-dependent areas of open and semi-open sections of skin considered in the model to estimate the iodine absorption into blood were taken from a companion paper (Kutsen et al. 2019, this issue). The clearance rates from the face and hands (Table 4) are greater than those from other sections of skin because they were more frequently washed. Further discussion is presented in the section on "Models used to estimate the internal contamination of the body".

Resuspension of deposited radioactive material—It was assumed that resuspension of deposited radionuclides could only occur in the absence of rain. The concentration of secondary aerosols in ground level air, indoors or outdoors, is proportional to the surface density of the radionuclide i on the lawn (outdoors) or on the floor of a building (indoors):

$$C_{i,\,second}(t) = \alpha \cdot Q_i(t)\,. \quad (17)$$

The resuspension coefficient α was taken to be 2.0×10^{-4} m^{-1} in accordance with Besnus et al. (1996), considering data of Bad'in et al. (1980a), Izrael et al. (1990), and Voillequé et al. (2002). The value of α was assumed to be independent both, radionuclide category and of time after fallout because, as Allott et al. (1992) indicated, once deposition has occurred, most of the radionuclides rapidly lose their individual identity, as they become absorbed on the host soil particles. Also, because no large precipitation occurred in the Republic of Belarus during the time period from 26 April to 30 June 1986, the transfer of the deposited activity from the upper layer of soil to deeper layers can be assumed to be negligible and the fraction of the deposited activity that is susceptible to be resuspended can be considered to be constant during the first two months after the accident (Izrael et al. 1990).

The daily concentration due to resuspension outdoors from natural processes (Eq. (17)) can be used to estimate the corresponding concentration in air indoors according to Eq. (5). The cumulative deposition density on body surfaces can be calculated using Eqs. (8) and (15), taking only dry deposition into account. Equation (8) can therefore be applied both to the primary radioactive material present in the radioactive plume emitted during the accident and to the secondary aerosols due to resuspension outdoors. However, the physical and chemical properties of the primary and secondary radionuclides are different. For example, the sizes of the secondary aerosol particles are larger than those of primary particles for radionuclides of volatile groups B, C, and D. Also, three forms of radioiodine are considered for the primary radioactive cloud while no gaseous radioiodine is considered for resuspension. Consequently, the primary and the secondary radionuclides have different deposition velocities to indoor surfaces. All secondary aerosols due to resuspension were classified in group A ("coarse").

The resuspension of radioactive material due to the outdoor activities of the subject was also considered. The surface contamination model used for this process is based on measurements described in detail elsewhere (Schwendiman 1958). The daily contamination of any section of body located at height h_j bove ground was estimated as:

$$dQ_{i,j}(t) = Q_{i,lawn}(t) \cdot P(H) \cdot e^{k_r \cdot [1 - SF_{size} \cdot (h_j/H)]}, \quad (18)$$

where k_r =2 is a parameter normalizing the general gradient of concentration from ground (outdoors) or floor (indoors) to standard height H; SF_{size} is a scaling factor that takes the age of the subject in account. Assuming that the proportions of the body do not change with the age of the child, the scaling factor can be estimated as the ratio of the child height and of the adult male height. Based on the results of measurements of height performed in 1986–1995 on approximately 450,000 Belarusians of different ages, evaluation of SF_{size} with age x in years is described by a purely empirical equation: $SF_{size} = -0.0013x^2+0.0585x+0.373$, ($r$=0.998); $P(H)$ is the daily probability (d^{-1}) for the contaminant particles to be deposited on surface level H above ground. For H = 1 m, the value of $P(H)$ = 0.04 d^{-1} according to Schwendiman (1958).

Transfer of the deposited activity on indoor surfaces to clothes and body sections—The transfer of radionuclide i during day t from a contaminated region j onto another region k of the body can be estimated as follows:

$$Q_{i,j,k}(t) = Tr_{i,j,k} \cdot Q_{i,j}(t), \quad (19)$$

where $Tr_{i,j,k}$ is the mean fraction of radionuclide i transferred from a contaminated region j onto another region k of the body.

The $Tr_{i,j,k}$ values vary in a wide range according to the type of surface, the particle size, the disturbance causing the transfer, and other factors. Experimental data presented for 5–10 μm particles (Andersson et al. 2002) show that the dry transfer fraction from some surface (cotton, wood) to skin can be estimated to be about 0.2 with a coefficient of variation of about 0.5. This figure agrees with the value of 0.20 ± 0.02 (one standard deviation) estimated for the transfer of radionuclides deposited on metal, plastic, or another smooth surface to dry smears taken five times from the same contaminated surface (Bad'in et al. 1980b). There is variation of the transferred fraction with particles size (Andersson et al. 2004): transfer was found to increase when the particle diameter decreased from 5 μm to 0.5 μm, with a maximum value of about 0.6 for diameters in the range from 0.5 to 1 μm. For the purposes of this study, values of 0.2, 0.06 and 0.01 were selected for $Tr_{i,j,k}$ for the radionuclide transfer from the body region of initial deposition to open, semi-open region and region of the body shielded by clothes from direct contamination, respectively.

Models used to estimate the internal contamination of the body

The modes of radionuclide intake that are considered to estimate the internal contamination of the body are: (1) inhalation, (2) milk consumption, and (3) skin absorption. Figure 4 shows the general scheme of internal contamination that was used. For intakes by inhalation, activities in the thyroid were calculated for ^{131}I and activities in lungs for the 17 radionuclides considered for external irradiation, whereas for intakes by ingestion activities in the thyroid were calculated for ^{131}I and activities in the whole body for the radiocaesium isotopes (^{134}Cs, ^{136}Cs, and ^{137}Cs). Activities in the thyroid due to transfer to blood of activities deposited on the skin were only calculated for ^{131}I.

Inhalation—The radionuclide concentrations in outdoor air were derived from Eqs. (1) and (2), while the indoor air concentrations were calculated using Eq. (5). The value of indoor occupancy was taken to be 0.833, i.e. four hours of the day spent outdoors and 20 hours inside buildings (home, school, office, etc.). For all of the considered radionuclides, the activity intakes were calculated using age-dependent breathing rates for different levels of physical activity (ICRP 1994).

Activities in lungs.: The activities of the inhaled radionuclides in lungs were calculated using the parameter values for deposition, retention in lung and absorption into blood of radionuclides inhaled in particulate form that are recommended in ICRP Publication 66 (ICRP 1994). By analogy with Fogh et al. (1999), the coarse aerosols (group A) were characterized by an AMAD of 5 μm, the fine aerosols (group B) were assumed to have an AMAD of 1 μm, and the geometric standard deviation (GSD) was taken to be 2.5 for both groups of aerosols (ICRP 1994). Radioisotopes of iodine and caesium were related to absorption type F (fast), radioisotopes of Zr, Ba, Te, La and Np to type M (moderate), and radioisotopes of Mo, Ru, and Ce to type S (slow).

Activities in the thyroid.: The absorption of inhaled gaseous radioiodine into blood was estimated according to ICRP Publication 71 (ICRP 1995). Age dependent uptake from blood by the thyroid and retention of radioiodine in the thyroid were estimated according to ICRP Publication 56 (ICRP 1990). Although the areas of Belarus affected by the Chernobyl accident had been a territory of mild to moderate iodine deficiency (VanMiddlesworth 2002), the uptake of iodine from blood by the thyroid was taken to be 30%, on average, as suggested by the ICRP Publication. It should be noted that it is challenging to consider the iodine deficiency around the time of the Chernobyl accident in the thyroid dosimetry because data on iodine intake for this time period are unavailable. Use of the thyroidal uptake recommended by ICRP is indirectly supported by the thyroid-mass values for Belarusian children, which were derived from measurements of thyroid volumes conducted in 1992–1996 by Sasakawa Memorial Health Foundation (Skryabin et al 2010). The results obtained there are consistent (within 30%) with the ICRP reference thyroid-mass values, for the majority of age groups.

Activities in other organs and tissues.: The activities in other organs and tissues were not calculated as they were much smaller than those in thyroid (for ^{131}I) or in lungs (for any of the other considered radionuclides).

