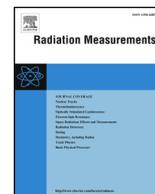




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## Assessing $^{131}\text{I}$ in thyroid by non-spectroscopic instruments - A European intercomparison exercise

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## ABSTRACT

One of the issues of the Open Project for the European Radiation Research Area (OPERRA) was human thyroid monitoring in case of a large scale nuclear accident. This issue was covered in task 5.4 as project “CaThyMARA” (Child and Adult Thyroid Monitoring After Reactor Accident), which included several aspects of thyroid monitoring, e.g. screening of facilities able to perform thyroid monitoring in the European countries, dose estimation, modelling of detector response, and two intercomparison exercises. The intercomparison described in this paper focused on thyroid monitoring by non-spectrometric instruments, including gamma cameras and other instruments that were considered available for measurements made by members of the public. A total of 12 facilities from 7 European countries have participated and 43 various measuring devices have been evaluated. The main conclusion of this intercomparison is that the ability to make assessments of  $^{131}\text{I}$  activity in the thyroid to the exposed population after an accidental release must, on the average, be considered as good among the European laboratories taking part in this study. This intercomparison also gave the participants the possibility to calibrate the measuring devices for thyroid measurements of children where this procedure was not available before. A comprehensive report of the intercomparison is given.

### 1. Introduction

In case of a nuclear accident, as Chernobyl and Fukushima, large amounts of radioiodine are released to the environment with the consequent risk of contamination of the population. The iodine is retained in the thyroid gland (target organ) during a few weeks after the intake.

Accidents involving releases of radioactive iodine can be significant sources of exposure of the thyroid gland and therefore deliver significant radiation doses to the exposed population. After the Chernobyl accident, many citizens received thyroid absorbed doses exceeding 1 Gy due to radioiodine intakes and more than 6000 thyroid cancers (mostly in children) were attributed to radioiodine. After the Fukushima

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accident about 98% of the effective doses received by emergency workers was attributable to radio-iodine (UNSCEAR, 2013). In such scenarios, in vivo measurements of  $^{131}\text{I}$  in thyroid are recommended to quickly identify the most contaminated people by gamma spectrometry at Whole Body Counters (WBC). However, this is unfeasible in case of large number of individuals potentially exposed. It is known that with proper calibration, many handheld instruments (e.g. dose rate meters and counters), stationary gamma spectrometers and gamma cameras can be used for this task for screening purposes (Nyander Poulsen et al., 2014; Scuffham et al., 2016). Lessons learned from the last accidents highlight the need for harmonizing procedures of calibration and measurement as well as establishing guidance for the triage in the early stage of the response to the accident.

One of the issues of the Open Project for the European Radiation Research Area (OPERRA) was various aspects of human thyroid monitoring in case of a large scale nuclear accident. OPERRA Task 5.4, the project “CaThyMARA” (Child and Adult Thyroid Monitoring After Reactor Accident) included several issues relating to thyroid monitoring, e.g. a survey of which facilities are able to perform thyroid monitoring in the European countries, methods for dose estimation, modelling of detector response, as well as two intercomparison exercises. This paper describes the intercomparison exercise that focused on thyroid monitoring by non-spectrometric instruments, including gamma cameras and other instruments that were considered available for measurements made by members of the public, based on a market survey (the details are given in SÚRO Report No (2017)). The use of data by members of the public has been used to a rather large extent in Japan following the Fukushima accident (Coletti et al., 2017; Brown et al., 2016).

The aim of the intercomparison was to evaluate the ability to provide accurate estimations of content of  $^{131}\text{I}$  in thyroid for children and adults and also to give laboratories the opportunity to calibrate or validate their non-spectrometric detector systems for such task.

