ORIGINAL ARTICLE

Thyroid doses in Ukraine due to ^{131}I intake after the Chornobyl accident. Report I: revision of direct thyroid measurements

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Abstract

The increased risk of thyroid cancer among individuals exposed during childhood and adolescence to Iodine-131 (^{131}I) is the main statistically significant long-term effect of the Chornobyl accident. Several radiation epidemiological studies have been carried out or are currently in progress in Ukraine, to assess the risk of radiation-related health effects in exposed populations. About 150,000 measurements of ^{131}I thyroid activity, so-called 'direct thyroid measurements', performed in May–June 1986 in the Ukrainian population served as the main sources of data used to estimate thyroid doses to the individuals of these studies. However, limitations in the direct thyroid measurements have been recently recognized including improper measurement geometry and unknown true values of calibration coefficients for unchecked thyroid detectors. In the present study, a comparative analysis of ^{131}I thyroid activity measured by calibrated and unchecked devices in residents of the same neighboring settlements was conducted to evaluate the correct measurement geometry and calibration coefficients for measuring devices. As a result, revised values of ^{131}I thyroid activity were obtained. On average, in Vinnytsia, Kyiv, Lviv and Chernihiv Oblasts and in the city of Kyiv, the revised values of the ^{131}I thyroid activities were found to be 10–25% higher than previously reported, while in Zhytomyr Oblast, the values of the revised activities were found to be lower by about 50%. New sources of shared and unshared errors associated with estimates of ^{131}I thyroid activity were identified. The revised estimates of thyroid activity are recommended to be used to develop an updated Thyroid Dosimetry system (TD20) for the entire population of Ukraine as well as to revise the thyroid doses for the individuals included in post-Chornobyl radiation epidemiological studies: the Ukrainian-American cohort of individuals exposed during childhood and adolescence, the Ukrainian in utero cohort and the Chornobyl Tissue Bank.

Keywords Chornobyl · Chernobyl · Radiation exposure · Thyroid dose · Iodine-131 measurement · Classical error · Berkson error

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Introduction

The increase in thyroid cancer incidence among individuals exposed as children and adolescents to Iodine-131 (^{131}I) is the main long-term effect of the Chornobyl (Chernobyl) accident (UNSCEAR 2011). Unsurprisingly, this radiation-related health effect generated great interest and led to a series of radiation epidemiological studies, to quantify the risk of radiation-related thyroid cancer among the residents of the most contaminated regions of the three most affected countries: Belarus, Ukraine and the Russian Federation (e.g. Astakhova et al. 1998; Cardis et al. 2005; Kopecky et al. 2006; Likhtarov et al. 2006; Tronko et al. 2006, 2017). One of the most advanced post-Chornobyl studies,

the Ukrainian-American cohort study of thyroid cancer and other thyroid diseases includes 13,204 individuals who were exposed between ages 0 and 18 years to ^{131}I from the Chornobyl fallout and who are followed up for thyroid cancer and other thyroid diseases through a standardized screening protocol (Stezhko et al. 2004). Individual doses due to ^{131}I intakes were reconstructed two times for all cohort members using dosimetry systems that were revised to take improvements into account (Likhtarev et al. 2006; Likhtarov et al. 2014).

The 'Thyroid Dosimetry 2010 system', TD10, is currently being used to assess the radiation-related risk of thyroid cancer and other thyroid diseases in the Ukrainian-American cohort (Brenner et al. 2011; Hatch et al. 2010; Little et al. 2014; Tronko et al. 2017). However, the limitations in the dosimetry system regarding direct thyroid measurements have been recently recognized. These limitations include an improper geometry of measurement and unknown true values of calibration coefficients for the measuring devices used at that time. A wide-scale radiation monitoring of ^{131}I activity in the thyroid, called 'the thyroid dosimetry monitoring', in the affected population was organized within a relatively short period of time immediately after the accident. This has led an increase in errors for some field measurements as compared to purely laboratory-based measurements. These errors were caused by the involvement of personnel in the thyroid dosimetry monitoring who had no or only minimal experience in radiation measurements, by the use of various types of devices that were not designed to measure radioactivity in the thyroid gland, and by application of simplified methods for express measurements (Likhtarov et al. 2015). It was already suspected that errors associated with the thyroid measurements may have resulted in unexpected differences in the radiation-related risk of thyroid cancer between two oblasts of the Ukrainian-American cohort study, Zhytomir and Chernihiv (Brenner et al. 2011).

The main sources of data used for estimation of thyroid doses in Ukraine were (1) radiation measurements (called 'direct thyroid measurements') that were conducted against the thyroids of all cohort members within the first two months after the Chornobyl accident (Likhtarev et al. 1993, 1995, 2015); (2) estimates of ^{131}I deposition on the ground at each location where the cohort members resided (Talerko 2005); and (3) estimates of age- and gender-specific thyroid masses for children and adolescents (Likhtarov et al. 2013a).

Recently, recognized limitations in the dosimetry system resulted in the revision of thyroid dosimetry in Ukraine including three parts: (1) a detailed analysis of 146,425 measurements resulting in a revision of the measured ^{131}I thyroid activity and in the identification of sources of shared and unshared errors associated with the measurements; (2) the creation of an improved 'Thyroid Dosimetry 2020 system' (named TD20 here and below) for the entire population

of Ukraine which is described by (Likhtarev et al. 2005); and (3) the recalculation of the thyroid doses for the members of the Ukrainian-American cohort (Likhtarov et al. 2014) that incorporated the revised ^{131}I thyroid activities and new sources of errors associated with direct thyroid measurements, for the Ukrainian in utero cohort (Likhtarov et al. 2011), and for the subjects included in the Chornobyl Tissue Bank (Likhtarov et al. 2013b). The present paper describes the first part of the revision of the thyroid dosimetry in Ukraine, while the other parts will be considered in separate papers.

Materials and methods

Thyroid dosimetry monitoring in Ukraine

The accident at the Chornobyl nuclear power plant led to the environmental release of 1.8 EBq of ^{131}I (UNSCEAR 2011) and, consequently, to radiation exposure to the local population. Wide-scale measurements of ^{131}I in the thyroid were organized and conducted in May and June 1986 under the supervision of the Ministry of Health of the Ukrainian Soviet Social Republic. About 150,000 measurements were performed in Ukraine during the thyroid dosimetry monitoring. One-third of these measurements were performed using single-channel gamma-spectrometers, namely the GTRM-01ts, NK, DSU-68, DSU-2 and UR devices, within the energy window of ^{131}I (364 keV, yield: 81.7%), including detectors equipped with lead collimators that significantly reduced the impact of radioactive background on the results of the measurements. The rest of the measurements were done with the SRP-68-01 device, a NaI(Tl) scintillation survey meter, which was commonly used in the former Soviet Union for geological exploration. Since this device was not equipped with a factory-made collimator, homemade collimators with a typical thickness of 4 mm and a length of 5–15 cm were used whenever possible. Exceptionally, measurements were also performed with a gamma-spectrometer, namely the PRL, and the DP-5 device including a Geiger-Mueller (GM) counter, which was designed for use by military and civil defense organizations. Table 1 provides characteristics of the thyroid monitoring by types and models of the devices used, the number of measurements, and duration of the monitoring.

The device's detector was placed against the neck, which was cleaned with alcohol, of the measured person. For some measurements done shortly after the accident, an additional measurement of the forearm was performed because it was assumed that the contamination of the forearm surface was the same as that of the neck. The results of these measurements, which were given in terms of the number of counted pulses or their intensity for the gamma-spectrometer or in

Table 1 Number of thyroid measurements[a] by Oblast and Kyiv city that were done in Ukraine and among the Ukrainian-American cohort members using various types of devices[b]

Oblast or raion	Ukraine						Ukrainian–American cohort					
	SRP-68-01	GTRM-01ts	NK	DSU-2	DSU-68	UR	SRP-68-01	GTRM-01ts	NK	DSU-2	DSU-68	UR
Dates of measurements in 1986	30 April–25 June[c]	17 May–30 June[d]	14 May–11 June	18–29 May	25–31 May	17 May–6 June	30 April–25 June	17 May–30 June	14 May–4 June	21–27 May	25–31 May	17–28 May
Zhytomyr Oblast												
Narodychy raion	15,745	4257	1	–	–	2	1203	134	1	–	–	–
Ovruch raion	22,116	48	10	1	1	2	2330	1	1	–	–	–
Other raions	9985	630	146	47	–	18	1	–	–	1	–	–
Entire Oblast	47,846	4935	157	48	1	22	3534	135	2	1	–	–
Kyiv Oblast												
Ivankiv raion	3,552	6	36	–	–	409	650	1	11	–	–	74
Poliske raion	2,268	18	1,310	377	–	430	228	–	121	27	–	30
Chornobyl raion	3,694	216	882	14	1	359	528	25	119	–	–	51
Pripyat-town	2,314	582	1,233	77	7	444	328	67	237	16	1	60
Other raions	13,417	34	2804	617	2	2389	2	–	1	–	–	–
Entire Oblast	25,245	856	6265	1085	10	4031	1736	93	489	43	1	215
Chernihiv Oblast												
Kozelets raion	8919	3202	1628	–	4	1	1381	366	235	–	–	–
Ripky raion	6828	93	2225	–	15	2628	894	18	298	–	2	441
Chernihiv raion	908	4941	6921	–	939	280	149	683	1199	–	152	27
Chernihiv city	72	3471	1901	–	3053	25	7	479	149	–	448	1
Other raions	25	5	5	1	–	15	–	–	–	–	–	–
Entire Oblast	16,752	11,712	12,680	1	4011	2949	2431	1546	1881	–	602	469
Vinnytsia Oblast	1626	–	–	–	418	–	–	–	–	–	–	–
Kyiv city	2106	174	110	209	8	2,316	1	4	–	–	–	–
Other Oblasts[e]	142	156	109	2	4	427	1	3	3	–	–	2
Total	93,717	17,833	19,321	1345	4452	9745	7703	1781	2375	44	603	686

[a] Numbers are given according to place of residence although persons could be measured in other areas of Ukraine

[b] Five measurements done by the DP-5 dose rate meter on 8–19 May 1986 and 7 measurements were done by the gamma-spectrometer PRL on 8–12 May 1986 among the Ukrainian-American cohort members are not shown in the table

[c] About 90% of measurements were done before 12 June 1986

[d] More than 99.95% of measurement were done before 11 June 1986

[e] Including 248 persons who were measured outside of Ukraine

terms of exposure rate for the SRP-68-01, performed by the same team in the same settlement on the same day using the same device were recorded in a so-called 'measuring list'. Every hour or every day, the background radiation in the measurement room was recorded. A cylindrical bottle with 10 ml of a reference solution of ^{131}I was used as primitive phantom (so-called 'bottle phantom') for routine checking of the sensitivity (hereinafter referred to as "calibration") of the measuring devices, if it was possible, either hourly or daily.

A typical measuring list contained up to 700 individual measurements, although in some cases, the number of measurements performed by the team during the day was more than 1000 (see Section on "Gamma-spectrometers" below). The following information was recorded in the measuring list: identification data of the measured individual (full name or last name and initials, date or year of birth), information on the team conducting the measurements, on type of measuring device, the results of the device's calibration, and the value of radiation background in the room.