Ingestion (cow's milk consumption)—Milk consumption was for most BelAm cohort members the predominant pathway of intake for ^{131}I and radiocaesium isotopes (^{134}Cs, ^{136}Cs, ^{137}Cs). The radionuclide concentrations in cow milk were estimated in accordance with the model ECOSYS-87 (Müller and Pröhl 1993), taking into consideration the pasture conditions in Belarus at the end of April and the beginning of May 1986:

- The radionuclide ground deposition density onto grassland was taken to be the same as that for "lawn" in each settlement.
- The pasture grass mass interception factor was taken to be 0.5 m^2 kg^{-1} (fresh mass) for dry deposition of particulate caesium and for all forms of iodine.
- The removal rate of iodine and caesium from grass due to weathering and gross dilution was taken to be 0.064 d^{-1}.
- The consumption rate of pasture grass by cows was based on the date when cows were first put on pasture in the considered settlement. The maximum pasture grass consumption by cow, M_{max}, was taken to be 40 kg d^{-1} (fresh weight) for a fully-grown pasture. However, a linear ramping up to the maximum was assumed for the consumption rate during the first week when the cows were put on pasture (Dreicer et al. 1990; Spirin and Isamov 1996):

$$M_{grass}(t_0 + TG > t \geq t_0) = M_{min} + (M_{max} - M_{min}) \cdot \frac{t - t_0}{TG - t_0}, \quad (20)$$

where M_{grass} is the daily pasture grass consumption by the cow during the ramping up period after the first day of the pasture season (kg d^{-1}); t_0 is the first day of pasture counted from 26 April 1986 (d); $TG = 7$ is the total duration of the ramping up period (d); $M_{min} = 4$ is the mass of pasture grass consumed by the cow during the first day of pasture (kg d^{-1}).

- The transfer factor from grass to cow's milk was taken to be 0.003 d L^{-1} both for iodine and for caesium (Müller and Pröhl 1993). A one-exponential model was taken for decrease of iodine concentration in milk (biological transfer rate of iodine was taken to be 1.0 d^{-1}). However, the two-exponential model of ECOSYS-87 was applied to caesium in cow's milk: the fraction of the fast component of caesium in milk with transfer rate 0.46 d^{-1} was taken to be 0.80, while the transfer rate of the slow component of caesium in milk was taken to be 0.046 d^{-1}.
- The calculated ^{131}I concentration in commercial cow's milk was restricted to 3,700 Bq L^{-1} in accordance with an order of Ministry of Health of the USSR (MH 1986). The radiocaesium concentration was decreased proportionally to radioiodine. There was no restriction for private cow's milk.
- The daily cow's milk consumption by people was taken to be 0.5 L d^{-1} for any age to facilitate the comparison with other pathways.

Activities of ^{131}I in the thyroid and of the radiocaesiums in the whole body.: The uptake and retention of radioiodine in the thyroid were estimated similarly to inhalation. Age-dependent two- or one-exponential retention function of whole body caesium was taken according to ICRP Publication 56 (ICRP 1990).

Skin absorption (for radioiodine isotopes only)—The percutaneous way of radioiodine intake was significant for gaseous iodine deposited on skin and taken to be negligible for the particulate forms. The absorption of any contaminant from the skin surface into blood depends on many factors (Ho 2002), including:

- the water- and oil-solubility of the contaminant,
- the pH index of the skin surface,
- the thickness of the outer layer of skin,
- the fractional area of hair follicles and of sweat ducts per unit area of skin, and
- the length of hair follicles and of sweat ducts.

According to Harshe et al. (1980), only 16% of iodine was found to be transferred from skin to blood six days after contamination by soluiodine (a complex of iodine with nonionic surfactant). This corresponds to an absorption rate of around 0.03 d^{-1}. However, about 90% of $Na^{131}I$ was found to be absorbed under the same conditions that correspond to absorption rate of around 0.4 d^{-1}. Most of the factors listed above are age- and gender dependent. Although a small number of measurements were made on human adult volunteers, the estimates of the absorption rate of iodine from skin to blood are associated with large uncertainties.

The application of 8 mL of 2% iodine tincture to skin was found to be an acceptable alternative to the ingestion of a KI pill (130 mg of stable iodine) to block the thyroid, because of the high rate of absorption of iodine from skin to blood (Miller et al. 1989). Unfortunately, the absorption rate was not estimated directly. However, measurements of stable iodine concentration in serum were done 26 hours after administration. The concentration in serum was lower by a factor of about 2 for cases of skin application in comparison to KI pill ingestion. Thus, the absorption rate should be estimated to be about to the limit 0.16 d^{-1}, if the whole blockage of thyroid was provided during all the time up to 26-h point of sampling and the clearance rate of blood was equal 1.9 d^{-1} according to ICRP Publication 56 (ICRP 1994) for blood-to-urine removal. For the same data, another absorption rate limit should be estimated to be about 0.22 d^{-1}, if the thyroid had not any blockage and the clearance rate of blood was 2.7 d^{-1} according ICRP Publication 56 (ICRP 1990). Other studies (e.g., Harrison 1963; Tas and Feige 1958; Zarković et al. 1965) showed that from 0.1 to 8.6 percent of ^{131}I from $Na^{131}I$ solution, which was applied on skin, passed skin barrier and could be detected in thyroid and urine. Following the review of several studies, Il'in et al. (1972) concluded that the radioiodine intake resulting from the skin-to-blood pathway is a few percent of the intake from inhalation of radioiodine vapor.

It is obvious that there are large methodical uncertainties in the studies aiming at the estimation of the contribution of percutaneous absorption to the intake of radioiodine

following exposure to a volume of contaminated air. Finally, considering the available experimental data, the absorption rate of gaseous iodine from skin to blood was tentatively estimated to be a relatively slow process with a central value of 0.25 d^{-1}.

Activities in the thyroid.: Age dependent uptake from blood by the thyroid and retention of radioiodine in the thyroid were estimated according to ICRP Publication 56 (ICRP 1990).

Results and discussion

The purpose of this article is to develop a methodology of evaluation of the external and internal contamination of 11,732 Belarusian-American cohort members at the time of their direct thyroid measurements. Because (a) the cohort members were children from 0 to 18 years old, so that a range of body sizes (for external contamination) and age-dependent parameter values (for internal contamination) had to be considered; and (b) many cohort members changed places of residence several times between the date of the accident and the date of the direct thyroid measurement, it is not possible to present individual results for every cohort member. Instead, in this paper, only example results will be given using several common assumptions:

- Constant residence in the same settlement from 26 April until the day of the direct thyroid measurement.
- Direct thyroid measurements performed at 12:00.
- Baths taken by the subjects on each Saturday at 19:00.
- Two types of conditions were considered regarding the contamination of the clothes at the time of the direct thyroid measurement:
  - **a.** the subject wore contaminated clothes that had not been washed since the accident; the decrease of their contamination was only due to natural processes;
  - **b.** the subject wore new, uncontaminated, clothes.
- Two sources of cow's milk were considered:
  - **a.** private (family cow), and
  - **b.** commercial (shops).
- No restriction was considered for the contamination of milk from private cows. A restriction on the ^{131}I concentration in commercial milk to a level below 3,700 Bq L^{-1} was considered to have started on 7 May 1986. If the calculated ^{131}I concentration in milk was higher than the limit, the ^{131}I concentration was considered to be 3,700 Bq L^{-1}, and the concentration of other radionuclides (isotopes of caesium) was decreased proportionally by the ratio of the calculated ^{131}I concentration and of 3,700 Bq L^{-1}.

Examples of results obtained for the residents of the town Khoiniki (administrative center of Khoiniki raion located in the southern part of Gomel Oblast) are given below. It should be

noted that similar calculations were done for all rural and urban populations of the most contaminated areas of Belarus.

Application of the method for residents of the town of Khoiniki

Detailed meteorological and environmental data are available for the town of Khoiniki (Drozdovitch et al. 2013b). Numerous results of measurements of radionuclides in soil, grass and milk samples collected in May-June 1986 allowed for obtaining reliable estimates of radionuclides mixture in ground deposition from the Chernobyl accident. The ^{137}Cs ground deposition density resulting from the accident was estimated to be 1.44×10^{5} Bq m^{-2}. This degree of contamination was not high enough to justify the relocation of its inhabitants soon after the accident, but it resulted in substantial levels of external and internal contamination. The daily fractions of the total ^{137}Cs fallout were 0.350 on 27 April, 0.548 on 28 April, 0.102 – 29 April, and 1.4×10^{-5} on 30 April 1986; no primary deposition occurred after 30 April 1986. As no precipitation was recorded from April 26 through 3 June 1986 by the meteorological station located in Khoiniki, only dry deposition was considered.

The modeled radioactive contamination of the body surface regions (#1 and #18) and clothes (Bq m^{-2}) normalized to ^{137}Cs ground deposition density (Bq m^{-2}) is shown in Fig. 5. The body surface contamination occurred both indoors and outdoors. Options “no washing”, “weekly washing”, and “washing twice daily” are presented to illustrate the periodic effect of washing. This effect is not visible for regions of the body other than the face because the fraction of fine aerosols in the primary contamination was high. It is important to note that the decrease in the primary contamination, presented in Fig. 5, was only due to the component of slow clearance after 30 April. Later on, the decrease in the contamination was just the same both for people who relocated to Khoiniki and for inhabitants who permanently resided in Khoiniki.