## 2. Materials & methods

The thyroid phantom (child and adult) used in the intercomparison was prepared by SCK-CEN, Belgium (Lebacqz et al., 2017), and is shown in Fig. 1. The diameter and the depth of the six drilled holes (Table 1) were chosen to mimic the size and position of the thyroid lobes of a 5-y old child, a 10-y old child and an adult, respectively. Three sets of vials, with volumes corresponding to the thyroid lobe mass for the respective age, according to ICRP Publication 89 (ICRP, 2002), were used to simulate the thyroid gland (Fig. 2).

The iodine radioisotope  $^{131}\text{I}$  is a fission product with a physical half live of 8.02 days. Due to the short half-life, it is recommended in intercomparison exercises that a surrogate of  $^{131}\text{I}$  should be used to

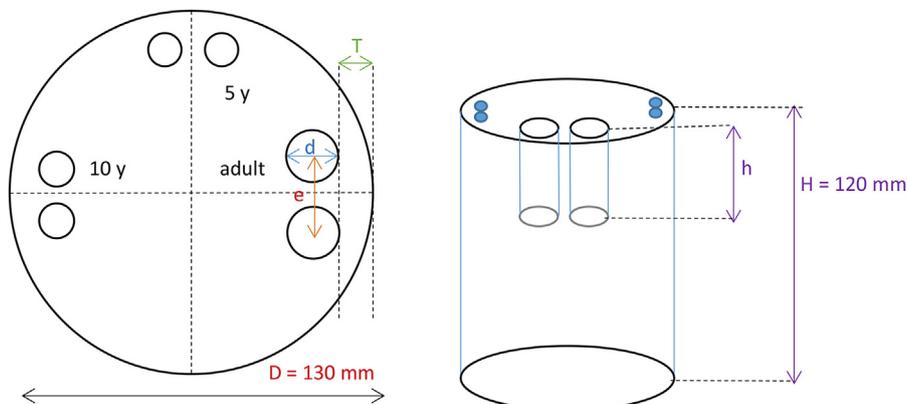


Fig. 1. Top and side view of the thyroid phantom showing the locations of the three pair of holes simulating the thyroid lobes for 5 and 10 y children, and adults. The dimensions of the phantom are given in Table 1.

Table 1  
Dimensions (mm) depicted in Figs. 1 and 2.

	5 y	10 y	Adult
e	24.0	26.0	34.0
d	17.7	17.7	24.1
T	10.0	12.0	15.0
h	40.0	60.0	70.0



Fig. 2. Vials with a solution of  $^{133}\text{Ba}$  and  $^{137}\text{Cs}$ , representing thyroid sizes for 5 y ( $2 \times 1.6$  ml), 10 y ( $2 \times 3.75$  ml) and adults ( $2 \times 9.5$  ml).

enable transportation between participating laboratories. Therefore, the vials were filled with a mock-iodine solution consisting of a mixture of  $^{133}\text{Ba}$  and  $^{137}\text{Cs}$ , prepared by IRSN, France, in a suitable proportion to simulate the gamma spectrum of  $^{131}\text{I}$ . The substitution of  $^{131}\text{I}$  by mock-iodine can be justified if the detector responds similarly to both sources and differences in emission probability for the various photon energies therefore have to be taken into account. For a spectroscopic device, it should be sufficient to compensate for the emission probability in the full energy peaks, but for a non-spectroscopic device, operating with energy windows, several photon energies may have to be matched. In order to compensate for the considerably higher X-ray photon yield of  $^{133}\text{Ba}$  compared to  $^{131}\text{I}$ , a filter of Ag is inserted in each of the holes in the phantom, thus covering the mock-iodine vial. To find a suitable distribution of activity between  $^{133}\text{Ba}$  and  $^{137}\text{Cs}$ , and taking account of the Ag-filter, the number of emitted photons in three energy intervals was chosen for comparison (Table 2). Ideally, the number of emitted photons per unit time from a mock-iodine source containing  $\alpha$  Bq of  $^{133}\text{Ba}$  and  $\beta$  Bq of  $^{137}\text{Cs}$ , and shielded by an Ag-filter, should be equal to the number of photons emitted per unit time from 1 Bq of  $^{131}\text{I}$  (without Ag-filter) in each of the three energy intervals, thus

$$\propto \cdot J_1 \cdot e^{-\mu_1 x} = I_1$$

**Table 2**

Energy groups,  $\Delta E$ , and corresponding sum of photon emission probabilities (denoted by  $I$  and  $J$ , for  $^{131}\text{I}$ ,  $^{133}\text{Ba}$  and  $^{137}\text{Cs}$ , respectively). Nuclear data by the Laboratoire National Henri Becquerel (LNHB, 2019).