Figure 1 shows the spatial distribution of the number of direct thyroid measurements done after the Chornobyl accident among people residing in the most contaminated study areas of Zhytomir, Kyiv, and Chernihiv Oblasts. It should be noted that numbers of measured individuals are given in terms of their places of residence at the time of the accident, and not in terms of the locations where the measurements

were done. Residents of Zhytomir, Kyiv, and Chernihiv Oblasts were measured in these oblasts in the places of residence as well as in other nine oblasts in Ukraine where they were evacuated or where they moved voluntarily shortly after the accident. In addition, local residents were measured in Vinnytsia and Lviv Oblasts.

Table 1 shows the number of measurements by Oblast, raion and city using different devices done in Ukraine and, partially, among the Ukrainian-American cohort members. Most of the measurements of the Ukrainian population and of the cohort members were performed by means of the SRP-68-01 device, 93,717 from 146,425 (64.0% of the total) and 7,703 from 13,204 (58.3% of the total), respectively.

Because measurements were performed during the same time period in 12 oblasts of Ukraine, corresponding to about half of the country, it was impossible to supply all measurement teams with the reference radiation sources for calibration of the devices. As a high priority, ^{131}I solutions for calibration were provided to the teams working with gamma-spectrometers. Table 2 shows that most of the gamma-spectrometers, i.e., 26 from 28 (92.9% of the total), were calibrated during the measurement campaigns at least once. In contrast, only 23 from 64 of the radiometer devices (35.9% of the total) were calibrated. Table 3 gives the number of the cohort members measured using calibrated gamma-spectrometers and the radiometers. In total, 51.8% (2844
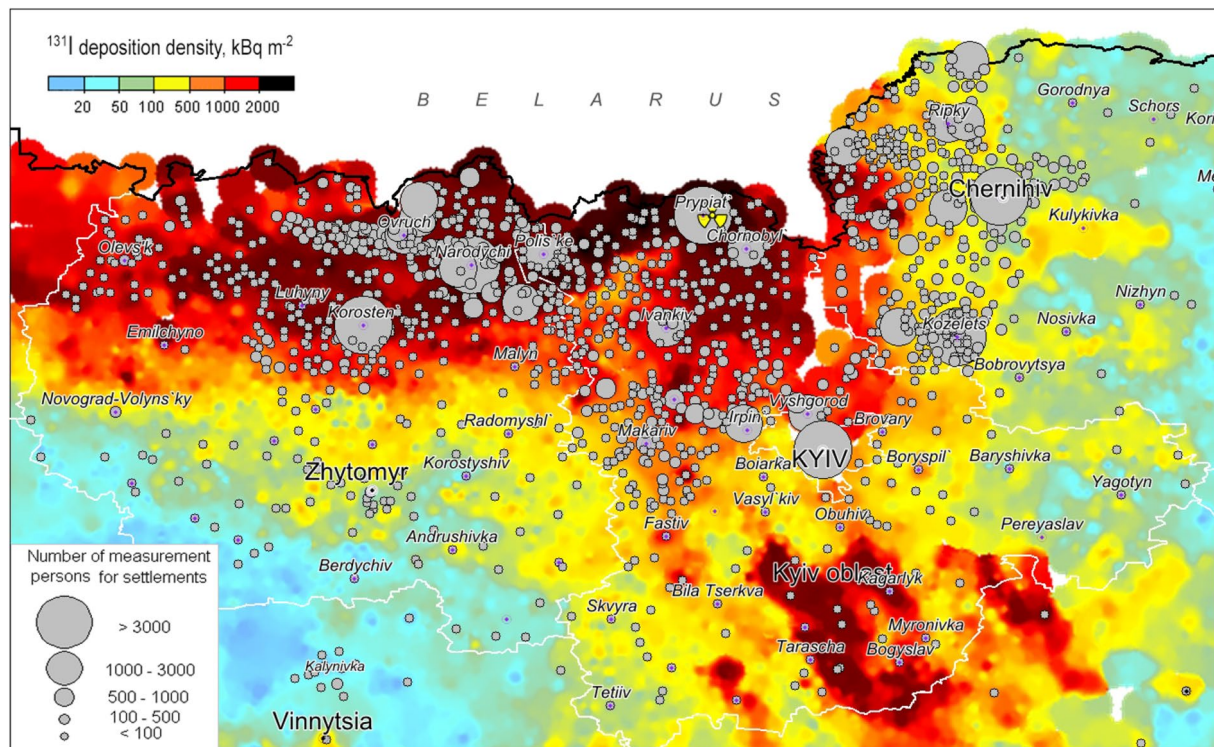
Fig. 1 Spatial distribution of the number of direct thyroid measurements done in Zhytomir, Kyiv, Chernihiv, and Vinnytsia Oblasts, after the Chornobyl accident

Table 2 Distribution of calibrated and unchecked gamma-spectrometers and radiometers by oblast where the measurements were performed

Oblast of measurement	Number of devices used for measurements		Number of measurements	Percent of calibrated measurements
	Total	Calibrated[a]		
Zhytomyr	2/11[b]	2/3	4882/42,584	100.0/2.7
Chernihiv	7/4	6/–	31,463/15,247	44.8/–
Odesa	2/13	2/12	1778/16,329	100.0/97.1
Crimea	7/8	7/4	5480/6556	100.0/53.2
Kyiv, including Kyiv city	6/5	4/–	5334/2006	84.9/–
Lviv	3/1	3/1	3253/1059	100.0/100.0
Khmelnytsk	1/6	1/–	45/3612	100.0/–
Vinnytsia	1/2	1/2	418/1626	100.0/100.0
Sumy	1/6	1/–	50/1757	100.0/–
Rivne	–/1	–/1	–/1655	–/100.0
Donetsk	–/5	–/–	–/1042	–/–
Zaporizhia	–/2	–/–	–/40	–/–
Total[c]	28/64	26/23	52,703/93,722	65.5/26.5

[a]Number of devices that were calibrated at least once

[b]Spectrometer/radiometer

[c]One spectrometer was used in three different oblasts: Kyiv, Chernihiv and Lviv Oblast. It was not calibrated in Chernihiv Oblast

Table 3 Number of measurements performed among the Ukrainian-American cohort members using the gamma-spectrometers and the SRP-68-01 devices

Oblast[a]	Number of devices used to measure the cohort members		Number of measured cohort members	
	Total	Calibrated[b]	Total	Calibrated
Kyiv, including Kyiv city	19/33[c]	15/14	852/1742	703/1345
Zhytomyr	6/13	6/1	138/3534	138/48
Chernihiv	10/7	9/–	4498/2431	1991/–
Total[d]	22/43	20/15	5496/7708	2844/1393

[a]Oblast of person residence on the 26th of April 1986, not oblast where the measurements were performed

[b]Device was calibrated at least once

[c]Spectrometer/radiometer

[d]Including nine individuals who resided in other oblasts at the time of the accident

from 5496) and 18.1% (1393 from 7708) cohort members were measured using calibrated gamma-spectrometers and SRP-68-01 devices, respectively.

Atmospheric modelling

The ^{131}I ground deposition densities were used to normalize the activities of ^{131}I measured in the thyroid (see Section on "Comparative analysis to evaluate correct geometry of measurement and calibration coefficient for unchecked devices"). The ^{131}I deposition density in Ukraine after the Chornobyl accident (Fig. 1) was calculated using the mesoscale atmospheric transport model LEDI. In that model, the ^{131}I atmospheric transport over the territory of Ukraine was simulated for the first 12 days after the accident, from 26 April to 7 May 1986 (period of deposition), with the ^{131}I source term developed by Talerko (2005) that accounted for variability with time of the release rate and the source effective height during the period of release. The meteorological data used in the model were generated by the Weather Research Forecast model (WRF) (Powers et al. 2017), version 3.9.1, adapted for Ukraine. The area covered a central part of Europe including Ukraine and Belarus (247×211 grid cells) with a horizontal resolution of 10×10 km and 28 vertical layers. The pressure of the top layer was set to 100 hPa and the minimum vertical grid step near the Earth's surface was about 50 m. Data from the European Center for Medium-Range Weather Forecasts were used as boundary conditions for the WRF simulations. Using meteorological fields with a high resolution in time and space from the WRF model allowed to improve the simulation of radionuclide dispersion and deposition of radionuclides in comparison to that of Talerko (2005). The updated daily values of the ^{131}I airborne concentration and ^{131}I ground deposition density for the period from 26 April to 7 May 1986 were calculated for 30,352 settlements in entire Ukraine, including 1263 settlements in Kyiv Oblast, 1717 in Zhytomyr Oblast and 1570 in Chernihiv Oblast (Talerko et al. 2020).

SRP-68-01 survey meter

Sixty-four percent of the direct thyroid measurements were made in Ukraine using the SRP-68-01 device (Fig. 2). This device included a NaI(TI) crystal ($\varnothing$30 mm $\times$ 25 mm length) that responded to the photons emitted by ^{131}I in the thyroid in terms of exposure rate expressed in μR h^{-1}. For the conversion of the recorded exposure rate to the ^{131}I activity in the thyroid at the time of the measurement, the so-called conversion coefficient, C_a, expressed in Bq per μR h^{-1} was used. The measured exposure rate included not only the signal due to ^{131}I in the thyroid, but also any signal due to (1) background radiation in the room where the measurements were made, (2) external contamination of the body and clothes of the investigated individual and (3) internal contamination of the body with ^{134}Cs, ^{136}Cs and ^{137}Cs. The contribution to the detector reading from other radionuclides, e.g., ^{95}Zr, ^{95}Nb, ^{103}Ru, ^{106}Ru, ^{140}Ba, ^{140}La, was assumed to be negligible and, therefore, was not considered in the study.

To minimize the background signals, the SRP-68-01 devices used in Ukraine were equipped with a hand-made cylindrical lead collimator, which surrounded the radiation detector while having a clear view of the thyroid. However, using such collimators did not completely eliminate the response of the SRP-68-01 device to sources other than ^{131}I in the thyroid. Instead, depending on the spectral characteristics of the background, the collimator weakened the background signal by a factor of up to 3.

Thyroid ^{131}I activity at the time of the direct thyroid measurement was calculated as:

$$Q = 10^{-3} \bullet C_b \bullet G \bullet \left(I_{\text{th}}^{\text{meas}} - f_{\text{sh}} \bullet I_{\text{bg}}^{\text{meas}} \right) = C_a \bullet I_{\text{net}}^{\text{meas}}, \qquad (1)$$

and

$$C_a = C_b \bullet G, \qquad (2)$$

where Q is the activity of ^{131}I measured in the thyroid (kBq);10^{-3} is a unit conversion coefficient (kBq Bq^{-1}); C_a is the conversion coefficient (Bq per μR h^{-1}); C_b is the

Fig. 2 The SRP-68-01 survey meter

calibration coefficient for the device obtained from the measurements of the reference radiation source with known ^{131}I activity (bottle phantom) (Bq per μR h^{-1}); G is the correction factor for C_b that accounts for the difference of the measurements geometry between the reference source (the bottle) and a person of different ages (unitless); $I_{\text{th}}^{\text{meas}}$, $I_{\text{bg}}^{\text{meas}}$ and $I_{\text{net}}^{\text{meas}}$ represent the readings of the device during the measurements of the thyroid gland, background, and net device indication, respectively (μR h^{-1}); f_{sh} is the fraction of gamma background radiation attenuated by the body of the measured individual that depends, in particular, on the anthropometric parameters of the body (unitless). The f_{sh}-values can vary between 1 (no attenuation) and 0 (full attenuation).