Examples of results obtained using the models of external and internal contamination for a 5-years old resident of the town Khoiniki are presented in Fig. 6: the activity of the radionuclide mixture deposited on the skin of the front quarter of the neck (external contamination) is compared with the ^{131}I activities in the thyroid (internal contamination) resulting from intakes via inhalation, percutaneous absorption, consumption of private cow’s milk, and consumption of commercial cow’s milk. As expected, the highest ratios are observed in early May, at a time when the external contamination due to short-lived radionuclides had decreased substantially while the ^{131}I activities had accumulated in the thyroid.

Validation of calculations by comparison with measurements

Measurements of external contamination of contaminated clothes and of various parts of the human body that could have been made in Belarus after the Chernobyl accident have not been found. Therefore, the models for external contamination cannot be empirically verified. However, measurements of internal contamination of the thyroid and of the whole body on the same group of people are available; they were used to validate the models of internal contamination.

Gamma-spectrometry measurements of ^{131}I in thyroid and ^{137}Cs body burden by means of stationary whole body counter were performed from 5 May through 25 June 1986 at the Institute of Radiation Hygiene (IRH) in Saint-Petersburg on individuals who arrived from the territories most contaminated following the Chernobyl accident (Lebedev and Yakovlev 1993). The measurements were made on thoroughly washed individuals wearing uncontaminated clothes. Figure 7 shows the activity ratios of ^{131}I in thyroid to ^{137}Cs body burden, either calculated using the models described in this paper or derived from the measurements performed among 56 Belarusians who came to Saint-Petersburg from Khoiniki or neighboring most contaminated raions of Gomel Oblast at various times after the Chernobyl accident.

As can be seen from the figure, a rather wide variability of ratios of ^{131}I activity in thyroid to ^{137}Cs body burden is observed among the measured individuals. There is a systematic difference within a factor of 1.5–2 between the trend line of measured ratios and the results calculated using the models. It should be noted that most of the measured individuals came to Saint-Petersburg from rural settlements of Khoiniki or other raions whereas the calculations apply to the residents of the town of Khoiniki. The differences between the measured and the modeled values may be due to difference in radioactive contamination between these settlements, as well as to deviations from model assumptions on behavioral habits and foodstuff-consumption rates, including self-limitation of the consumption of locally produced foodstuffs. However, the ratios of ^{131}I activity in thyroid to ^{137}Cs body burden calculated using the models are within the values of the ratios measured at different dates.

The results obtained using the models described in this paper were also compared with those obtained by Ulanovsky et al. (2004). Very similar models were used in the two studies with the notable exception of the ratio of ^{131}I-to-^{137}Cs activity in deposition, which was taken to be 39 in the present study while a value of 16 was used by Ulanovsky et al. (2004). Increasing the results of Ulanovsky et al. (2004) by a factor of 2.4 (=39/16) would results in a good agreement between two studies (see Fig. 7).

Influence of the external and internal radioactive contamination of the body on the results of the thyroidal ^{131}I measurements

The models described above were developed to estimate the levels of external and internal contamination for all Belarusian cohort members at the time of their direct thyroid measurement. Models of external contamination were used to estimate the activities of ^{95}Zr, ^{95}Nb, ^{99}Mo, ^{103}Ru, ^{106}Ru, ^{132}Te, ^{131}I, ^{132}I, ^{133}I, ^{134}Cs, ^{136}Cs, ^{137}Cs, ^{140}Ba, ^{140}La, ^{141}Ce, ^{144}Ce, ^{239}Np on all surfaces of the body, which was divided into 19 regions, including face, hair, neck, arms, hands, and trunk. The activities were calculated for each day between 26 April and 30 June 1986 for individuals of different ages: newborns, children aged 1, 5, 10, and 15 y; and adults. Sixteen areas in Belarus with different scenarios (wet / dry) and radionuclide mix in deposition were considered in the calculations. Twelve different scenarios of residence were considered in the study, including permanent residence at the settlement and relocation from the settlement of residence on 26, 27, 28, 29, 30 April or 2, 5, 10, 15, 20, or 25 May 1986. The results presented in this and a companion paper (Kutsen et

al. 2019, this issue) were combined to calculate the contributions of the external and internal contamination of the body to the radiation signal of thyroid detectors. For the purposes of the dose assessment, it was assumed, because the majority of measurements were made two weeks or later after the accident, that the cohort members at the times of the direct thyroid measurements wore clothes that had been washed every week.

Figure 8 shows, as an example, the variation with time of the contribution of thyroidal ^{131}I activity to the exposure rate measured near the thyroid among persons of different ages who resided in Khoiniki without relocation. This example is given for a measurement made by means of a SRP-68–01 device among persons who consumed private cow's milk. As can be seen from the figure, according to the model calculations, ^{131}I activity in thyroid contributed more than 75% to the response of thyroid detector from 4 May through 18 May 1986 when the majority of the BelAm cohort members were measured in Belarus (Drozdovitch et al. 2013a).

As another example, the variation with time of the relative contributions to the exposure rate measured by the SRP-69–01 and the DP-5 devices near the thyroid of the external contamination of the subject, of his or her internal contamination, and of the ^{131}I activity in his or her thyroid, is presented in Fig. 9 (reproduced from Drozdovitch et al. (2013a) for the SRP-68–01 device). In this example, the measurement is made on a five-year-old child who resided in Khoiniki raion, which was the raion most populated with BelAm cohort subjects. As can be seen from the figure, if the child was measured between 5 May and 20 May 1986, the relative contributions of the ^{131}I activity in the thyroid to the signal measured near the thyroid were around 80% and 70% for the SRP-69–01 and the DP-5 devices, respectively. For measurements that took place at the beginning of June 1986 or later, ^{131}I in the thyroid contributed less than half to the device response, the remainder of the signal being caused mainly by the caesium radioisotopes incorporated in the body. At all times, the contribution from external contamination was relatively low (about 20% in this example).

Summary and conclusions

Reliable estimates of thyroidal ^{131}I activity derived from the results of direct thyroid measurements are very important for the estimation of individual thyroid doses and, consequently, of the risk of radiation-related thyroid cancer and other thyroid diseases resulting from the Chernobyl accident. Unfortunately, the direct thyroid measurements made in Belarus within the first few weeks after the Chernobyl accident were of poor quality and fraught with methodological problems due to the very large numbers of measurements that had to be administered in a very short time by a limited number of radiation professionals and appropriate radiation detectors. An important issue was the contamination of the persons that were measured, resulting in an overestimation of the thyroidal ^{131}I activities derived from the direct thyroid measurements. This paper presents a detailed assessment of the evaluation of the external and internal contamination of the Belarusian subjects of an epidemiological study at the time of their direct thyroid measurement.

The models used to estimate the external contamination include (a) the estimation of the radionuclide concentrations in outdoor air; (b) the transfer of the outdoor contamination to

indoor air; (c) the deposition of the radionuclides on the ground and other surfaces, both indoors and outdoors; (d) the resuspension of the deposited radioactive material; and (e) the transfer of the deposited radioactive material to the body surfaces and clothes, both indoors and outdoors. Models of internal contamination also were used to estimate (a) the activities of ^{131}I in thyroid and of 17 radionuclides in lungs following intakes by inhalation; (b) the activities of ^{131}I in thyroid and of ^{134}Cs, ^{136}Cs, and ^{137}Cs in the whole body following intakes due to consumption of contaminated cows' milk; and (c) the activities of ^{131}I in thyroid following absorption into blood of radioiodine deposited on the skin. The uncertainties in the estimated activities were not estimated in a quantitative manner, because of the complexity of the models, the large variability of some of the parameter models according to poorly known environmental conditions (for example, the radionuclide composition in fallout or their deposition velocities), and the numerous simplifying assumptions that were made (for example, on the lifestyle of the subjects). However, the partial validation of the results related to internal contamination shows that the estimates obtained in this paper are reasonably reliable. It is subjectively estimated that the overall uncertainties on the radionuclide activities in skin, clothes, and in the body are within a factor of 2 to 3 around the central estimates.

The results presented in this and a companion paper (Kutsen et al. 2019, this issue) were used to eliminate the contributions of the external and internal contamination of the body from the radiation signal of thyroid detectors, and, in turn, to improve the estimates of the ^{131}I activities in the thyroids of all subjects of the epidemiologic study of thyroid cancer and other thyroid diseases.

The models described in this paper, as well as the results of their application, constitute a resource that could be applied on data obtained after the Fukushima accident and could be also used in case of future hypothetical large-scale radiation accidents in which only suboptimal radiation measurements could be performed due to lack of time, personnel, and appropriate equipment.

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Appendix

Appendix.

List of parameters used in the models of external and internal radioactive contamination of the body and of the clothes.