	$^{131}\text{I}$		$^{133}\text{Ba}$		$^{137}\text{Cs}$	
	$\Delta E$ (keV)	$\Sigma I$	$\Delta E$ (keV)	$\Sigma J$	$\Delta E$ (keV)	$\Sigma J$
Group 1	80.2	0.026	79.6–80.99	0.36		
Group 2	284.3–364.5	0.88	276.4–383.8	0.96		
Group 3	636.9–722.9	0.091			661.6	0.85

**Table 3**

Activity of the manufactured sources and the activity ratio. Values shown are arithmetic means for the two duplicate vials used in the intercomparison. The relative uncertainty ( $k = 1$ ) for the activity is  $\pm 3\%$ .

	Activity (Bq) 1 Jan 2016		$\alpha/\beta$
	$^{133}\text{Ba}$	$^{137}\text{Cs}$	
5 y	2601.3	294.4	8.837
10 y	5911.1	668.9	8.837
Adult	14735.3	1667.5	8.837

**Table 4**

Mean mock-source activity, given by the sum of activities for  $^{133}\text{Ba}$  and  $^{137}\text{Cs}$ , and apparent  $^{131}\text{I}$  activity. The activities are given to the reference date 1 Jan 2016. The uncertainty in apparent activity is about 10% ( $k = 2$ ), taking into account the uncertainty in  $^{133}\text{Ba}$  and  $^{137}\text{Cs}$  activity (3% each) and different contribution from Compton continuum for the real source compared to the theoretical source (2%).

	Mock-source activity (Bq)	Apparent $^{131}\text{I}$ activity (Bq)
5 y	2896	2483
10 y	6580	5642
Adult	16403	14063

$$\propto J_2 \cdot e^{-\mu_2 x} = I_2$$

$$\beta \cdot J_3 \cdot e^{-\mu_3 x} = I_3$$

where  $x$  is the thickness of the Ag-filter and  $\mu_i$  is the linear attenuation coefficient for Ag at the mean photon energy in each of the three intervals.  $J_i$  and  $I_i$  are the sum of the photon emission probabilities for the mock-source and  $^{131}\text{I}$  source in interval  $i$ , respectively. By solving these three equations simultaneously, we get  $x = 0.1$  cm,  $\alpha = 1.04$  Bq and  $\beta = 0.12$  Bq, and the ideal ratio  $\alpha/\beta$  is thus 8.667. This means that a source of 1.16 Bq of mock-iodine, shielded by 0.1 cm Ag will give the same photon flux as a 1 Bq source of  $^{131}\text{I}$  (Isaksson et al., 2017). However, this ratio as well as the activities for the real sources differed slightly from the ideal source. In total, two phantoms with corresponding sets of vials were distributed among the participants and an example from one of these sets is given in Table 3.

When recalculating the photon flux from the real sources we found it difficult to keep the same ratio for both sets of sources. We therefore decided to use the energy interval 284.3–364.5 keV (Group 2 in Table 2) to find the apparent  $^{131}\text{I}$  activity. These activities are given in Table 4 and are based on the number of photons directly outside the Ag filter; no attempts have thus been made to compensate for different scattering properties in the phantom material.