The fractions of gamma background attenuation by the body, f_{sh}, for an adult subject, i.e., for the subject with maximal adsorption of gamma radiation, were experimentally obtained by the authors of this work for the SRP-68-01, NK and GTRM-01ts devices. The resulting values were found to be 0.94–0.95 for the SRP-68-01 device and 0.88–0.96 for gamma-spectrometers for 'standard' geometry of measurements, i.e. for the case when the detector was located against the neck. These experimental values are in good agreement with f_{sh} values in the range of 0.87–1.0 that were estimated by Bratilova et al. (2003), Pitkevich et al. (1996), and Zvonova et al. (1997).

It should be noted that the net reading of the devices (Eq. 1) might include the background signal due to external contamination of the body and clothes of the person and due to internal contamination of the body with ^{134}Cs, ^{136}Cs and ^{137}Cs. However, it was considered here that the background signal due to the external contamination of the body and clothes was negligible, because (1) the neck of a measured individual was cleaned with a cotton wool wet in alcohol, and the contribution to the SRP-68-01 response from other contaminated parts of the body was much lower even for a device without a collimator (Kutsen et al. 2019); and (2) the vast majority of direct thyroid measurements in Ukraine were done three weeks or more after the accident and, therefore, individuals wore clean clothes because they are expected to wash their clothes at least every week. This was especially likely for individuals evacuated shortly after the accident to non-contaminated regions of Ukraine where they were measured (Table 2). The contribution to the response of the measuring device of ^{131}I activity distributed in the human body outside the thyroid gland, even for non-collimated NaI(TI) detectors, was estimated to be about 1−3% (Gómez-Ros et al. 2019). Therefore, for devices equipped with collimators, the contribution of extrathyroidal iodine activity was insignificant from a practical point of view.

Contribution of the internal contamination of the body with ^{134}Cs, ^{136}Cs and ^{137}Cs to the device response was accounted as follows (Likhtarov et al. 2014):

$$Q_I = Q \bullet \left(1 - B_{Cs}(t_m)\right), \tag{3}$$

where $B_{Cs}(t_m)$ is the relative contribution of the cesium radioisotopes to the reading of the device at the time of measurement t_m (unitless).

The value of C_b was determined by measurement of the reference radiation source with known ^{131}I activity. From Eq. (1), assuming $G=1$ and $f_{sh}=1$ for the bottle phantom, the value of the calibration coefficient was calculated as:
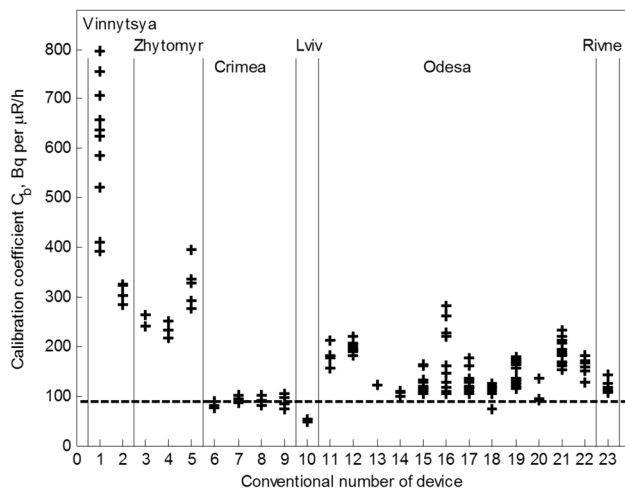
$$C_b = 10^3 \bullet \frac{Q_{ref}}{I^{meas}_{ref} - I^{meas}_{bg}}, \tag{4}$$

where Q_{ref} is the activity of ^{131}I in the reference radiation source (kBq); 10^3 is a unit conversion coefficient (Bq kBq^{-1});

**Fig. 3** Individual daily C_b-values (crosses) for the 23 SRP-68-01 devices that were calibrated in 1986. Dashed line shows C_b-value experimentally obtained by the authors (MC, SM) in 1999 for the SRP-68-01 device with a zero shift of the collimator and a bottle with ^{131}I solution

I^{meas}_{ref} is the device reading during the measurement of the reference source (μR h^{-1}).

The response of a device to a radioactive source may vary for different reasons, i.e., the parameters of the electronic components of the device might depend on temperature, due to changes in battery voltage, etc. Therefore, to minimize measurement errors, it was recommended to calibrate the device immediately prior to the measurements of the individuals. This in turn implied that the measured team should have a reference radiation source available.

The SRP-68-01 device, for which the C_b-value was estimated from measurements of a bottle phantom that contained a ^{131}I solution with known activity, is referred to as 'calibrated', in this study. For some devices, for various reasons, C_b-values were not measured; these devices are referred to as 'unchecked'. Figure 3 shows the C_b-values for 23 SRP-68-01 devices that were calibrated in May–June 1986 at least once. Dashed line in the figure shows the C_b-value experimentally obtained by the authors (MC, SM) in 1999 for the SRP-68-01 device with a zero shift of the collimator and a bottle with ^{131}I solution.

Gamma-spectrometers

Table 4 provides technical characteristics of five types of gamma-spectrometers and the number of individuals measured during thyroid monitoring in Ukraine in May–June 1986. Only one type of gamma-spectrometer, GTRM-01ts, was designed to measure ^{131}I activity in the thyroid (Fig. 4). Two types of devices, namely the GTRM-01ts and the DSU-2, allowed for the measurement of activity in the thyroid using the mode 'measured percentage of the activity of ^{131}I source'. Such measurements were considered in the present study to be the most reliable, since any problems with calibration procedure and errors related to different durations of measurement of the individuals and the calibration source were eliminated. Twelve percent of all gamma-spectrometry

Table 4 Technical characteristics of gamma-spectrometers and number of individuals measured during thyroid monitoring in May–June 1986

Name of the device	NaI(Tl) crystal (mm)	Technical characteristics	Number of measurements done by device	
			calibrated	Unchecked
GTRM-01ts	∅ 40×40	Designed to measure ^{131}I activity in the thyroid. Plastic cap on the collimator indicated the position of the thyroid gland. Mode 'measured percentage of the activity of ^{131}I source' was available	13,781	4052
NK	∅ 40×40	Multiple-purpose device (origin: Hungary) that included different collimators and counting units	9679	9642
DSU-2	∅ 63×63	Mode 'measured percentage of the activity of ^{131}I source' was available	1345	–
DSU-68	∅ 40×40	–	2953	1499
UR	∅ 63×63	Multiple-purpose device that included a set of collimators and counting blocks, including blocks with needle (pulse counting intensity) and digital indication	6768	2977
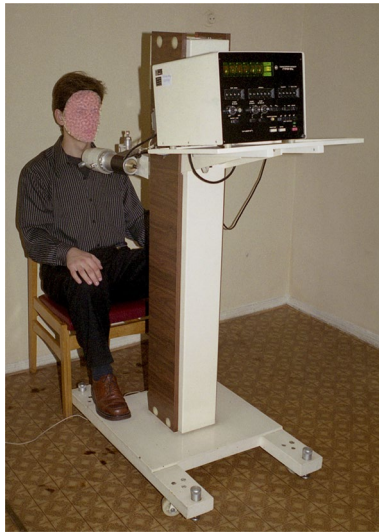
Fig. 4 Gamma-spectrometer GTRM-01ts and individual during ^{131}I thyroid activity measurement

Fig. 5 Gamma-spectrometer NK and individual during ^{131}I thyroid activity measurement

measurements was done in Ukraine using this mode, including 100% of all gamma-spectrometry measurements done in Zhytomyr Oblast.

For the NK (Fig. 5) and DSU-68 devices, as well as for some UR devices, the results of the measurements were recorded as number of counts accumulated during the time of measurement, which was from 15 to 120 s (some of the GTRM-01ts and DSU-2 devices were used in this 'accumulated number of counts' mode too), while for the other UR devices, the results of the measurements were recorded from the needle indicator as counts s^{-1}. Unfortunately, some gamma-spectrometers were not calibrated during the measurement campaign using the bottle phantom

with ^{131}I solution. Instead, the C_b-values were assigned for each of these devices by dosimetrists overseeing the thyroid dosimetry monitoring campaign. In the present study, these devices and measurements were considered to be unchecked.

Age-dependent conversion coefficients for gamma-spectrometers and the SRP-68-01 device

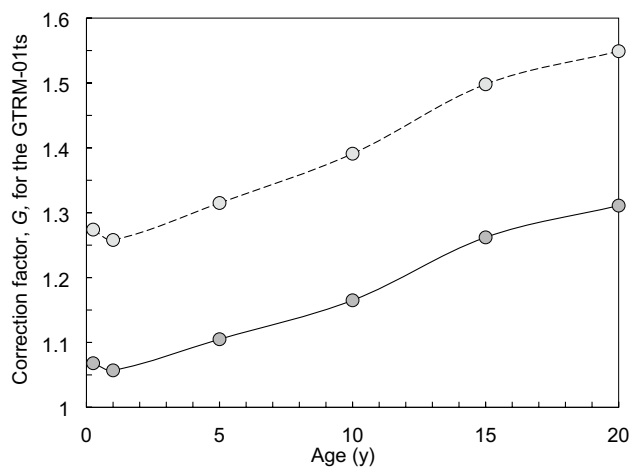The conversion coefficient, C_a, of a device depends on anthropometry of the measured individual, i.e., thyroid size and thickness of tissue overlying the thyroid gland, which both depend on age (e.g., Beaumont et al. 2018; Venturini 2003; Vilardi et al. 2018; Yunoki 2019). Age-specific C_a-values for the most commonly used Ukraine gamma-spectrometers (i.e., the GTRM-01ts and the NK which accounted for 70.5% of all spectrometric measurements) were calculated in this study using Monte Carlo simulations of photon and electron transport (Briesmeister 1997). To perform a Monte Carlo simulation, a family of Oak Ridge National Laboratory (ORNL) phantoms (Cristy 1980; Cristy and Eckerman 1987) was used. The phantoms represent the newborn; children aged 1 years, 5 years, 10 years, 15 years; and adults. Ulanovsky and Eckerman (1998) modified the ORLN phantoms to make them anatomically more realistic in the neck area. Response of the GTRM-01ts and the NK gamma-spectrometric devices on ^{131}I thyroid activity was calculated for different scenarios of collimator position: specifically, in the simulations the collimator was positioned at distances of zero, 0.5 cm and 1 cm from the skin of the neck.

The calibration coefficient, C_b, of the devices was also calculated for the reference radiation source, a glass bottle with 10 mL of ^{131}I solution, which was used for calibration in May–June 1986. Based on these calculations, age-dependent values of the correction factor, G, that accounts for the difference of the response of a device to the reference ^{131}I source and to individuals of different ages (Eq. 2) were obtained.