Parameter	Value	Symbol	Units	Equation	Variable by
Time-integrated outdoor air concentrations of ^{137}Cs	calculated	$DC_{137,out}(t)$	Bq d m^{-3}	(1)	settlement and day t
Total ^{137}Cs ground deposition density in the settlement	DB[a]	Q_{137}	Bq m^{-2}	(1)	constant for settlement
Fraction of ^{137}Cs that deposited during day t	DB	$f_{137}(t)$	unitless	(1)	settlement and day t
Deposition velocity of ^{137}Cs under dry conditions	100	$V_{137,1awn,dry}$	M d^{-1}	(1)	constant
Deposition velocity of ^{137}Cs with precipitation	290	$V_{137,1awn,wet}$	m d^{-1} mm^{-1}	(1)	constant
Amount of precipitation during day t	DB	$R(t)$	mm	(1)	settlement and day t
The time-integrated outdoors air concentration of radionuclide i other than ^{137}Cs during day t	calculated	$DC_{i,out}(t)$	Bq d m^{-3}	(2)	settlement and day t
Ratio of the time-integrated concentrations of radionuclide i and of ^{137}Cs in ground level air during day t	DB	$r_{i,137}(t)$	unitless	(2)	settlement and day t
Ratio of radionuclide i and of ^{137}Cs in the released activities	DB	$r_{0i,137}(t)$	unitless	(3)	constant
Radioactive decay constants of radionuclide i	DB	λ_i	d^{-1}	(3)	constant
Radioactive decay constants of ^{137}Cs	6.3×10^{-5}	λ_{137}	d^{-1}	(3)	constant
Time-integrated indoor air concentrations of radionuclide i	calculated	$DC_{i,in}(t)$	Bq dm^{-3}	(5)	settlement and day t
Filtration factor at time t	1.0	$FC_i(t)$	unitless	(5)	day t
Rate of air exchange in the building	20	λ_{exch}	D^{-1}	(5)	constant
Rate of removal of radionuclide i from indoor air to deposition onto all surfaces inside the building	calculated	h,d	d^{-1}	(5)	category of radionuclide i
Effective deposition velocity of radionuclide i onto horizontal and vertical surfaces in the room	Table 3	$V_{i,in}$	md^{-1}	(7)	category of radionuclide i
Ratio of the area of all surfaces to the volume of the room	2	A/V	m^{-1}	(7)	constant
Fraction of day when the subject stays indoors	0.833	f_{in}	unitless	(8)	constant
Fraction of sleeping time	0.5	f_{sleep}	unitless	(8)	constant
Deposition density of radionuclide i on lawn at day t	calculated	$Q_{i,lawn}(t)$	Bq m^{-2}	(10)	settlement and day t
Cumulative deposition density of radionuclide i on the lawn at time T	calculated	$Q_{i,lawn}(T)$	Bq m^{-2}	(13)	settlement

Parameter	Value	Symbol	Units	Equation	Variable by
Fast component of clearance of body surfaces	Table 4	F_{fast}	unitless	(14)	category of radionuclide *i*
Slow component of clearance of body surfaces	Table 4	F_{slow}	unitless	(14)	category of radionuclide *i*
Retention function of fast component of clearance of body surfaces	calculated	$ret_{i,j,fast}(T,t)$	unitless	(15)	
Retention function of slow component of clearance of body surfaces	calculated	$ret_{i,j,slow}(T,t)$	unitless	(15)	
Reduction washing factor	Table 4	$bath_j$	unitless	(16)	body surface *j*
Number of bathes or the number of times hands of face were washed		N_{bath}	unitless	(16)	
Clearance rate of radionuclide *i* from surface *j*	Table 4	$\lambda_{i,j}$	D^{-1}	(16)	category of radionuclide *i* and body surface *j*
Outdoor or indoor air concentration of secondary aerosols due to resuspension	calculated	$C_{i,second}(t)$	Bq d m^{-3}	(17)	
Resuspension coefficient	2.0×10^{-4}	α	m^{-1}	(17)	constant
Daily probability for the contaminant particles to be deposited on surface level ***H*** above ground	0.04	$P(H)$	d^{-1}	(18)	constant
Parameter normalizing the general gradient of concentration from (outdoors) or floor (indoors) to	2	k_r	unitless	(18)	constant
Standard height	1	H	m	(18)	constant
Scaling factor for the standard height	calculated	SF_{size}	unitless	(18)	age of the subject
Fraction of radionuclide *i* transferred from a contaminated region *j* onto another region *k* of the body	0.2, 0.06, 0.01	$Tr_{i,j,k}$	unitless	(19)	type of body surface: open, semi-open or shielded
Daily pasture grass consumption by the cow during the ramping up period after the first day of the pasture season	calculated	M_{grass}	Kg d^{-1}	(20)	day *t*
Maximum daily pasture grass consumption by cow (fresh weight)	40	M_{max}	Kg d^{-1}	(20)	constant
Mass of pasture grass consumed by cow during the first day of pasture (fresh weight)	4	M_{min}	Kg d^{-1}	(20)	constant
Duration of ramping up period in pasture grass consumption by cow	7	TG	d	(20)	constant

[a] Database

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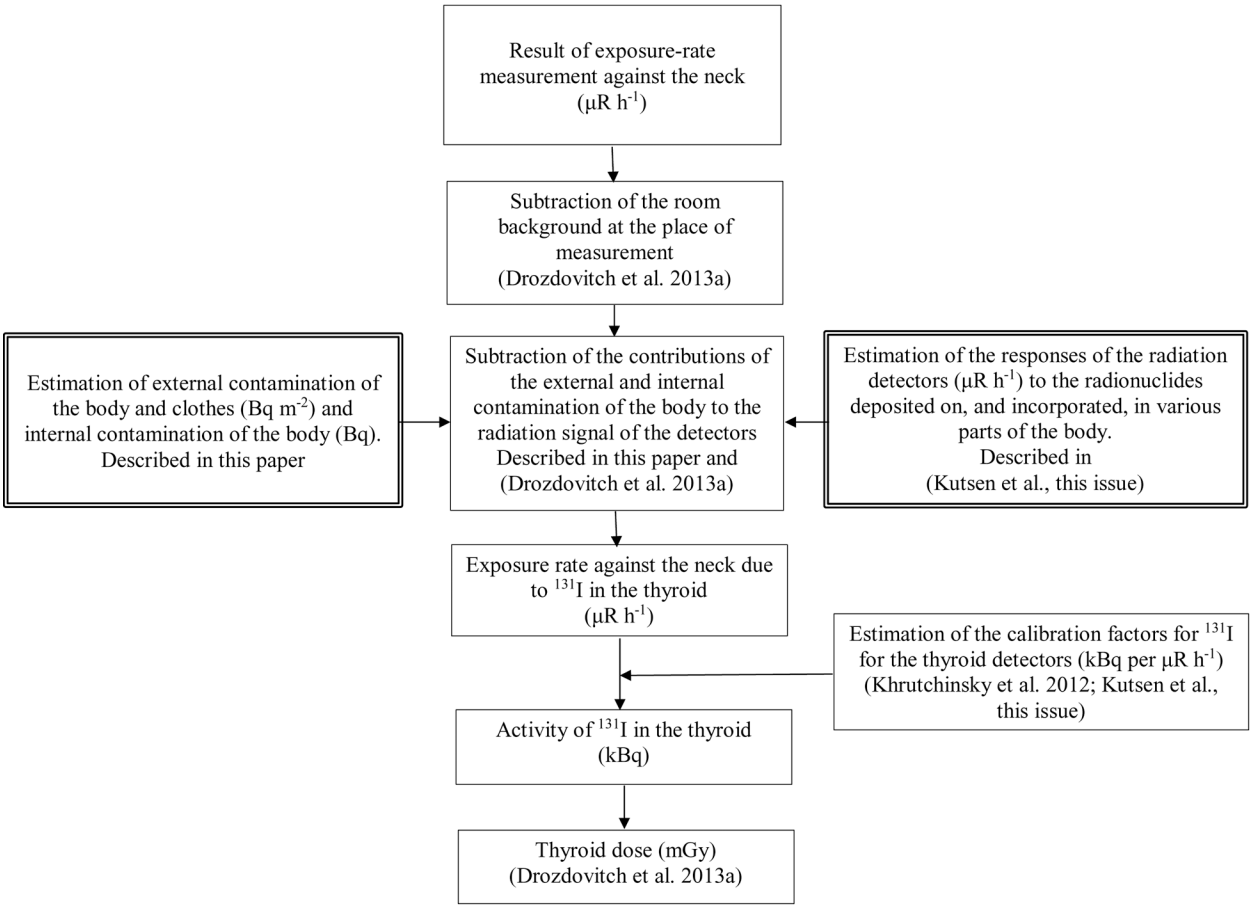

Fig. 1.

Scheme of the thyroid dose assessment based on direct thyroid measurements for the subjects of Belarusian-American screening cohort.