The results were evaluated against criteria from the ‘‘Relative Bias’’ statistic parameter according to ANSI/HPS N13.30/ISO 12790-1 standards (American National Standards Institute, 2011; International Organization for Standardization, 2010):

$$B_{ri} = \frac{A_i - A_{ai}}{A_{ai}}$$

**Table 5**

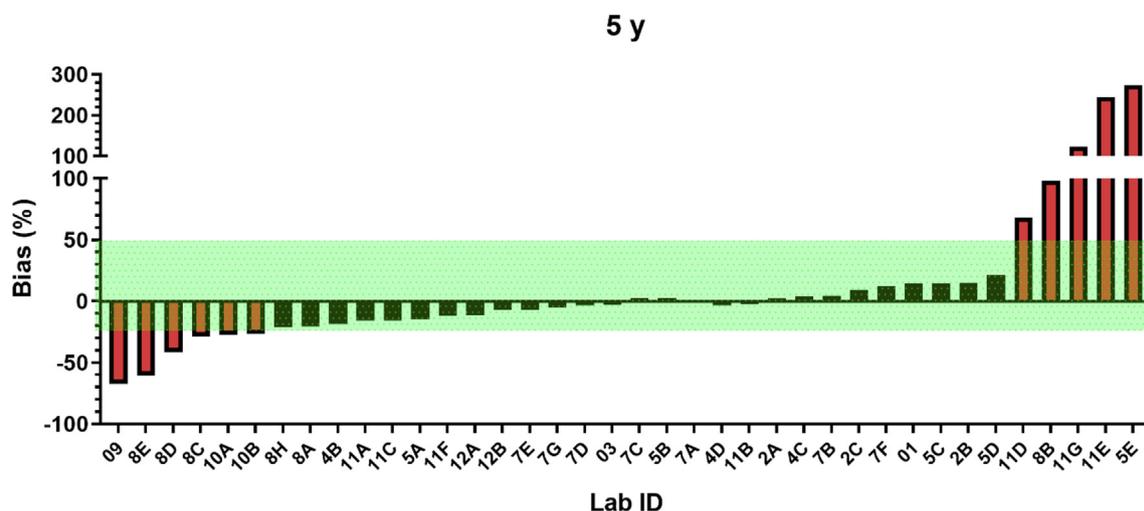
Facility code, type of instrument and calibration method for participants in the intercomparison exercise. NR indicates that no information was reported by the participant.

Facility Code	Measurement device	Calibration Source	Calibration phantom
1	Dosimeter	Iodine	Adult
2A	Count Rate Meter	No Iodine	Age specific
2B	Count Rate Meter	No Iodine	Age specific
2C	Gamma Camera	No Iodine	Age specific
3	Count Rate Meter	Iodine	Adult
4A	Dosimeter	Iodine	Age specific
4B	Dosimeter	Iodine	Age specific
4C	Count Rate Meter	Iodine	Age specific
4D	Count Rate Meter	Iodine	Age specific
5A	Dosimeter	Iodine	Age specific <sup>a</sup>
5B	Count Rate Meter	Iodine	Age specific <sup>a</sup>
5C	Dosimeter	Iodine	Age specific <sup>a</sup>
5D	Dosimeter	Iodine	Age specific <sup>c</sup>
5E	Count Rate Meter	Iodine	Age specific <sup>a</sup>
6A	Count Rate Meter	No Iodine	Adult
6B	Gamma Camera	No Iodine	Adult
7A	Dosimeter	Iodine	Age specific
7B	Dosimeter	Iodine	Age specific
7C	Dosimeter	Iodine	Age specific
7D	Count Rate Meter	Iodine	Age specific
7E	Dosimeter	Iodine	Age specific
7F	Dosimeter	Iodine	Age specific
7G	Dosimeter	NR	NR
8A	Gamma Camera	Iodine	NR
8B	Dosimeter	No Iodine	Adult
8C	Dosimeter	No Iodine	Adult
8D	Count Rate Meter	No Iodine	Age specific
8E	Count Rate Meter	No Iodine	Age specific
8F	Count Rate Meter	No Iodine	Age specific
8G	Count Rate Meter	No Iodine	Age specific
8H	Count Rate Meter	No Iodine	Age specific
9	Count Rate Meter	Iodine	Adult
10A	Count Rate Meter	Iodine	Age specific
10B	Count Rate Meter	Iodine	Age specific
11A	Dosimeter	Iodine	Age specific
11B	Dosimeter	Iodine	Age specific
11C	Dosimeter	Iodine	Age specific
11D	Dosimeter	Iodine	Age specific
11E	Dosimeter	Iodine	Age specific
11F	Count Rate Meter	Iodine	Age specific
11G	Dosimeter	Iodine	Age specific
12A	Count Rate Meter	Iodine	Age specific
12B	Count Rate Meter	Iodine	Age specific