Figures 6 and 7 show the age-dependent values of the correction factor, G, for the GTRM-01ts and NK devices for two measurement geometries: the collimator touching the neck of a measured individual, and the collimator being at a distance of 1.0 cm from the neck surface. The G values obtained for the GTRM-01ts and NK devices can be used for other devices of that type and position of the collimator, independently from duration of measurement, width of energy window, etc.

Monte Carlo calculations of age-specific C_a-values were not performed for other types of gamma-spectrometers (i.e., the DSU-2, DSU-68 and UR devices), as such calculations are time-consuming. However, comparison of

Fig. 6 Correction factor, G, for the GTRM-01ts device. Solid line corresponds to collimator with plastic cap touching the neck, dashed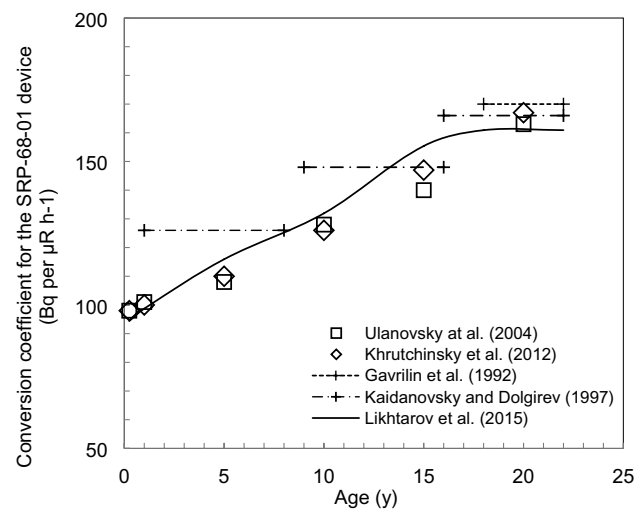 line—collimator at distance 1 cm from the neck surface. Statistical uncertainties in the Monte Carlo simulations were less than 1%

Fig. 8 Conversion coefficients, C_a, for the SRP-68-01 device without a collimator

C_a-values obtained from measurements performed by Gavrilin et al. (1992) for adults and Kaidanovsky and Dolgirev (1997) for individuals of different ages with those obtained using Monte Carlo calculations (Khrutchinsky et al. 2012; Ulanovsky et al. 2004) and using an analytical approach (Likhtarov et al. 2015). The figure demonstrates that there is reasonable agreement between experimentally obtained and calculated age-dependent C_a-values.

As was mentioned above, the SRP-68-01 devices used for direct thyroid measurements in Ukraine were equipped with a hand-made cylindrical lead collimator that surrounded the radiation detector. Figure 9 shows a sketch of the SRP-68-01 detector block with a lead collimator surrounding the radiation detector. Measurements at different locations were done by the SRP-68-01 device with different collimator shift, which is defined as distance between the edge of the detector and the edge of the collimator (Fig. 9). The calibration coefficient, C_b, of the SRP-68-01 device was calculated in this study using Monte Carlo simulations of photon and electron transport for the reference radiation source, a glass bottle with 10 mL of ^{131}I solution. Table 5 compares the calibration coefficients, C_b, simulated and measured for different shifts of the collimator. The results obtained show a reasonable agreement between the simulated and measured values of the dose rate within the measurement error of the SRP-68-01 device of 13% (DM 1986) and the statistical uncertainty of the Monte Carlo simulations (see Table 5).

Experimental data on age-dependent conversion coefficients, C_a, were not available for the SRP-68-01 device with the collimator. Figure 10 shows the age-dependent C_a-values for different shifts of the collimator that were calculated by Likhtarov et al. (2015) using an analytical approach.

Fig. 7 Correction factor, G, for the NK device. Solid line corresponds to collimator touching the neck, dashed line—collimator at a distance of 1 cm from the neck surface. Statistical uncertainties in the Monte Carlo simulations were less than 1%

C_a-values for the GTRM-01ts and NK devices obtained in this study with those calculated by Likhtarov et al. (2015) showed reasonable agreement. Therefore, for the DSU-2, DSU-68 and UR devices, the age-dependent C_a-values calculated by Likhtarov et al. (2015) were used in the present study.

In contrast to gamma-spectrometers, for which age-dependent C_a-values had to be calculated, experimental data on age-dependent C_a-values were available for the SRP-68-01 device (Gavrilin et al. 1992; Kaidanovsky and Dolgirev 1997). Figure 8 compares the age-dependent
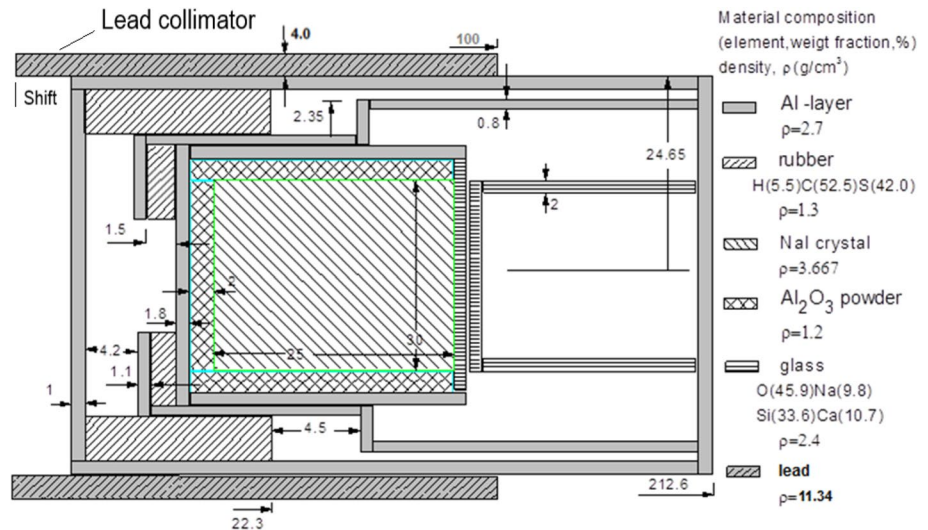
Fig. 9 Sketch of the SRP-68-01 detector block with lead collimator surrounding the radiation detector
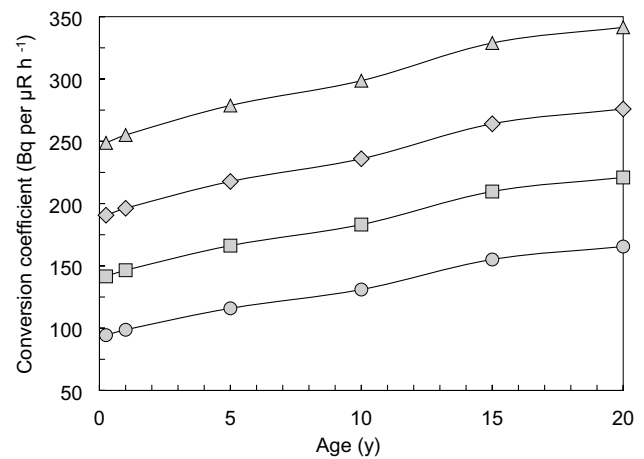

Table 5 Calibration coefficients, C_b, of the SRP-68-01 device, for the reference radiation source, a glass bottle with 10 mL of ^{131}I solution, simulated and measured for different shifts of the collimator

Shift of collimator (cm)	Calibration coefficient, C_b, (Bq per μR h^{-1})	
	Monte Carlo simulations	Results of measurements
0	89 ± 3[a]	92 ± 12[b]
1	143 ± 4	141 ± 18
2	208 ± 4	192 ± 25
3	286 ± 4	250 ± 35
5	476 ± 5	417 ± 54

[a]Statistical uncertainty in the Monte Carlo simulations depends on the shift of the collimator

[b]Measurement error is 13%, which is the typical 1-sigma uncertainty in a measurement made by the SRP-68-01 instrument (DM 1986)

Fig. 10 The age-dependent calibration coefficients, C_a, for the SRP-68-01 device with different shifts of a collimator (Likhtarov et al. 2015): crosses—0 cm, squares—1 cm, diamonds—2 cm, triangles—3 cm

Comparative analysis to evaluate the correct measurement geometry and calibration coefficients for unchecked devices

Comparative analyses were conducted as described below, whenever possible, to evaluate the correct measurement geometry and calibration coefficients for (1) unchecked SRP-68-01 devices and (2) for gamma-spectrometers, for cases in which the correct measurement methodology could not be applied (e.g., when a device was used to measure more than 700 persons per day, see below). The measurement geometry includes the position of the detector with regard to an individual during the thyroid measurement procedure, the anthropometric parameters

of the measured individual, the size and shift of the collimator, and other factors.

The following hypothesis was the basis for the analysis: the frequency distributions of ^{131}I activity in the thyroid measured by different devices in residents of the same or closely located settlements should show a similar pattern. Frequency distribution of ^{131}I thyroid activity derived from reliable measurements, i.e., done using a calibrated gamma-spectrometer or an SRP-68-01 device, was considered in this study to be 'trusted'. The analysis indicated that the distribution of measured ^{131}I thyroid activity among the residents of the same settlement is approximately log-normal with a geometric standard deviation (GSD) of approximately 2.0. Therefore, if the median of the distribution obtained using

an 'unchecked' device, e.g., an uncalibrated SRP-68-01 with unknown shift of the collimator, deviated by more than a factor 1.5 from those obtained using a trusted device, this justified the need to adjust the C_b-values of an unchecked device, since such a deviation is statistically significant even for a small numbers of measured individuals.

Correction was done using a device-dependent adjustment factor (AF) that was calculated in the present study. Frequency distributions of ^{131}I thyroid activity measured by trusted and unchecked devices were compared for the same 'cluster', i.e., using ^{131}I activities measured by the same devices in the same settlement or group of settlements.

To decrease dispersion of the distribution of ^{131}I thyroid activity, measurement results obtained for individuals aged from 8 to 16 years old were included in the analysis, because they were most frequent. To ensure that the results of such a comparative analysis were reliable and the AF-values obtained were correct, the following measurements were included:

1. Measurements performed between 18 May and 5 June 1986 when most direct thyroid measurements were done in Ukraine. Because of the short half-life of ^{131}I in the thyroid, which is defined by its physical half-life of 8.02 days, ^{131}I thyroid activities were recalculated to the same date of 31 May 1986 to ensure correct comparison of activities measured within a period of 19 days. Measurements done before 18 May and later than 5 June were not included in the analysis.

2. To increase the number of measurements included in analysis, activities of ^{131}I in the thyroid measured in residents of neighboring settlements were normalized to the ^{131}I ground deposition density in the settlement of residence, which was calculated using an improved atmospheric dispersion model (Talerko et al. 2020) (see above).