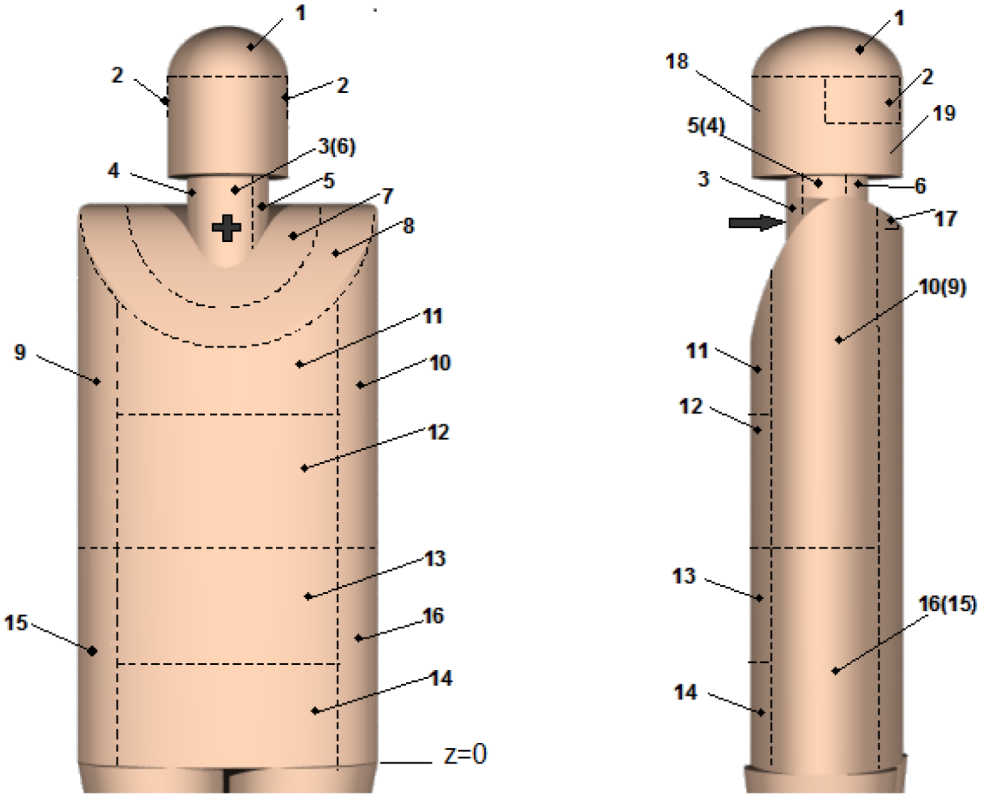

Fig. 2.

Division of phantom into regions: 1- top of the head; 2-sides of the head; 3-front side of neck; 4(5)-right (left) side of neck; 6-back of neck; 7- chest area close to the neck fragment of chest; 8- chest area more distant from the neck; 9(10)-upper right (left) arm; 11-front trunk (upper part); 12-front trunk (middle part 1); 13-front trunk (lower part 2); 14-front trunk (lower part); 15(16)-lower right (left) arm; 17-upper part of shoulder blades; 18-face; 19-occiput. Arrow indicates position of the thyroid detector during the measurement.

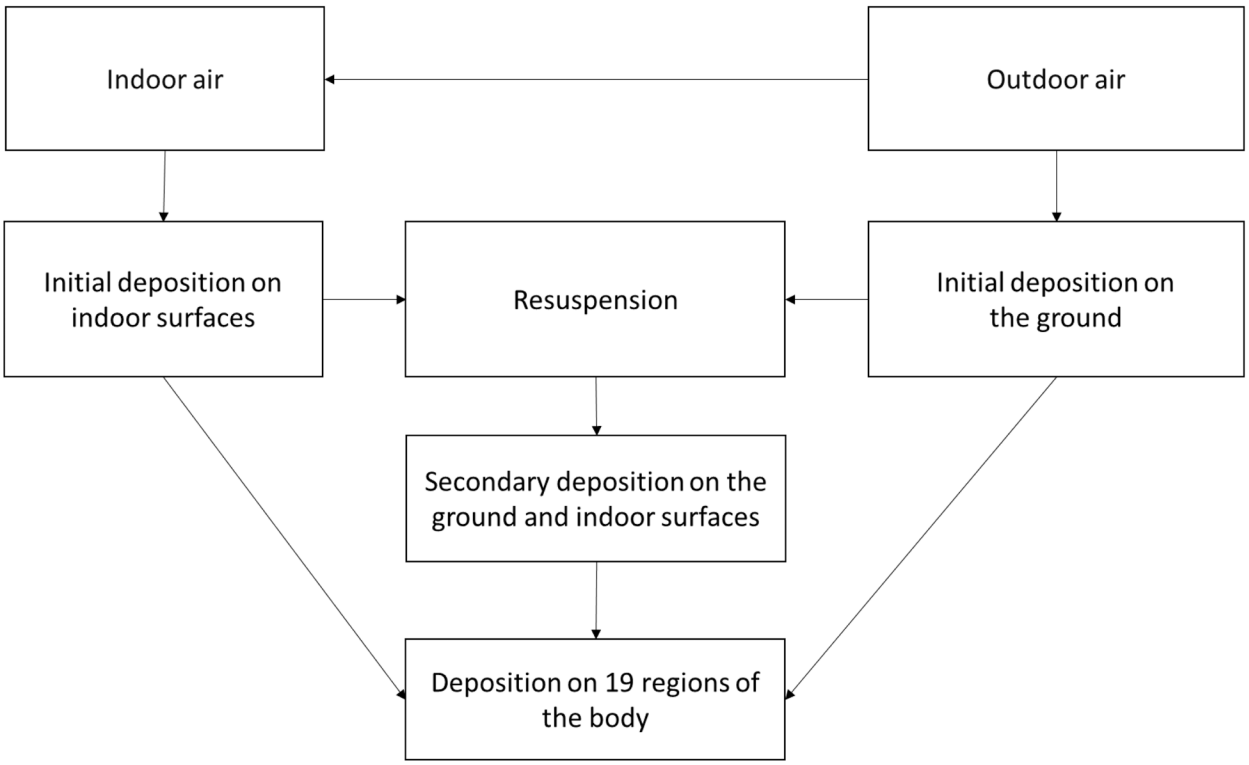

Fig. 3.
Schema of the main processes of external contamination of body surfaces and clothes.

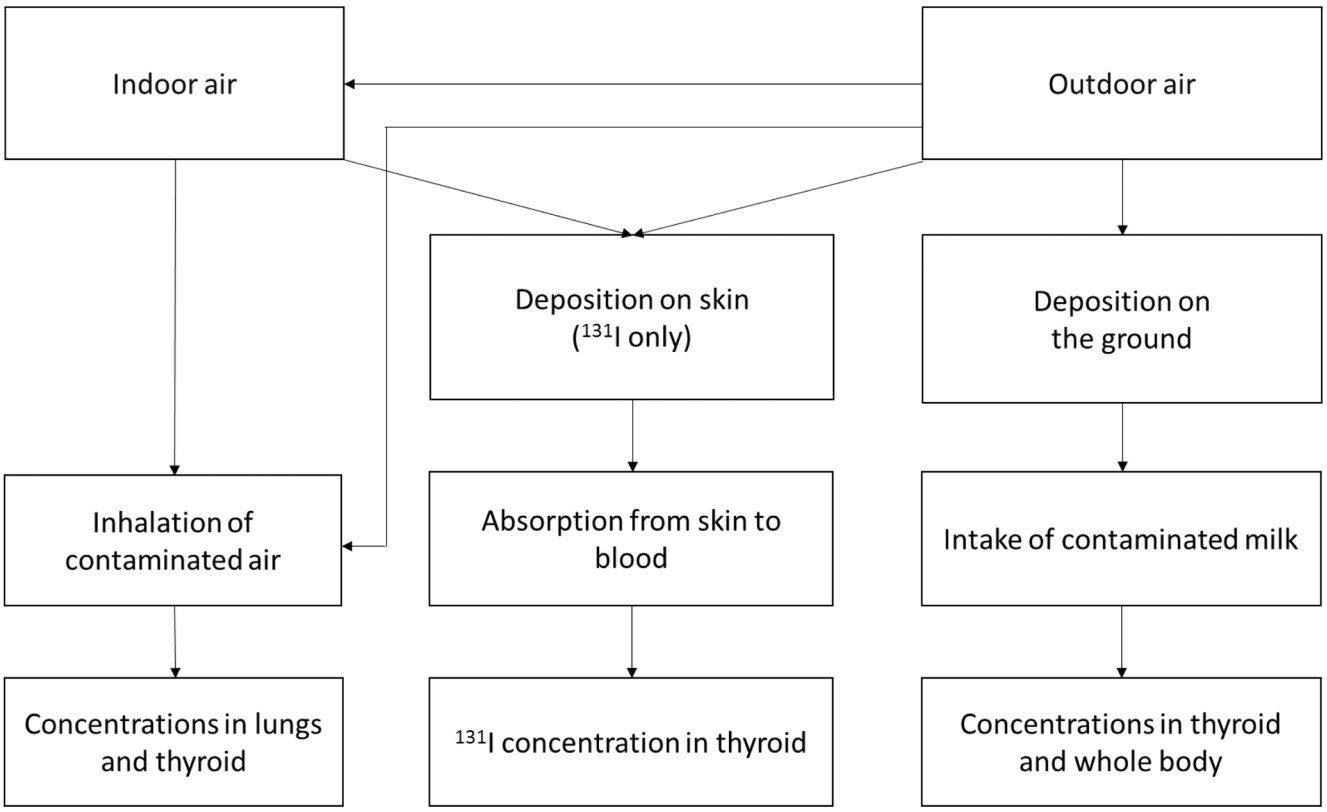

Fig. 4.
Schema of the main processes of internal contamination of the body following intakes of radionuclides.