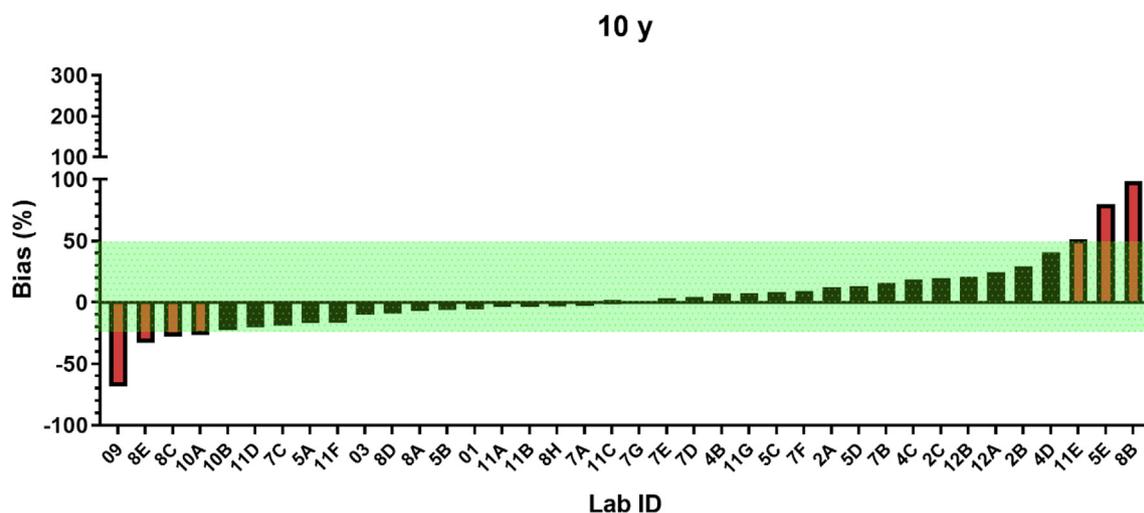
<sup>a</sup> No age categories were given, but instead different depth of organ, constant volume.

where  $B_{ri}$  is the relative bias statistic for the  $i$ th measurement in a category with respect to the known/correct value;  $A_i$  is the value of the  $i$ th measurement and  $A_{ai}$  is the known/correct value. For performance testing purposes, the acceptable relative bias shall be within  $-0.25$  to  $+0.50$ , when  $A_{ai}$  is at or above the MTL (Minimum Testing Level). A negative bias thus means that the reported result underestimates the true activity.

The enrolling of participants to the intercomparison was made by sending out an invitation to research institutes and hospitals. A form for the expression of interest was attached, requesting basic information such as availability of equipment and special calibration and measurements procedures for thyroid monitoring of children, and followed by a more comprehensive technical questionnaire and general instructions for those willing to participate. The technical questionnaire requested detailed information about the detection systems used, method and frequency of calibration, detection limit, etc. In total 12 participants from 7 countries took part in the intercomparison, and 43 instruments were used. The participants were requested to submit their results as activity of  $^{131}\text{I}$ .