The device's adjustment factor was calculated as weighted mean of the ratios of 'trusted' to 'unchecked' median of the distribution of ^{131}I thyroid activity measured at all clusters where the unchecked device was used:

$$\mathrm{AF}_j = \frac{\sum_i \frac{\hat{Q}_i^{\mathrm{trust}}}{\hat{Q}_{i,j}^{\mathrm{uncheck}}} \cdot W_{i,j}}{\sum_i W_{i,j}} = \frac{\sum_i A_{i,j} \cdot W_{i,j}}{\sum_i W_{i,j}}, \text{where } W_{i,j} = \frac{N_i^{\mathrm{trust}} \cdot N_{i,j}^{\mathrm{uncheck}}}{N_i^{\mathrm{trust}} + N_{i,j}^{\mathrm{uncheck}}}, \tag{5}$$

where AF_j is the adjustment factor for the j-th unchecked device (unitless); $\hat{Q}_i^{\mathrm{trust}}$ is the median of the distribution of the trusted ^{131}I thyroid activity measured in the i-th cluster that was recalculated to 31 May 1986 and normalized to ^{131}I ground deposition density (Bq per kBq m^{-2}); $\hat{Q}_{i,j}^{\mathrm{uncheck}}$ is the median of the distribution of ^{131}I thyroid activity measured by the j-th unchecked device in the i-th cluster that was

recalculated to 31 May 1986 and normalized to ^{131}I ground deposition density (Bq per kBq m^{-2}); N_i^{trust} and $N_{i,j}^{\mathrm{uncheck}}$ is the number of measurements that were done in i-th cluster by the trusted and j-th unchecked devices, respectively. A cluster was used to calculate the device adjustment factor if more than ten measurements of ^{131}I thyroid activity had been performed by each trusted and unchecked device.

If AF_j was less than 1.5, which corresponds to a shift of the collimator for the SRP-68-01 device of less than 1 cm, calibration coefficients for this device were considered to be correct. Otherwise, the C_b-values for the unchecked device was corrected as:

$$C_b^{j,\,\mathrm{corr}} = C_b^j \bullet AF_j, \tag{6}$$

where $C_b^{j,\,\mathrm{corr}}$ is the corrected calibration coefficient for the j-th unchecked device (Bq per μR h^{-1}); C_b^j is the calibration coefficient used for the j-th unchecked device in TD10 (Bq per μR h^{-1}).

The correct geometry of measurement, i.e., the correct shift of the collimator, was evaluated for the SRP-68-01 device by a comparison of the estimated corrected calibration coefficient, $C_b^{j,\mathrm{corr}}$, and calibration coefficients obtained for different shifts of a collimator (Table 5). Values of correction factor, G, which were appropriate for a given measurement geometry, were then used to calculate conversion coefficients, C_a, to be used to derive ^{131}I thyroid activity in individuals of different ages.

Figure 11 shows the schema of the comparative analysis and estimation of the AF-values. The revision of direct thyroid measurements and introduction of adjustment factors resulted in new sources of shared and unshared errors related to the estimates of ^{131}I thyroid activity in the calculation of individual stochastic doses.

Estimation of measurement errors

Errors in the measured ^{131}I thyroid activity were estimated for each individual measurement. The classical and Berkson components of the errors were evaluated separately using a combination of models, because the influence of these components on estimates of radiation-related risk of thyroid cancer following exposure to ^{131}I are considerably different (Masiuk et al. 2016, 2017). The following errors in the measurement of the ^{131}I thyroid activity were identified:

1. Unshared classical errors including those associated with the device response during the measurements of ^{131}I in the thyroid and in the bottle phantom containing a reference ^{131}I solution, the device's measurement errors and the errors associated with age-depended individual