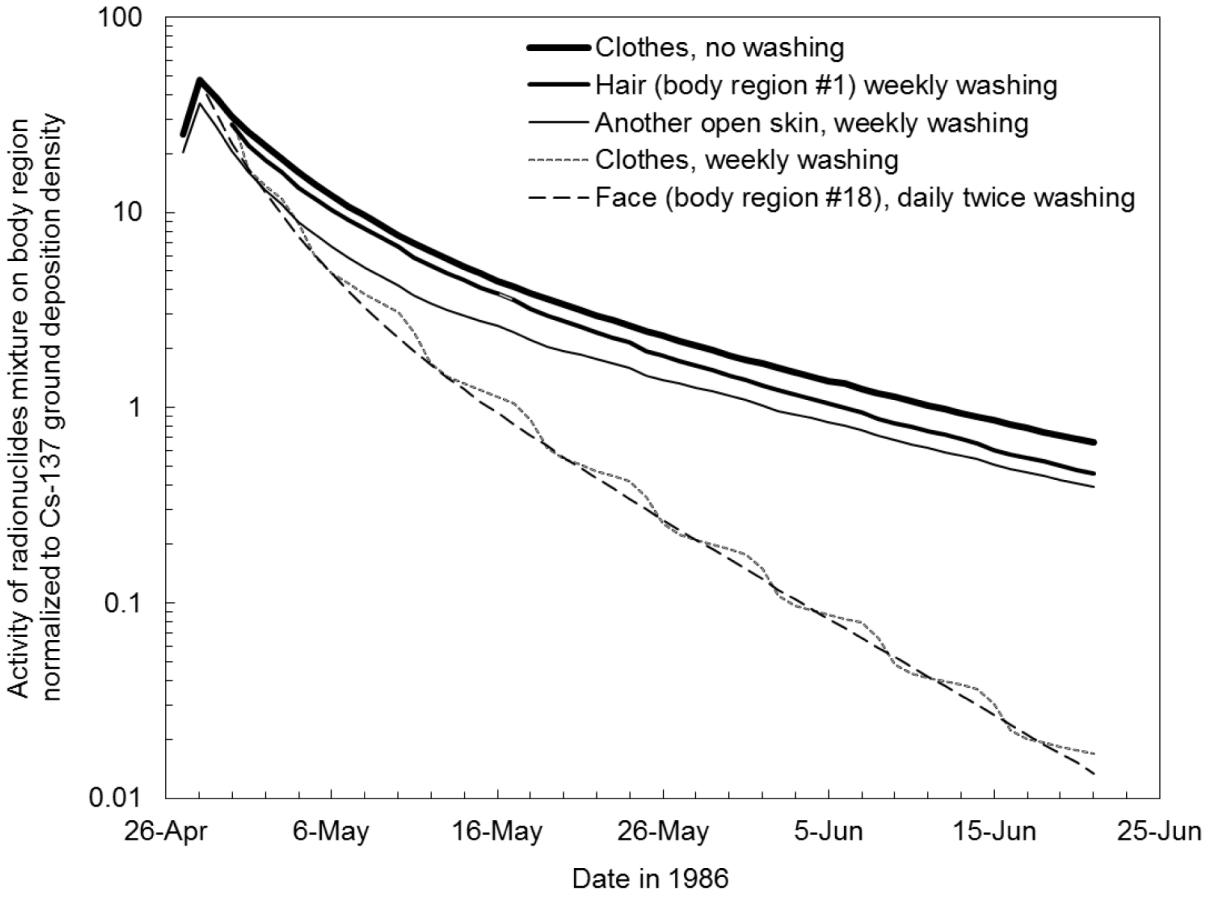

Fig. 5.

Modeled radioactive contamination of the body surface regions (#1 and #18) and clothes related to ^{137}Cs ground deposition density at scenario of dry fallout in town Khoiniki, Gomel Oblast, Belarus during 27–30 April 1986. No resuspension was considered here.

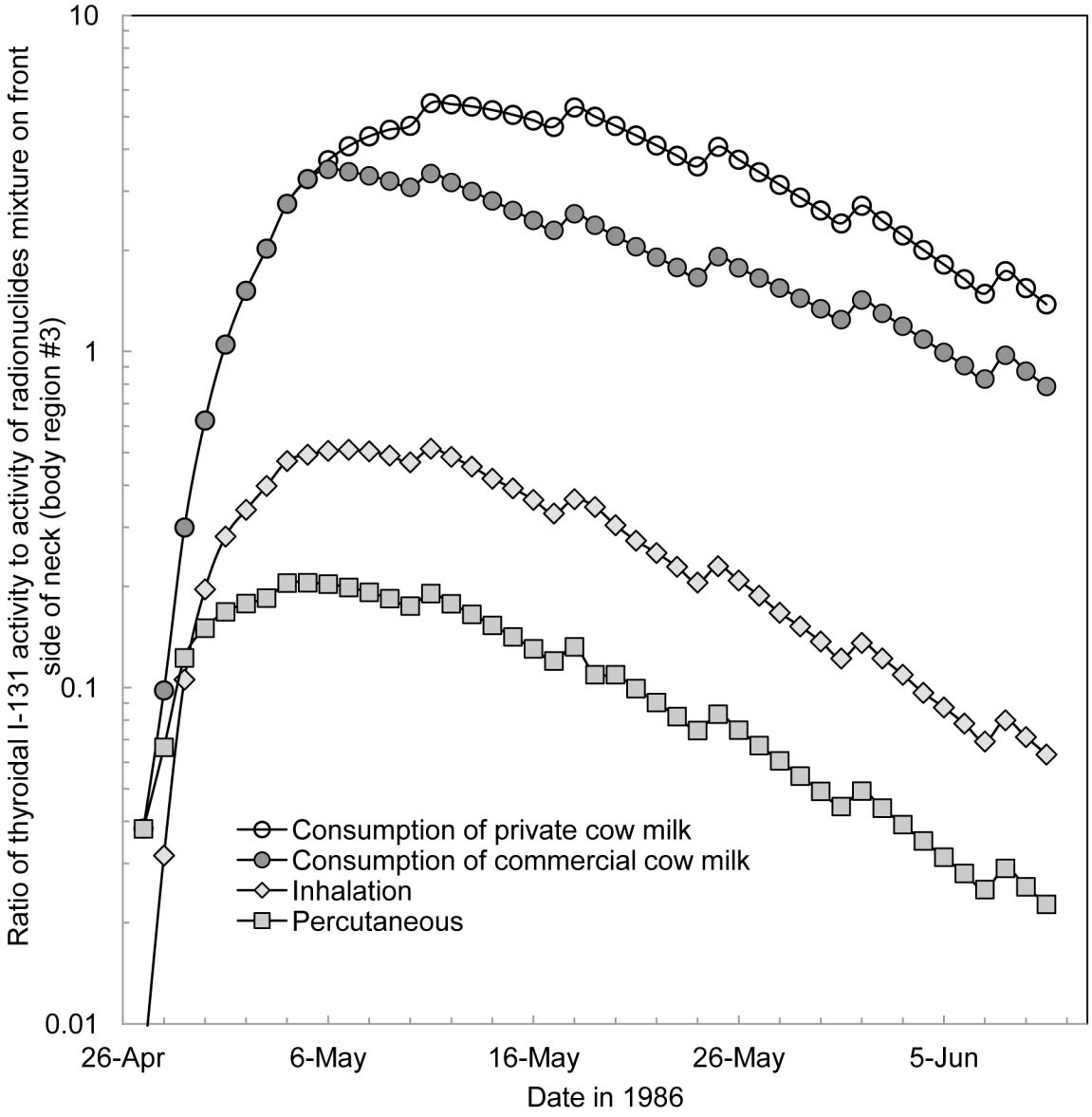

Fig. 6.

Ratio of the thyroidal ^{131}I activity to activity of the radionuclides mixture on skin near thyroid by pathway of ^{131}I intake calculated for a 5-year old resident of the town Khoiniki.