**Fig. 3.** Relative bias (%) for the measurements of the 5-y phantom. The green area marks the acceptable interval  $-25\%$  to  $+50\%$ . Bars corresponding to values outside the acceptable interval are marked in red. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)



**Fig. 4.** Relative bias (%) for the measurements of the 10-y phantom. The green area marks the acceptable interval  $-25\%$  to  $+50\%$ . Bars corresponding to values outside the acceptable interval are marked in red. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

### 3. Results and discussion

Different devices were used, such as dose rate meters, count rate meters and gamma cameras. Some of them have been calibrated for a specific age, while others have only been calibrated for adults (Table 5).

Although the vials contained a mock-iodine solution, the participants were requested to submit their results as activity of  $^{131}\text{I}$ , i.e. the apparent  $^{131}\text{I}$  activity at a specific date, representing the activity of the mock-iodine in each vial. Previous to the discussion, it is important to highlight that some participants had calibrated their equipment with  $^{131}\text{I}$  and obtained good results in the intercomparison exercise. This can be considered as confirmation of the good design of the mock iodine source utilized. For the 5-y phantom, 38 results were reported since each participant could measure with different kinds of instruments. The mean was 12.97%, with 11 results outside the acceptable interval  $-25\%$  to  $+50\%$  (Fig. 3). However, the mean bias decreased to 9.04% if only instruments previously calibrated with  $^{131}\text{I}$  were considered. In this case, 6 results were outside the acceptable interval.

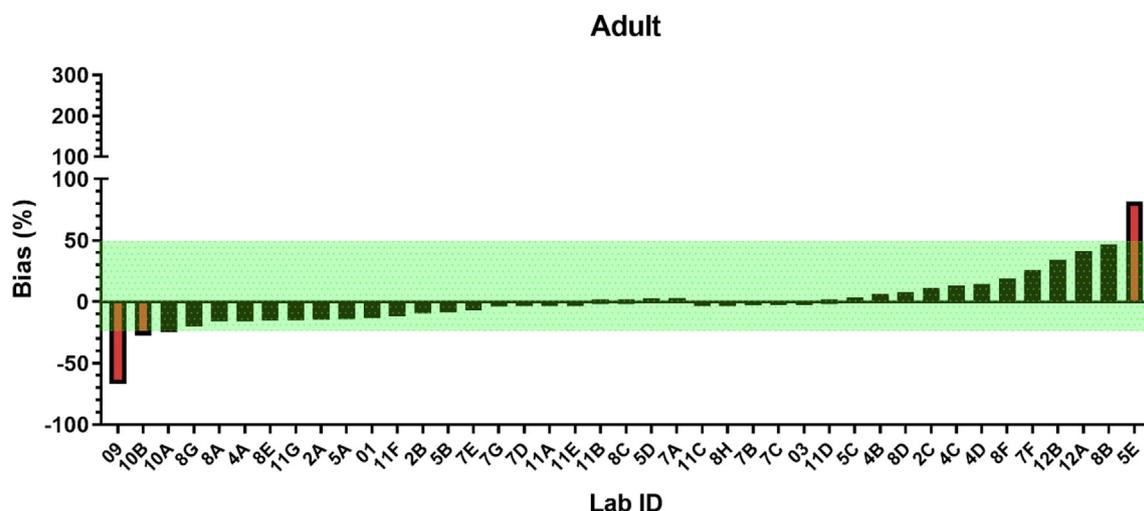
Also, 38 results were reported for the 10-y phantom, with a mean bias of 4.12% (Fig. 4). Of these, 7 of the results were outside the acceptable interval. Considering the subset of previously calibrated

instruments as above, the mean bias was reduced to 1.98%, with 4 results outside the acceptable interval.

A slightly larger number of results, 41, were reported for the adult phantom (Fig. 5). The mean bias was 0.2% with only 3 results outside the acceptance interval. For the previously calibrated instruments, the mean bias decreased to  $-0.06\%$ , still with 3 results found to be outside the acceptance interval.