Fig. 11 Schema of the comparative analysis applied in the present study to estimate adjustment factors for unchecked devices


```
┌─────────────────────────┐                                          ┌─────────────────────────┐
│ 'Trusted' activity of    │                                          │ Activity of 131I in the  │
│ 131I in the thyroid      │                                          │ thyroid measured by the  │
│ measured in residents of │                                          │ j-th unchecked device in │
│ the i-th cluster         │                                          │ residents of the i-th    │
└─────────────────────────┘                                          │ cluster                  │
              ↑                  ┌──────────────────────────┐         └─────────────────────────┘
              │                  │ Recalculation of 131I    │                     │
              │                  │ activity in the thyroid  │                     │
              │←─────────────────│ measured in the i-th     │────────────────────→│
              │                  │ cluster, recalculated to │                     │
              │                  │ 31 May 1986              │                     │
              ↓                  └──────────────────────────┘                     ↓
┌─────────────────────────┐                                          ┌─────────────────────────┐
│ 'Trusted' 131I activity  │                                          │ 131I activity in the     │
│ in the thyroid measured  │                                          │ thyroid measured by the  │
│ in residents of the i-th │                                          │ j-th unchecked device in │
│ cluster, recalculated to │                                          │ residents of the i-th    │
│ 31 May 1986              │                                          │ cluster, recalculated to │
└─────────────────────────┘                                          │ 31 May 1986              │
              ↑                  ┌──────────────────────────┐         └─────────────────────────┘
              │                  │ Normalizing of 131I      │                     │
              │                  │ activity in the thyroid  │                     │
              │←─────────────────│ to 131I ground           │────────────────────→│
              │                  │ deposition density in    │                     │
              │                  │ every settlement of the  │                     │
              │                  │ i-th cluster             │                     │
              ↓                  └──────────────────────────┘                     ↓
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'Trusted' activity of ^{131}I in the thyroid measured in residents of the i-th cluster

Activity of ^{131}I in the thyroid measured by the j-th unchecked device in residents of the i-th cluster

Recalculation of ^{131}I activity in the thyroid measured in the i-th cluster, recalculated to 31 May 1986

'Trusted' ^{131}I activity in the thyroid measured in residents of the i-th cluster, recalculated to 31 May 1986

^{131}I activity in the thyroid measured by the j-th unchecked device in residents of the i-th cluster, recalculated to 31 May 1986

Normalizing of ^{131}I activity in the thyroid to ^{131}I ground deposition density in every settlement of the i-th cluster

Distribution of 'trusted' ^{131}I activity in the thyroid measured in residents of the i-th cluster, recalculated to 31 May 1986 and normalized to ^{131}I ground deposition density, $\hat{Q}_i^{trust}$

Distribution of ^{131}I activity in the thyroid measured by the j-th unchecked device in residents of the i-th cluster, recalculated to 31 May and normalized to ^{131}I ground deposition density, $\hat{Q}_i^{uncheck}$

Adjustment factor of j-th unchecked device: central value, AF_j, and parameters of distribution (Eq. (5))

anthropometric characteristics (thyroid volume, thyroid location, etc.).

2. Unshared Berkson errors associated with the deviation of the thyroid detector's position from the proper measurement geometry.

3. Shared Berkson errors associated with the result of the comparative analysis and with the estimates of the AF-values used to correct the calibration coefficients of the unchecked gamma-spectrometers and SRP-68-01 devices.

The Appendix provides a detailed description of the methods used to define and calculate these errors. It also provides the mathematical model of measurement results including classical and Berkson errors that can be used for dose calculations and for further radiation-related thyroid cancer risk analysis.

Results

Comparative analysis to evaluate the correct measurement geometry and calibration coefficients for unchecked SRP-68-01 devices

Measurements among residents of Chernihiv Oblast

Comparative analysis, which was conducted for the unchecked SRP-68-01 devices used for direct thyroid measurements in Chernihiv Oblast, showed that the collimators were used without shift meaning that the edge of the collimator coincided with the edge of the detector. This fact was confirmed by witnesses and participants of the thyroid dosimetry monitoring campaign. There were two more uncalibrated SRP-68-01 devices with unknown shift of the collimator, i.e., unchecked SRP-68-01 devices that were used in Sumy Oblast to measure residents of Chernihiv Oblast (Table 6). An adjustment factor AF=1.55 was obtained for both devices, and the corrected calibration coefficient was

Table 6 Comparative analysis for the unchecked SRP-68-01 device that was used in Sumy Oblast to measure residents of two raions in Chernihiv Oblast

Raion	Number of i-th cluster	Unchecked SRP-68-01		Trusted gamma-spectrometer				Ratio of $\widehat{Q}_i^{\text{trust}}$ to $\widehat{Q}_i^{\text{uncheck}}$
		Number of measurements	$\widehat{Q}_i^{\text{uncheck}}$(Bq per kBq m^{-2})	Type of device	Serial number of device	Number of measurements	$\widehat{Q}_i^{\text{trust}}$(Bq per kBq m^{-2})	
Ripky	1618	58	126	UR	912001	73	219	1.73
	1624	20	10.1	UR	912001	24	19.1	1.90
	1627–1	23	99	UR	912001	29	180	1.82
	1627–3	28	42	UR	912001	32	81	1.94
Chernihiv	2109	118	267	UR	912001	41	246	0.92
				NK	81031	77	363	1.36
	2114	83	317	NK	71070	86	413	1.30
	2127	63	263	NK	71077	66	506	1.92
				NK	81031	51	429	1.63
				UR	912001	32	370	1.41
	2129	31	520	NK	81031	42	833	1.60
Adjustment factor[a], *AF*								1.55
Shift of the SRP-68-01 device's collimator according to Table 5								1.0 cm

[a] Adjustment factor is the weighted mean of the ratios of $\widehat{Q}_i^{\text{trust}}$ to $\widehat{Q}_i^{\text{uncheck}}$ (see Eq. (5))

estimated using Eq. (6) to be $C_b^{j,\text{corr}}$=143 Bq μR h^{-1}. According to Table 5, this calibration coefficient corresponded to a collimator shift of 1 cm.

Measurements among residents of Zhytomyr Oblast

Eleven SRP-68-01 devices were used for thyroid dosimetry monitoring among residents of Zhytomyr Oblasts; only three of these devices were calibrated (once on 20 May 1986) and their collimators were shifted by 3 cm (Table 5). Comparative analysis was conducted only for three out of eight unchecked SRP-68-01 devices, because only an insufficient number of measurements were done by means of the trusted devices (gamma-spectrometers GTRM-01ts and calibrated SRP-68-01 devices). As a result, the measurement geometry was evaluated for three devices with a collimator shift of 0, 1 and 3 cm. Corrected calibration coefficients, $C_b^{j,\text{corr}}$, and appropriate age-dependent correction factors, G, were assigned to these SRP-68-01 devices. For the other five unchecked SRP-68-01 devices, a zero shift of the collimator was assumed.

Measurements among residents of Kyiv Oblast

It was impossible to conduct a reliable comparative analysis of measurements performed on children and adolescent residents of Kyiv Oblast because they (1) moved shortly after the accident to the non-contaminated Odesa Oblast and Crimea and were measured there, and (2) the dates of relocation from the contaminated settlements were unknown for

individuals who were not included in the Ukrainian-American cohort. However, about 96% of the Ukrainian-American cohort members measured on Crimea and in Odesa Oblast were measured by calibrated SRP-68-01 devices. The same geometry of measurement was assigned for the rest of residents of Kyiv Oblast measured by SRP-68-01 devices, i.e., a zero collimator shift was assumed for Crimea and a 1 cm collimator shift was assumed for Odessa Oblast (see Fig. 3; Table 5).

Comparative analysis to evaluate the correct measurement geometry for gamma-spectrometers

To evaluate possibly unreliable results of the gamma-spectrometry measurements, the number of measurements done using the same device at the same day was analyzed. The minimal time required to measure one individual, including the correct positioning of the individuals and detector against the neck, was at least 1 min. Consequently, a maximum of about 700 individuals could be measured during an extended 12-h working day. However, for some days, up to 2000 persons were measured by the same device in Kozeletsky raion in Chernihiv Oblast (Table 7). It is emphasized that it was not possible to complete such a large number of measurements without violation of the requirements for standard measurement procedures, including correct positioning of the individuals and the detector against the thyroid. Improper measurement geometry may have resulted in an inaccurate estimate of ^{131}I activity in the thyroid as the calibration coefficient used was obtained assuming the standard measurement geometry.

Table 7 Number of daily measurements performed by the GTRM-01ts device in Kozeletsky raion in Chernihiv Oblast

Date in 1986	Number of measurements per day	Time needed to perform the measurements (h)[a]
24 May	394	6.6
25 May	624	10.4
26 May	516	8.6
27 May	580	9.7
28 May	414	6.9
29 May	381	6.4
30 May	798	13.3
31 May	1918	32.0
1 June	799	13.3

[a]Assuming "at least one minute per measurement"

The comparative analysis showed that calibration coefficients for two gamma-spectrometers GTRM-01ts used in Chernihiv Oblast were underestimated. These devices were characterized by an unrealistically large number of daily measurements done between 30 May and 1 June 1986 (Table 7), and an improper measurement geometry could be the possible reason for the underestimation of the calibration coefficients. Table 8 provides an example of the comparative analysis performed for one of the GTRM-01ts devices with an underestimated calibration coefficient that was used in Kozeletsky raion in Chernihiv Oblast. As a result of the comparative analysis, an adjustment factor $AF = 4.55$ was obtained for the GTRM-01ts device, and the corrected calibration coefficient was calculated using Eq. (6). The same analysis was done for the second GTRM-01ts device with an underestimated calibration coefficient, for which a factor

$AF = 2.0$ was obtained. It should be noted that no problems were identified during the comparative analysis for the other gamma-spectrometers used for the thyroid dosimetry monitoring in Ukraine.

^{131}I activity in the thyroid

Table 9 compares the ^{131}I thyroid activities for all 146,425 measured residents of Ukraine that were estimated in this study with those previously estimated by Likhtarov et al. (2005). The highest ^{131}I thyroid activity at the time of measurement was found in residents of Zhytomyr, Kyiv, and Chernihiv Oblasts, with arithmetic means among the measured individuals of 18 kBq, 12 kBq, and 10 kBq, respectively. The lowest ^{131}I thyroid activity (arithmetic mean of 0.29 kBq) was observed among residents of Lviv Oblast, located about 450 km to the west of the Chornobyl nuclear power plant. The revised activity of ^{131}I in the thyroid among residents of Kyiv city was lower than that for Kyiv Oblast, 2.3 kBq vs. 12 kBq. This observation can be explained by the fact that residents of Kyiv city did not consume fresh cow's milk while milk from trade network was controlled for the permissible level of radioactivity of 3.7 kBq L^{-1} starting 6 May 1986 (MH 1986). Rather wide frequency distributions of ^{131}I thyroid activity were observed in different oblasts with GSD values that varied from 2.4 to 5.3. On average, in Vinnytsia, Kyiv, Lviv, and Chernihiv Oblasts, and in the city of Kyiv, the revised values of the ^{131}I thyroid activities were found to be 10–25% higher than previously estimated by Likhtarov et al. (2005), while in Zhytomyr Oblast, the values of the revised activities were found to be about 50% lower.