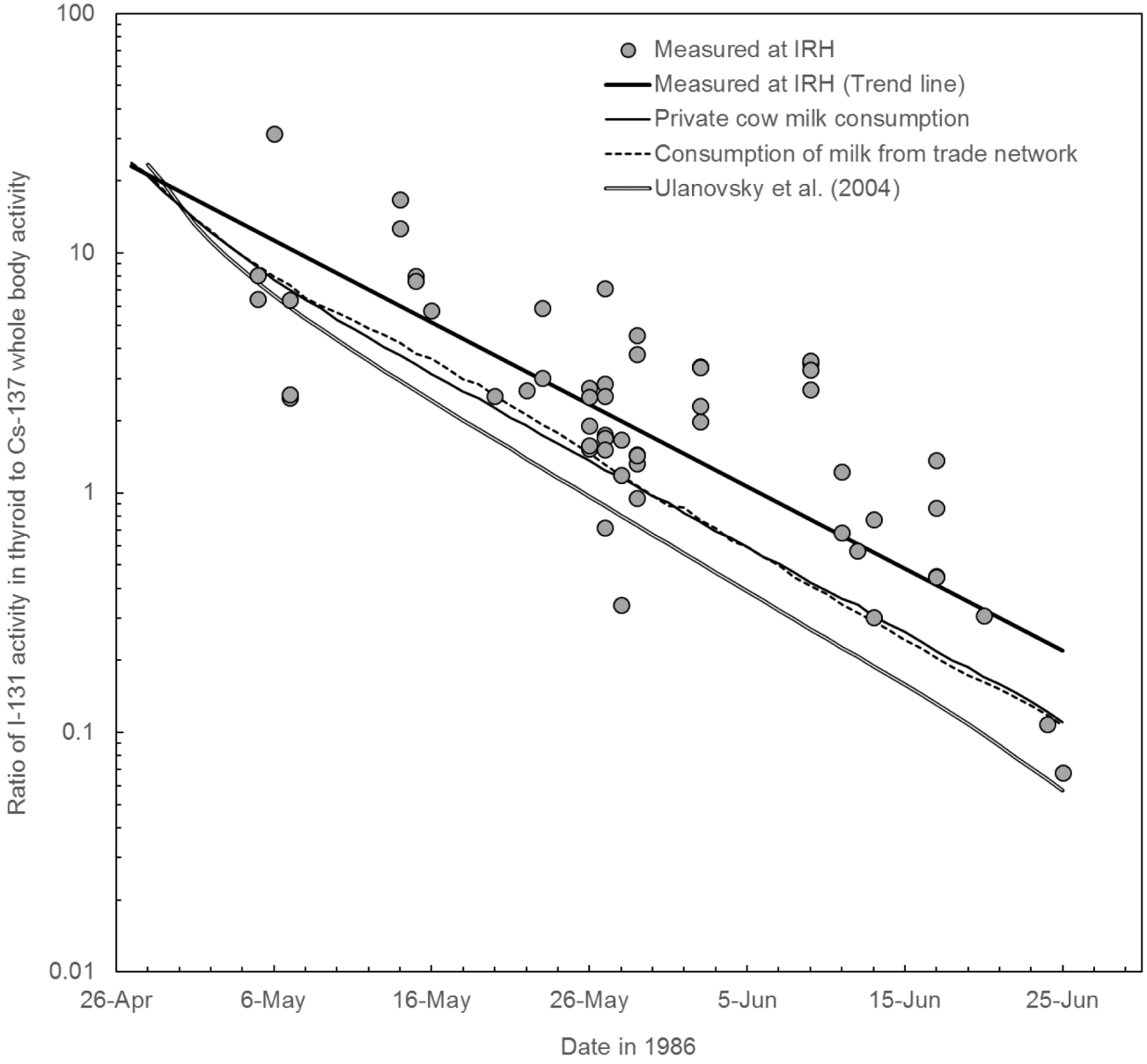

Fig. 7.

Ratios of ^{131}I activity in thyroid and of ^{137}Cs body burden, either calculated using the models described in this paper and by Ulanovsky et al. (2004) or derived from the measurements performed at Institute of Radiation Hygiene (IRH, Saint-Petersburg) among Belarusians who had arrived in Saint-Petersburg from Khoiniki and neighboring raions of Gomel Oblast (Lebedev and Yakovlev 1993).

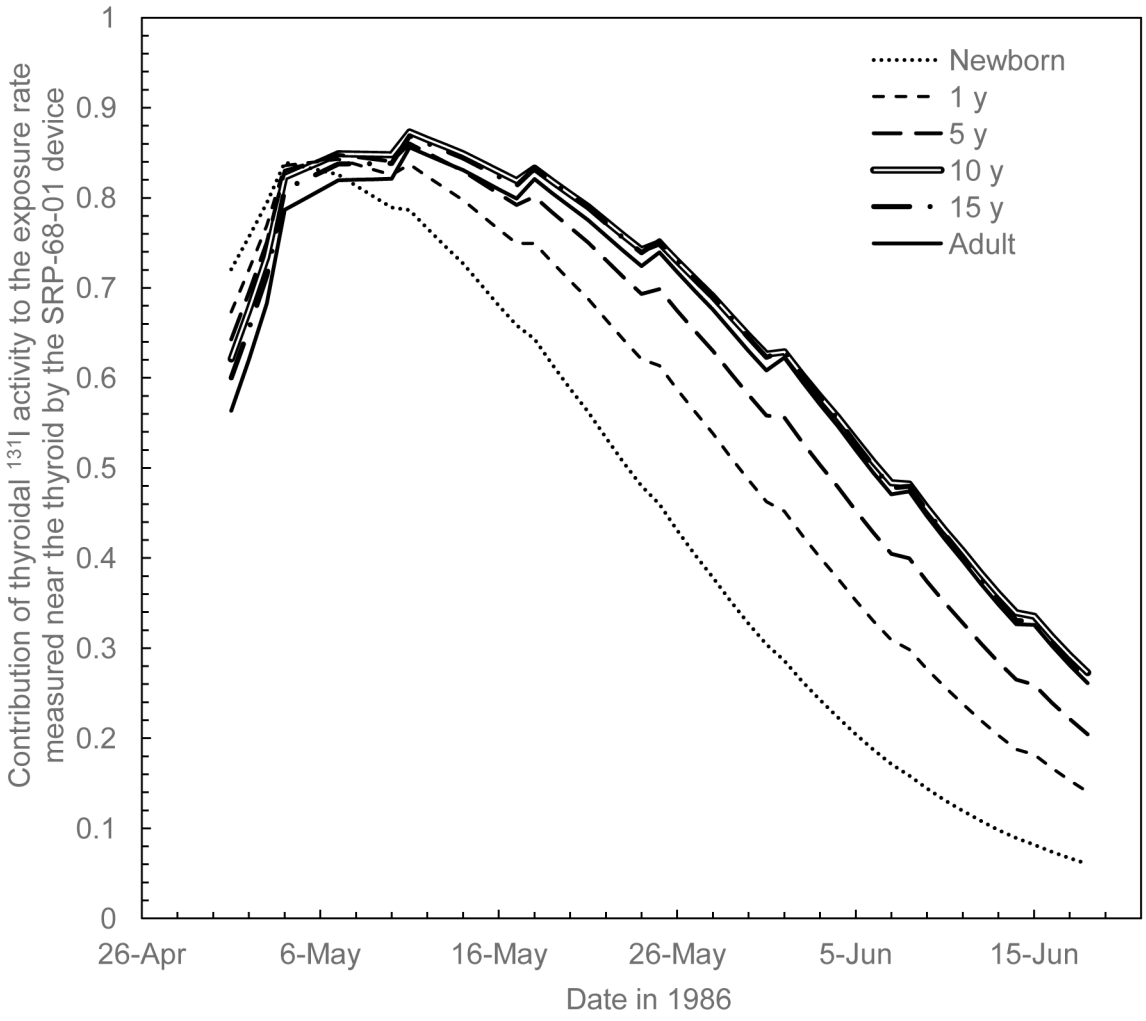

Fig. 8.

Variation with time of contribution of thyroidal ^{131}I activity to the exposure rate measured near the thyroid by the SRP-68–01 device among persons of different ages who resided in Khoiniki.