Several types of instruments were used in the intercomparison and calibration factors for some of these are given in Table 6. These instruments are chosen due to good performance for all three phantom sizes (relative bias between  $-10\%$  and  $+10\%$ ) and reported calibration factors. The data from Table 6 may thus serve as examples of calibration factors for these types of instruments.

Two participants provided data based on measurements with gamma cameras. The relative bias for one of these labs were  $+9\%$ ,  $+20\%$  and  $+11\%$  for the 5 y, 10 y and adults phantom, respectively. This gamma camera was calibrated for each specific age, using different phantoms. The results from the other lab showed relative bias of  $-21\%$ ,  $-7\%$ ,  $-16\%$  for the three phantom sizes, respectively. Both of these participants used a General Electric Medical Systems INFINIA HAWKEYE-4.



**Fig. 5.** Relative bias (%) for the measurements of the adult phantom. The green area marks the acceptable interval  $-25\%$  to  $+50\%$ . Bars corresponding to values outside the acceptable interval are marked in red. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

**Table 6**

List of instruments and calibration factors, together with the distance from the neck during measurement and measurement time. The instruments given here are chosen due to reported results with relative bias between  $-10\%$  and  $+10\%$ . Also shown are results from one gamma camera where calibration factors were reported. Although some of the listed instruments may be used as spectrometers, none of these were used in this mode during this intercomparison. A similar intercomparison with spectroscopic devices were also performed during the CaThyMARA project. These results have been submitted for publication.

Instrument	Calibration factor ( $^{131}\text{I}$ )			Distance (cm)	Measurement time (s)
	5 y	10 y	Adult		
S.E.A. SCINTO Thyroid	41 kBq per $\mu\text{Sv}\cdot\text{h}^{-1}$	41 kBq per $\mu\text{Sv}\cdot\text{h}^{-1}$	46 kBq per $\mu\text{Sv}\cdot\text{h}^{-1}$	0	10
Berthold Technologies LB 124	92 Bq·cps $^{-1}$	101 Bq·cps $^{-1}$	111 Bq·cps $^{-1}$	5	60
XRF Corporation ICS-4000	699 Bq·cps $^{-1}$	694 Bq·cps $^{-1}$	952 Bq·cps $^{-1}$	10	300
Automess 6150 AD-b	53.9 kBq per $\mu\text{Sv}\cdot\text{h}^{-1}$	56.9 kBq per $\mu\text{Sv}\cdot\text{h}^{-1}$	75.1 kBq per $\mu\text{Sv}\cdot\text{h}^{-1}$	0	90
Canberra EasySpec	11.7 Bq·cps $^{-1}$	14.3 Bq·cps $^{-1}$	18.7 Bq·cps $^{-1}$	0	120
Canberra EasySpec	27.2 kBq per $\mu\text{Sv}\cdot\text{h}^{-1}$	46.8 kBq per $\mu\text{Sv}\cdot\text{h}^{-1}$	66.3 kBq per $\mu\text{Sv}\cdot\text{h}^{-1}$	0	120
Georadis s.r.o. RT-30	64 kBq per $\mu\text{Sv}\cdot\text{h}^{-1}$	64 kBq per $\mu\text{Sv}\cdot\text{h}^{-1}$	83 kBq per $\mu\text{Sv}\cdot\text{h}^{-1}$	0	*
General Electric Medical Systems INFINIA HAWKEYE-4	19.0 Bq·cps $^{-1}$	20.9 Bq·cps $^{-1}$	21.7 Bq·cps $^{-1}$	10	300

\*Three consecutive readings.

Two devices (count rate meter) were outside the acceptance interval in all measurements, 6 devices were outside the acceptance interval in 2 measurements, 4 devices were outside the acceptance interval in 1 measurement and 30 meet the acceptance criteria of ISO 28218.

In general, the lowest bias was obtained by laboratories using specific age calibration. In some cases, their instruments were calibrated for specific ages and their results were out of range, while others were calibrated for adults and their results were inside of the range. In case of activity in adult phantom there were fewer laboratories out of range, which means that analysing with a suitable efficiency is important to obtain more realistic activity results.