Figure 12 shows the overall frequency distribution of ^{131}I thyroid activities at the time of measurement (logarithm

**Table 8** Example of comparative analysis for the GTRM-01ts device

Raion	Number of i-th cluster	Unchecked GTRM-01ts		Trusted SRP-68-01 devices			Ratio of $\widehat{Q}_i^{\text{trust}}$ to $\widehat{Q}_i^{\text{uncheck}}$
		Number of measurements	$\widehat{Q}_i^{\text{uncheck}}$ (Bq per kBq m^{-2})	Serial number of device	Number of measurements	$\widehat{Q}_i^{\text{trust}}$ (Bq per kBq m^{-2})	
Kozeletsky	0703	280	22.4	957	642	100.1	4.48
		280	22.4	906	10	56.2	2.51
	0704	40	43.7	957	54	179.5	4.10
	0707	11	68.3	957	89	219.2	3.21
	0710	12	42.9	957	77	187.2	4.37
	0713	62	22.3	957	19	124.6	5.60
	0718	45	32.5	957	25	239.2	7.35
	0723	57	79.7	906	51	186.6	2.34
	0725	88	49.1	957	41	360.8	7.35
	0734	64	46.3	957	24	157.4	3.40
	0735	16	42.8	957	39	197.4	4.61
Adjustment factor [a], AF							4.55

[a] Adjustment factor is the weighted mean of the ratios of $\widehat{Q}_i^{\text{trust}}$ to $\widehat{Q}_i^{\text{uncheck}}$ (see Eq. (5))

Table 9 Parameters of distribution of [131]I thyroid activity by oblast of residence at the time of the accident (ATA): arithmetic mean (AM), stan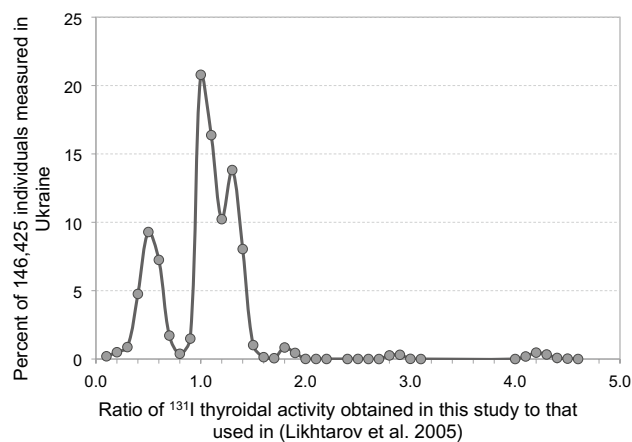dard deviation (SD), geometric mean (GM) and geometric standard deviation (GSD), obtained in the present study and comparison with those previously estimated by Likhtarov et al. (2005)

Oblast of residence ATA	N	Dates of measurements in 1986	[131]I activity in the thyroid (kBq)									
			This study					Previously estimated by Likhtarov et al. (2005)				
			5–95%	AM	SD	GM	GSD	5–95%	AM	SD	GM	GSD
Kyiv	37,504	30 April–30 June	0.95–41	12	25	5.4	3.3	0.80–36	10	22	4.6	3.2
Kyiv city	4923	16 May–30 June	0.48–6.2	2.3	4.2	1.4	2.4	0.41–6.2	2.1	7.7	1.2	2.4
Zhytomyr	53,009	15 May–30 June	0.81–70	18	37	7.2	4.1	0.93–143	34	78	11	4.5
Chernihiv	48,105	30 April–30 June	0.70–35	10	27	4.3	3.5	0.44–28	8.0	23	3.2	3.6
Vinnytsia	1989	17 May–10 June	0.19–31	7.4	10.3	2.7	5.3	0.22–20	5.9	7.3	2.6	4.3
Lviv	475	27 May–20 June	0.026–0.94	0.29	0.33	0.16	3.1	0.037–0.81	0.25	0.29	0.16	2.5
Total[a]	146,425	30 April–30 June	0.69–49	13	31	5.2	3.9	0.56–68	18	51	5.3	4.4

[a]Including 420 individuals who resided outside the listed Oblasts
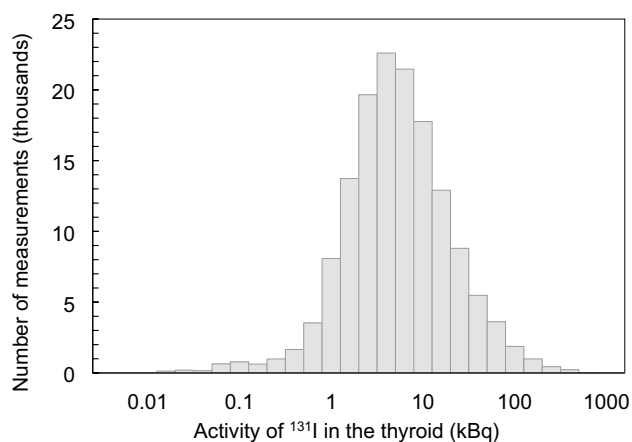
Fig. 12 Frequency distribution of revised [131]I thyroid activity at the time of measurement (logarithm of actual value) among 146,425 individuals measured in Ukraine

Fig. 13 Frequency distribution of the ratios of [131]I thyroid activity obtained in the present study to that previously assessed by Likhtarov et al. (2005) for all 146,425 individuals measured in Ukraine

of actual value) among 146,425 individuals measured in Ukraine. This distribution is close to a log-normal distribution with a geometric mean (GM) equal to 5.2 kBq and GSD equal to 3.9. The range of [131]I activities in the thyroid measured in the Ukrainian residents is rather wide, and the 90% confidence interval is 0.69–49 kBq. Figure 13 shows the frequency distribution of the ratios of [131]I thyroid activity obtained in the present study to that previously assessed by Likhtarov et al. (2005) for all 146,425 individuals measured in Ukraine. Values of the ratio of less than 1 represent residents of Zhytomyr Oblast, while values of the ratio between 1 and 2 represent individuals from Kiev, Vinnitsa and Chernihiv Oblasts. For some residents of Chernihiv Oblast, this ratio exceeds 2.

Activity of [131]I in the thyroid for the members of the Ukrainian-American cohort, which was revised as described above (TD20), was compared with that previously used for TD10 by Likhtarov et al. (2014). Figure 14 compares the [131]I activity in the thyroid, TD20 vs. TD10, by Oblast of residence of the Ukrainian-American cohort members. For about 40% of the cohort members residing in Zhytomir Oblast, the revised values of [131]I thyroid activity (TD20) were found to be 2–3 times lower as compared to those used for previous dose assessment in TD10. For cohort members who resided in Kiev and Chernihiv Oblasts, the revised values of [131]I activity in the thyroid tended to be higher than those from TD10.

Errors in measurements of [131]I thyroid activity

Table 10 provides device- and Oblast-specific classical and Berkson relative errors associated with the measurements of
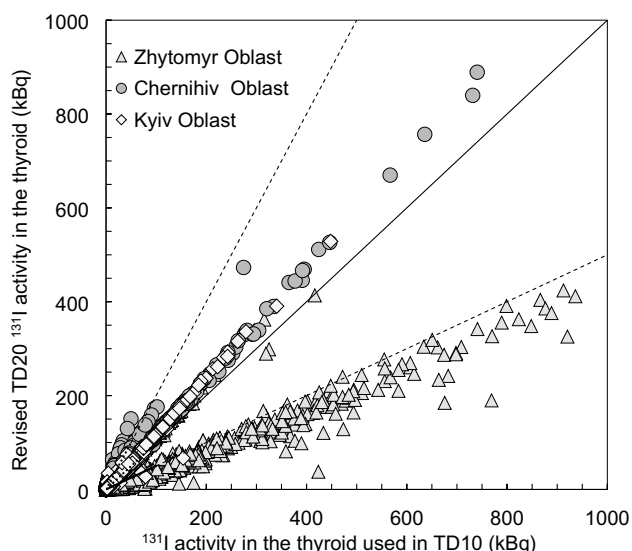
Fig. 14 Comparison ^{131}I activity in the thyroid, TD20 vs. TD10, by Oblast of residence of the Ukrainian-American cohort members. Solid line represents a ratio of TD10-to-TD20 ^{131}I activity in the thyroid equal to 1. Dashed lines show a factor of 2 difference between the two sets of activities

^{131}I activity in the thyroid for the members of the Ukrainian-American cohort. The highest classical errors in measured ^{131}I thyroid activity were in Chernihiv Oblast, including about 0.5 for the measurements done by the UR and SRP-68-01 devices. For the entire Ukrainian-American cohort, devices ranked highest to lowest in terms of classical errors are: the UR, SRP-68-01, GTRM-01ts, NK, DSU-68 and DSU-2.

The highest relative Berkson errors in ^{131}I thyroid activity measurements were observed for the Chernihiv Oblast, including 0.86 for the measurements done by the GTRM-01ts device. Such high errors were observed because the calibration coefficients for two gamma-spectrometers GTRM-01ts which were used in Chernihiv Oblast to measure ^{131}I activities in 930 out of 1546 individuals (60.2% of the total

measured by this type of device) were underestimated by a factor of 4.55 (see Section on "Comparative analysis to evaluate the correct measurement geometry for gamma-spectrometers" above). For the entire Ukrainian-American cohort, devices ranked highest to lowest in terms of Berkson errors are: the GTRM-01ts, SRP-68-01, NK, UR, DSU-2 and DSU-68.

Discussion

The present paper describes an in-depth analysis and revision of directly measured thyroid ^{131}I activities done in the Ukrainian population in May–June 1986. The variation of the C_b-values for the SRP-68-01 device that was observed in TD10 from one oblast to another (Fig. 3) might be caused, to a large extent, by different positions of the lead collimators that affected the distance between the thyroid gland and the radiation detector. For example, a shift of the collimator from the edge of the detector of up to 3 cm resulted in changes of the C_b-values for the SRP-68-01 device of up to factor of 2.5.

A substantial age dependence of a device's conversion coefficients was observed for the SRP-68-01 device, and a ratio of C_a-value for an adult individual to that for a 1-year-old child was 1.75 (Fig. 8). It should be noted that the Hitachi-Aloka TCS-161/171/172 survey meter including a NaI(Tl) crystal ($\varnothing$25.4 mm × 25.4 mm length), similar to the crystal size of the SRP-68-01 device, was used after the Fukushima-Daiichi accident in Japan for measuring the ^{131}I thyroid activity, has a similar age dependence of C_a-values (Kim et al. 2020).

For gamma-spectrometers, the age dependence was less pronounced, and the corresponding ratio was 1.23 and 1.13 for the GTRM-01ts and NK devices, respectively (Figs. 6, 7). Such dependence on age is quite typical for devices including a NaI(TI) crystal with a diameter and thickness of about 2 inches (Li et al. 2019).

Table 10 Classical and Berkson relative errors (ratio of standard deviation to central value of ^{131}I activity) associated with the measurements of ^{131}I activity in the thyroid for the members of the Ukrainian-American cohort using various devices

Oblast	SRP-68-01	GTRM-01ts	NK	DSU-2	DSU-68	UR
Classical errors						
Kyiv and Kyiv city	0.29	0.14	0.17	0.10	–	0.24
Zhytomyr	0.42	0.12	0.09	–	–	–
Chernihiv	0.50	0.32	0.30	–	0.26	0.50
Entire cohort	0.42	0.29	0.27	0.10	0.26	0.42
Berkson errors						
Kyiv and Kyiv city	0.10	0.04	0.05	0.02	0.02	0.05
Zhytomyr	0.13	0.04	0.06	–	–	–
Chernihiv	0.13	0.86	0.05	–	0.02	0.05
Entire cohort	0.12	0.76	0.05	0.02	0.02	0.05

The shape, size and position of the lobes and the isthmus of the thyroid gland are variable (e.g., Hiatt and Gartner 2010). Therefore, the position of the thyroid gland relative to the center of the detector could also vary between individuals who underwent direct thyroid measurements. For the SRP-68-01 device without a collimator, shifts of up to 2 cm in position of the thyroid relative to the detector resulted in only small relative error estimates for the ^{131}I thyroid activity, i.e., up to 0.07 (ratio of standard deviation to central value). However, for the thyroid detectors with collimators, the radiation from ^{131}I in the thyroid could be shielded by the collimator to different degrees depending on the position of the collimator relative to the thyroid of the measured individual. This leads to increasing measurement uncertainty that is hard to quantify for a particular individual, because of the unknown position and shape of his or her thyroid gland.

Comparative analysis showed a significant variability between individual ^{131}I thyroid activities that were derived even from calibrated measurements. As a rule, the frequency distribution of ^{131}I thyroid activities among individuals measured within the same cluster was approximately lognormal and characterized by a GSD value that was typically greater than 2. The observed variability in ^{131}I thyroid activities is determined by the variability in parameters values of ecological and biokinetic models that define the transfer of ^{131}I through the chain 'ground deposition' → 'vegetation' → 'milk, milk products' → 'human's thyroid'. The dosimetry model used to calculate thyroid doses in the Ukrainian-American cohort uses more than 50 parameters (Likhtarov et al. 2014). Differences in parameters of the frequency distributions of ^{131}I thyroid activity measured by trusted and unchecked devices in the same cluster might be caused by statistical uncertainty and variability of values of the parameters considered in the dosimetry model. Therefore, adjustment factors were calculated as a weighted mean of the ratios of 'trusted' to 'unchecked' median of the distribution of ^{131}I thyroid activity measured among more than ten persons in one cluster (see Eq. 5).

Conclusion

This paper describes an in-depth analysis and revision of 146,425 measurements of ^{131}I thyroid activity in the Ukrainian population done in May–June 1986. A comparative analysis was conducted, where possible, to evaluate the measurement geometry and calibration coefficients for unchecked SRP-68-01 devices and gamma-spectrometers. New sources of shared and unshared errors associated with ^{131}I thyroid activity measurements were identified. The revised activities of ^{131}I in the thyroid and associated classical and Berkson measurement errors were derived from direct thyroid measurements done in Ukraine. The results

obtained provide the basis for the assessment of updated thyroid doses in Ukraine due to ^{131}I intake. The revision of ^{131}I thyroid activities will result in substantial changes of the thyroid dose estimates for the entire population of Ukraine (Likhtarev et al. 2005) as well as for the subjects of the Ukrainian-American cohort (Likhtarov et al. 2014), the Ukrainian in utero cohort (Likhtarov et al. 2011), and individuals included in the Chornobyl Tissue Bank (Likhtarov et al. 2013b).

Appendix. Errors in measured ^{131}I thyroid activities

Estimation of classical measurement errors for ^{131}I in the thyroid

It is known that at the fixed intensity of emission for a radioactive source, the probability to register k counts using a measuring device for measuring time t is defined by the Poisson distribution (Molina 1973). For a quite large intensity, the Poisson distribution is close to a normal distribution and can be written as:

$$I_{\text{th}}^{\text{meas}} \sim N\left(I_{\text{th}}^{\text{tr}}, \sigma_{\text{th}}^2\right), I_{\text{bg}}^{\text{meas}} \sim N\left(I_{\text{bg}}^{\text{tr}}, \sigma_{\text{bg}}^2\right), \tag{A1}$$