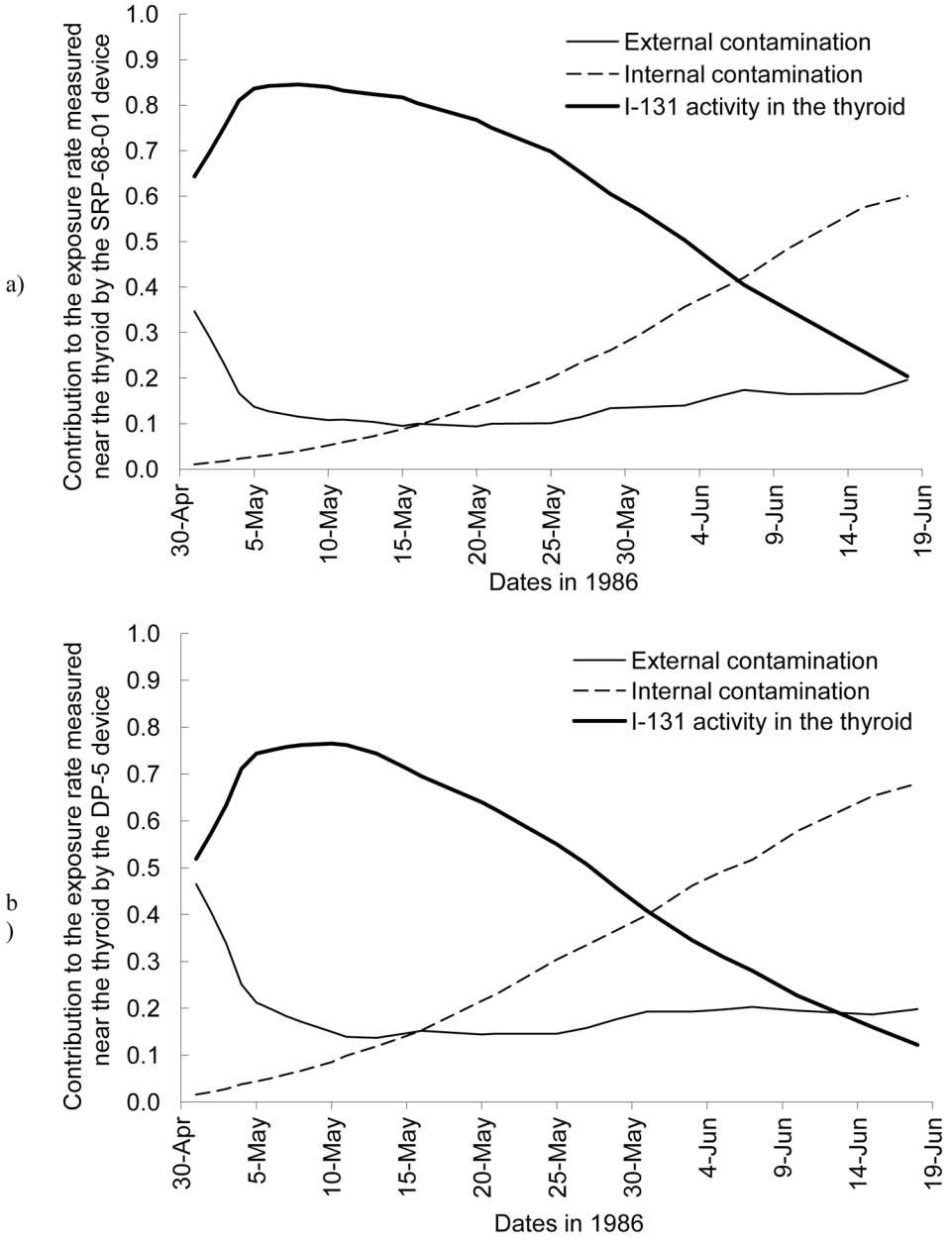

Fig. 9.

Variation with time of the contribution of the external and internal contamination of the subject, and of the ^{131}I activity in the thyroid to the exposure rate measured near the thyroid for a 5-year-old child who resided in Khoiniki raion by (a) the SRP-68–01 device and (b) the DP-5 device.

Table 1.

Characteristics of the gamma-emitting radionuclides that were considered in this study to estimate the external and internal radioactive contamination.

Radio-nuclide	Half-life[a]	Principal decay product (yield)	Mean photon energy[a] (MeV)	Organ(s) considered in internal contamination model	Activity released (PBq)[b]
^{95}Zr	64.03 d	^{95}Nb (0.993)	0.7421	Lungs	84
^{95}Nb	34.99 d		0.7656	Lungs	-
^{99}Mo[c]	65.94 h		0.4992	Lungs	>72
^{103}Ru[c]	39.26 d		0.5014	Lungs	>168
^{106}Ru[c]	373.59 d		0.6022	Lungs	>73
^{132}Te	3.204 d	^{132}I (1.0)	0.1992	Lungs	~ 1,550
^{131}I	8.02 d		0.1639	Thyroid, lungs	~ 1,760
^{132}I	2.30 h		0.7649	Thyroid, lungs	-
^{133}I	20.8 h		0.5845	Thyroid, lungs	910
^{134}Cs	2.065 y		0.6976	Whole body, lungs	~ 47
^{136}Cs	13.16 d		0.7464	Whole body, lungs	36
^{137}Cs[c]	30.17 y		0.6617	Whole body, lungs	~ 85
^{140}Ba	12.75 d	^{140}La (1.0)	0.3253	Lungs	240
^{140}La	1.68d		1.078	Lungs	-
^{141}Ce	32.508 d		0.1454	Lungs	84
^{144}Ce[c]	284.91 d		0.1239	Lungs	~ 50
^{239}Np	2.357 d		0.1875	Lungs	400

[a] ICRP (2008).

[b] UNSCEAR (2011).

[c] Includes a contribution from the short-lived progeny ^{99m}Tc, ^{103}Rh, ^{106}Rh, ^{137m}Ba and ^{144}Pr of ^{99}Mo, ^{103}Ru, ^{106}Ru, ^{137}Cs, and ^{144}Ce, respectively.

Table 2.

Availability of deposition density (measured and calculated) and meteorological data in Belarus for the subjects of Belarusian-American (BelAm) cohort study for the period from 26 April to 30 June 1986.

Parameters	Percentage (number) of settlements in the study area[a]	Percentage (number) of BelAm cohort subjects
Measured		
^{137}Cs ground deposition density in settlement	100.0 (1,077[b])	100.0 (11,732[c])
Daily ^{131}I and ^{137}Cs ground deposition density	0.28 (3)	18.0 (2,115)
Activity of radionuclide *i* in soil	39.4 (424)	58.2 (6,827)
Activity of radionuclide *i* in grass	5.1 (55)	24.8 (2,911)
Daily ^{137}Cs concentration in air	0.09 (1)	17.5 (1,504)
Daily ^{131}I aerosol concentration in air	0.09 (1)	17.5 (1,504)
Detailed data on precipitation: time of beginning, duration and amount	2.5 (27)	33.5 (3,933)
Daily amount of precipitation	8.2 (88)	14.2 (1,666)
Calculated		
Daily ^{131}I ground deposition density	100.0 (1,077)	100.0 (11,732)
Daily ^{137}Cs ground deposition density	100.0 (1,077)	100.0 (11,732)

[a] Gomel and Mogilev oblasts and Minsk-city.

[b] Number of settlements in the study area.

[c] Number of BelAm cohort subjects.

Table 3.

Central estimates of the dry and wet deposition velocity on outdoor or indoor surfaces relative to the ^{137}Cs dry deposition velocity onto lawn surfaces taken to be 100 m d^{-1} (Andersson et al. 2002, 2004; Fogh et al. 1999; Roed and Cannell 1987; Underwood 1990). Radionuclide categories A – D are described in the text

Surface	Category of radionuclides	Deposition velocity relative to the ^{137}Cs dry deposition velocity onto lawn surfaces for	
		Dry deposition	Wet deposition (per 1 mm of rain)
Urban lawn	A	5	14
	B	1	2.9
	C	9.6	1.0
	D	0.10	0.01
Open skin, hair, and clothes (outdoors)	A	6.0	1.4[a]
	B	1.2	0.3[a]
	C	14	-[b]
	D	0.07	-[b]
Indoor surfaces	A	0.4	-
	B	0.08	-
	C	0.6 (0.15)[c]	-
	D	0.008	-
Open skin and hair (indoors)	A	10	-
	B	0.86	-
	C	8.6	-
	D	0.09	-
Clothes (indoors)	A	2.3	-
	B	1.5	-
	C	8.6	-
	D	0.09	

[a]Value for hair and clothes only; wet deposition of any radionuclide on skin was considered to be negligible.

[b]Wet deposition of gaseous radioiodine (categories C and D) onto any surface of body and clothes was considered to be negligible.

[c]Value for isotopes of ruthenium.

Table 4.

Parameter values of the retention functions used in the body surface contamination model that were estimated on the basis of data provided in Andersson et al. (2002) and Fogh et al. (1999).

Parameter	Category of radionuclides	Surface	Component of clearance	
			Fast	Slow
Fraction of cleared component	A	Any	0.85	0.15
	B		0.15	0.85
	C		-	1
	D		-	1
Clearance rate (d^{-1})	A, B	Any, besides face and hand	4.0	0.023
		Face and hand	6.3	0.093
	C,D	Any open and semi-open section of skin besides face and hand	-	0.273
		Face and hand	-	0.343
Reduction washing factor	Any	Skin and hair	0.1	0.63
		Clothes	0.63	0.63