Some of the instrument types used in this intercomparison also appeared in a previous intercomparison (Nyander Poulsen et al., 2014). Among those listed in Table 6, this includes Berthold LB 124 ( $n = 4$ ) and Automess 6150 AD-b ( $n = 1$ ). The phantoms used in the study by Nyander Poulsen et al. (child, young and adult) were similar in size to those used in this study. The mean values of the calibration coefficients, in Bq·cps $^{-1}$ , for the Berthold LB 124 instruments when measuring close to the neck (0 cm) were 58, 72 and 105 for child, young and adult phantoms, respectively. When measuring at 10 cm from the neck, the corresponding values were 402, 431 and 575. The coefficient of variation varied between 22% and 37%. The calibration factors, in kBq per  $\mu\text{Sv}\cdot\text{h}^{-1}$ , for Automess 6150 AD-b at 0 cm distance from the neck were 65.5, 82.3 and 134 for child, young and adult phantoms, respectively.

The above given calibration factors from the study by Nyander

Poulsen et al. include a correction for mock-iodide since the participants did not report the measured activities as  $^{131}\text{I}$ . Correction factors are given by Nyander Poulsen et al. and for this comparison, data for Berthold LB 124 has been divided by a factor 2 and data for Automess 6150 AD-b has been multiplied by a factor 3. The calibration factors reported for Berthold LB 124 in this study are given at a measurement distance of 5 cm from the neck and thus not directly comparable to the data from Nyander Poulsen et al. However, the mean values of calibration coefficients for 0 cm and 10 cm shows good resemblance with data given by Nyander Poulsen et al. for the 5 y and 10 y phantoms. The agreement is rather good for the Automess 6150 AD-b regarding the smallest phantom, but the calibration factors deviates considerably for the 10 y and adult phantoms. A calibration factor for Automess 6150 AD-b has also been given in Rahola et al. (2006) as 45 kBq per  $\mu\text{Sv}\cdot\text{h}^{-1}$  for children and measurement in contact with the neck.

IAEA (2017) defines operational intervention levels (OIL) for a number of measured quantities, where a default OIL value indicates that a predetermined response action needs to be implemented. For thyroid monitoring, the default OIL value equals  $0.5\mu\text{Sv}\cdot\text{h}^{-1}$  above background ( $\text{OIL}_{8\gamma}$ ). In order to be useable under conditions with increased background, the calibration factor for the instrument used must not be too high in comparison to the factor of the “baseline instrument” used by the IAEA to relate the dose rate from the thyroid with the generic criterion for taking urgent action to reduce the risk of stochastic effects in the thyroid. No instrument in this intercomparison can be

used directly with this default OIL8<sub>γ</sub> value unless further considerations are made. The instrument Safecast bGeigie Nano, developed for environmental measurement by members of the public, was found to have a calibration factor of 26 kBq per  $\mu\text{Sv}\cdot\text{h}^{-1}$  for small children, which is lower than any of those given in Table 6. The relative bias for this instrument when measuring the 5-y phantom was  $-12\%$ .

#### 4. Conclusion

An intercomparison exercise has been organized for the adult and children thyroid measurement with non-spectroscopic devices. A total of 12 facilities from 7 European countries have participated and 43 various measuring devices have been evaluated. The main conclusion of this intercomparison is that the ability to make assessments of  $^{131}\text{I}$  activity in the thyroid to the exposed population after an accidental release must, on the average, be considered as good among the European laboratories taking part in this study. Results were successful for the measurements on the children and adult phantoms with most of devices, which means a satisfactory level of preparedness in Europe. This intercomparison also gave the participants the possibility to calibrate the measuring devices for thyroid measurements of children where this procedure was not available before. The presented calibration factors for some non-spectrometric instruments may serve as general indication for labs having the same type of instrument.

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