where $N(m, \sigma^2)$ is a normal distribution with expectation value m and variance σ^2, $I_{\text{th}}^{\text{meas}}$ and $I_{\text{bg}}^{\text{meas}}$ are intensities of a radioactive source (providing the reading of a device in terms of pulses per second) registered during the measurement of thyroid and background, respectively, $\sigma_{\text{th}}^2 = \frac{I_{\text{th}}^{\text{tr}}}{t_{\text{th}}}$ and $\sigma_{\text{bg}}^2 = \frac{I_{\text{bg}}^{\text{tr}}}{t_{\text{bg}}}$ are the variances of corresponding measurement errors, t_{th} is the duration of a thyroid measurement, and t_{bg} is the duration of a background measurement. Index 'tr' denotes the true value, while 'meas' denotes the measured value.

In addition to the statistical error of registration, the values $I_{\text{th}}^{\text{meas}}$ and $I_{\text{bg}}^{\text{meas}}$ include an instrumental error, with variance σ_{dev}^2. The full variances of the measurement errors for both thyroid and background are as follows:

$$\widehat{\sigma}_{\text{th}}^2 = \frac{I_{\text{th}}^{\text{meas}}}{t_{\text{th}}} + \sigma_{\text{dev}}^2 \text{ and } \widehat{\sigma}_{\text{bg}}^2 = \frac{I_{\text{bg}}^{\text{meas}}}{t_{\text{bg}}} + \sigma_{\text{dev}}^2. \tag{A2}$$

Based on the calibration method used one can write down the approximate relation:

$$C_a^{\text{meas}} \approx C_a^{\text{tr}} \bullet \left(1 + \delta_C \bullet \gamma_1\right), \quad \gamma_1 \sim N(0,1), \tag{A3}$$

where C_a is the conversion coefficient (Eq. 2) and δ_C is the relative error of the conversion coefficient, which includes the error of the ^{131}I activity in the bottle source used for the

device calibration, the device's error of the measurement, and the error of the age-dependent factor G.

Using Eqs. (A1)−(A3), ^{131}I activity in the thyroid estimated from the direct thyroid measurement (see Eq. 1) can be presented as:

$$Q^{\text{meas}} \approx C_a^{\text{tr}} \bullet \left(1 + \delta_C \bullet \gamma_1\right) \bullet \left(I_{\text{th}}^{\text{tr}} - f_{\text{sh}} \bullet I_{\text{bg}}^{\text{tr}} + \sigma_n \bullet \gamma_2\right), \quad (A4)$$

where $\sigma_n = \sqrt{\hat{\sigma}_{\text{th}}^2 + f_{\text{sh}}^2 \bullet \hat{\sigma}_{\text{bg}}^2}$ and $\gamma_2 \sim N(0,1)$.

Then, Eq. (A4) can be written as:

$$Q^{\text{meas}} \approx C_a^{\text{tr}} \left(I_{\text{th}}^{\text{tr}} - f_{\text{sh}} \bullet I_{\text{bg}}^{\text{tr}} + \left(I_{\text{th}}^{\text{tr}} - f_{\text{sh}} \bullet I_{\text{bg}}^{\text{tr}}\right)\delta_C \bullet \gamma_1 + \sigma_n \bullet \gamma_2 + \delta_C \bullet \sigma_n \bullet \gamma_1 \bullet \gamma_2\right). \quad (A5)$$

Because $Q^{\text{tr}} = C_a^{\text{tr}} \bullet (I_{\text{th}}^{\text{tr}} - f_{\text{sh}} \bullet I_{\text{bg}}^{\text{tr}})$, Eq. (A5) can be expressed as:

$$Q^{\text{meas}} \approx Q^{\text{tr}} + C_a^{\text{tr}} \bullet \left(\sigma_n \bullet \gamma_2 + \left(I_{\text{th}}^{\text{tr}} - f_{\text{sh}} \bullet I_{\text{bg}}^{\text{tr}}\right) \bullet \delta_C \bullet \gamma_1 + \delta_C \bullet \sigma_n \bullet \gamma_1 \bullet \gamma_2\right) \approx Q^{\text{tr}} + \sigma_Q^{\text{tr}} \bullet \gamma, \quad (A6)$$

where $\sigma_Q^{\text{tr}} = C_a^{\text{tr}} \bullet \sqrt{\sigma_n^2 + \sigma_n^2 \bullet \delta_C^2 + (I_{\text{th}}^{\text{tr}} - f_{\text{sh}} \bullet I_{\text{bg}}^{\text{tr}})^2 \bullet \delta_C^2}$ and $\gamma \sim N(0,1)$.

As $I_{\text{th}}^{\text{tr}}$ and $I_{\text{bg}}^{\text{tr}}$ are unknown, σ_Q^{tr} can be written as:

$$\sigma_Q^{\text{meas}} = C_a^{\text{meas}} \bullet \sqrt{\sigma_n^2 + \sigma_n^2 \bullet \delta_C^2 + (I_{\text{th}}^{\text{meas}} - f_{\text{sh}} \bullet I_{\text{bg}}^{\text{meas}})^2 \bullet \delta_C^2}. \quad (A7)$$

The error factor δ_C of the age-dependent conversion coefficient C_a, (Eq. 2) can be calculated as:

$$\delta_C = \sqrt{\delta_b^2 + \delta_G^2}, \quad (A8)$$

where δ_b is the relative error factor of the device's calibration using a bottle phantom, and δ_G is the relative error factor of the age-dependent factor G.

The goal of the calibration of a device using a bottle phantom is to determine its sensitivity, i.e. to find out the values $I_{\text{ref}}^{\text{meas}} - I_{\text{bg}}^{\text{meas}}$ caused by radioactivity Q_{ref} of a reference radiation source. Therefore, δ_b is specified as:

$$\delta_b = \sqrt{\delta_{\text{ref}}^2 + \left(\frac{\sigma_S}{I_{\text{ref}} - I_{\text{bg}}}\right)^2}, \quad (A9)$$

where δ_{ref} is the relative error factor of activity for the reference radioactive source, which is known from the technical documentation of the provider (Production Association "Isotope"); σ_S is the error factor in measuring the intensity of the reference source.

Because the process of calibration using a bottle phantom is the same as the process of measurement of radioactivity in the thyroid, the error factor σ_S can be calculated as:

$$\sigma_S = \sqrt{\hat{\sigma}_{\text{ref}}^2 + \hat{\sigma}_{\text{bg}}^2}, \quad (A10)$$

where $\hat{\sigma}_{\text{ref}}^2 = \frac{I_{\text{ref}}^{\text{meas}}}{t_{\text{ref}}} + \sigma_{\text{dev}}^2$ is the error variance of measuring the intensity of the reference source during the measurement time t_{ref}.

For devices with missing information about the calibration, δ_b was, based on expert judgement, estimated to be 30%. The value of the relative error factor δ_G for the SRP-68-01 device was estimated from empirical data. According to Kaidanovsky and Dolgirev (1997) this factor depends on thyroid mass and is in the range of 15–18%. Since the scintillation crystals of the gamma-spectrometers were located significantly farther from the thyroid than that for the SRP-68-01 device, the influence of measurement geometry was less and δ_G was estimated for spectrometers, again based on expert judgement, to be 5%. This error factor was mainly due to variations in thyroid volume and thyroid position.

Based on Likhtarov et al. (2013c), the following observation model of thyroid radioactivity with classical additive error was selected in the present study:

$$Q^{\text{meas}} = Q^{\text{tr}} + \sigma_Q^{\text{meas}} \bullet \gamma. \quad (A11)$$

Censoring

Measurements of ^{131}I thyroid activity were considered reliable if the probability to detect a net signal, which is the difference between thyroid signal and background signal, with the assumption that its true value equals zero, was not more than 25%. This is equivalent to the condition $I_{\text{th}}^{\text{tr}} - f_{\text{sh}} \bullet I_{\text{bg}}^{\text{tr}} \geq 0.68 \bullet \sigma_n$, where σ_n is defined by Eq. (A4), i.e., the critical limit of ^{131}I in the thyroid was accepted to be $0.68 \bullet \sigma_n$. The result of a measurement providing less than the critical limit was replaced by half of the critical limit. It was accepted that $I_{\text{th}}^{\text{tr}} - f_{\text{sh}} \bullet I_{\text{bg}}^{\text{tr}} = 0.34 \bullet \sigma_n$ under the condition $I_{\text{th}}^{\text{tr}} - f_{\text{sh}} \bullet I_{\text{bg}}^{\text{tr}} < 0.68 \bullet \sigma_n$.

Estimation of Berkson errors due to deviation from the proper measurement geometry

Results of direct thyroid measurements conducted in May–June 1986 were associated with Berkson uncertainties (Masiuk et al. 2013, 2017) arising from deviation of the

detector from the proper measurement geometry (where the lower edge of the detector is close to the lower point of the neck). Khrutchinsky et al. (2012) considered deviations from the standard measurement geometry for the SRP-68-01 device without collimator by 0–1 cm shifts in horizontal and vertical direction, and inclinations in horizontal and vertical planes; for such scenarios the relative error was estimated to vary from 0.24 to 0.51 depending on the age of the measured individual.

Following Khrutchinsky et al. (2012), a zero shift of the detector from the neck in horizontal direction was considered to be the proper measurement geometry. It was assumed that with 95% probability the shift of the detector in horizontal direction was in the range 0–2 cm from the neck. It was also assumed to have censored log-normal distributions of the shift with GSD = 2. The impact of a shift in vertical direction along the neck and its inclinations in horizontal and vertical planes were neglected. The uncertainty associated with detector deviation from the proper measurement geometry was considered to be unshared Berkson errors (Drozdovitch et al. 2015; Simon et al. 2015).

Let GF_j be the factor due to shift of the j-th device's detector in the horizontal direction away from the neck. For the sake of simplification the detector is calibrated in such way that GF_j is equal to unity for the proper measurement geometry, i.e., $GF_j\big|_{S=0} = 1$. Then GF_j can be approximated by a function which is quadratic in the shift S:

$$GF_j = a_j \bullet S^2 + b_j \bullet S + 1. \tag{A12}$$

Model of mixed classical and Berkson errors in thyroid measurements

Factors that are characterized by uncertainty of Berkson type are denoted as:

$$F_{ij} = GF_{ij} \bullet AF_j^{\text{meas}}, \tag{A13}$$

where GF_{ij} is the factor due to the shift of the j-th device's detector in the horizontal direction away from the neck of the i-th individual; AF_j^{meas} is the adjustment factor for the j-th device.

The resulting thyroid radioactivity for the i-th individual measured by j-th device can be represented as:

$$A_{ij}^{\text{meas}} = F_{ij}^{\text{meas}} \bullet Q_i^{\text{meas}}, \tag{A14}$$

where Q_i^{meas} is the measured ^{131}I thyroid activity associated with the classical additive error (Eq. (A11)).

The unknown true radioactivity A_i^{tr} is expressed as:

$$A_{ij}^{\text{tr}} = F_{ij}^{\text{tr}} \bullet Q_i^{\text{tr}}. \tag{A15}$$

The connection between F_{ij}^{tr} and F_{ij}^{meas} is determined by a Berkson multiplicative error:

$$F_{ij}^{\text{tr}} = F_{ij}^{\text{meas}} \cdot \delta_{F,ij}, \tag{A16}$$

where error $\delta_{F,ij}$ has a log-normal distribution $\log(\delta_{F,ij}) \sim N(-\frac{\sigma_{F,ij}^2}{2}, \sigma_{F,ij}^2)$ with an expectation value $E\delta_{F,ij} = 1$; F_{ij}^{meas} and $\delta_{F,ij}$ are stochastically independent, and $\sigma_{F,ij}^2$ is the variance of $\log(\delta_{F,ij})$. Values of F_{ij}^{meas} and $\sigma_{F,ij}^2$ can be obtained by the two-dimensional Monte Carlo procedure described in (Simon et al. 2015).

According to Eq. (A11), the measured ^{131}I activity in the thyroid, Q_i^{meas}, can be expressed as:

$$Q_i^{\text{meas}} = Q_i^{\text{tr}} + \sigma_{Q,i}^{\text{meas}} \bullet \gamma_i, \quad i = 1, ..., N, \tag{A17}$$

where $\gamma_1, ..., \gamma_N$ are the independent standard normal variables; $\sigma_{Q,i}^{\text{meas}}$ are the individual standard deviations of errors of direct thyroid measurements that were estimated according to Eq. (10). The values $\sigma_{Q,i}^{\text{meas}} \bullet \gamma_i$ and Q_i^{tr} are independent random variables.

Substituting Eqs. (A17)–(A15) and denoting $\overline{A}_{ij}^{\text{tr}} = F_{ij}^{\text{meas}} \bullet Q_i^{\text{tr}}$ results in:

$$\begin{aligned} A_{ij}^{\text{meas}} &= F_{ij}^{\text{meas}} \bullet Q_i^{\text{meas}} = F_{ij}^{\text{meas}} \bullet \left(Q_i^{\text{tr}} + \sigma_{Q,i}^{\text{meas}} \bullet \gamma_i \right) \\ &= F_{ij}^{\text{meas}} \cdot Q_i^{\text{tr}} + F_{ij}^{\text{meas}} \bullet \sigma_{Q,i}^{\text{meas}} \bullet \gamma_i. \end{aligned} \tag{A18}$$

Random variables $\{\delta_{F,ij}, i \geq 1\}$, $\{\gamma_i, i \geq 1\}$ and random vectors $\{(F_i^{\text{meas}}, Q_i^{\text{tr}}), i \geq 1\}$ are jointly independent, but F_i^{meas} and Q_i^{tr} can be correlated. If the notations $\sigma_{ij} = F_{ij}^{\text{meas}} \bullet \sigma_{Q,i}^{\text{meas}}$ and $\overline{A}_{ij}^{\text{tr}} = F_{ij}^{\text{meas}} \cdot Q_i^{\text{tr}}$ are introduced, Eqs. (A14) – (A18) become the following:

$$A_{ij}^{\text{meas}} = \overline{A}_{ij}^{\text{tr}} + \sigma_{ij} \bullet \gamma_i, \tag{A19}$$

$$A_{ij}^{\text{tr}} = \overline{A}_{ij}^{\text{tr}} \bullet \delta_{F,ij}. \tag{A20}$$

Equations (A19) and (A20) compose a dose observations model with a classical additive error and a Berkson multiplicative error (Masiuk et al. 2016, 2017).

Finally, the relationship between expectations of true radioactivity and radioactivity measured with Berkson error can be expressed as:

$$E\left(\overline{A}_{ij}^{\text{tr}} \big| A_{ij}^{\text{meas}} \right) = E\left(A_{ij}^{\text{tr}} \big| A_{ij}^{\text{meas}} \right). \tag{A21}$$

Compliance with ethical standards

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