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CALIBRATION STANDARD FOR USE IN GAMMA SPECTROMETRY AND LUMINESCENCE DATING

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Key words: GAMMA SPECTROMETRY, LUMINESCENCE DATING **Abstract:** In our TL dating laboratory the dose rates are determined from high-resolution gamma spectrometry measurements. A Canberra spectrometer with HPGe detector and Marinelli geometry (0.5 l capacity) is used. Spectral analysis is based on the Sampo90 computer program. The users have to supply the program with the detector efficiency as a function of gamma ray energy. Long-term changes in sensitivity of gamma-ray detectors lead to a drift in detector efficiency over a period of time. A uranium pitchblende source is investigated here for use in the system calibration. This calibrated source is a tempting alternative, especially for TL dating procedure, because of its long half-life and wide range of gamma-ray energies. Moreover, for a standard geometry (Marinelli) and matrix (quartz), the self-absorption and coincidence corrections for the calibrated and measured samples are comparable. The numerical deconvolution of spectral multiplets from the uranium pitchblende source with strongly overlapping peaks is verified experimentally. Several multiplets are used in the calibration procedure.

1. INTRODUCTION

Most of natural radioactive elements are members of one of three radioactive series – uranium, actinium and thorium and from nonseries nuclides, mainly ⁴⁰K and ⁸⁷Rb. These natural sources of alpha, beta and gamma radiation represent a continuous exposure of minerals. Natural luminescence (thermally and optically stimulated) is mainly the result of the exposure to radiation from uranium and thorium series and from potassium decay in roughly equal proportions. In our dating laboratory the dose rates are determined from high-resolution gamma spectrometry measurements (Oczkowski and Przegiętka, 1998). A Canberra spectrometer with HPGe detector and Marinelli geometry is used. Spectral analysis is based on the Sampo90 computer program and it is focused on the radionuclide and activity determination.

In general, there are two approaches to calibration for natural radioactivity analyses. The best one is direct calibration where the standards are uranium and thorium series emitters, and ⁴⁰K. If one had such a calibration source he would not need to use the absolute efficiency curve. In this case the certified activity and the number of counts in the peak replaces the absolute efficiency for each peak for activity calculation. The main uncertainties arise from the certification of the standard. The next approach to calibration problem is to measure an absolute efficiency curve over the whole energy region. For efficiency calibration one can use any source with known nuclide activity and gamma emission probability. In this case not only the accuracy of the activity standardisation but also the uncertainties arising from the emission probability become important. In order to calculate activities of the nuclide, the user have to supply the Sampo90 computer program with detector efficiency as a function of the gamma ray energy (efficiency curve).

To determine the energy dependence of the detector efficiency a set of several primary gamma-ray sources (Debertin and Helmer, 1988) is needed to cover the energy range of interest. If a moderate precision is sufficient (about 5-10%), efficiency calibration with a mixed-radionuclide or multienergy source and a simple interpolation procedure may yield an adequate accuracy. However, a major disadvantage of multinuclide commercial standards is that some of nuclides are short-lived, so that such a source is only useful over a period of a few months. Moreover, below 100 keV there is no primary calibration radionuclide. Since particularly in the low energy region the major and minor element composition of the standard matrix and even grain size should be similar to the samples being dated, it is difficult to meet these requirements with commercially available sources. In general it is doubtful whether an ideal standard for TL dating exists (Aitken, 1985).

Several authors have suggested the applicability of uranium pitchblende ore or ²²⁶Ra in equilibrium with daughters as a calibration source for the efficiency measurements (Waldort and Doust, 1968; Wallace and Coote, 1969; Zobel et al., 1977 and Momeni, 1982), in particular because of their numerous, intense gamma ray energies and long half-life (1602 years for ²²⁶Ra). Since the peaks range from the X-ray region to a few MeV, calibration curves can be easily obtained. Despite the fact that this material provides a cheap and rapid method for the calibration of germanium spectrometers, in absolute gamma spectrometry, its use was usually limited to the channelenergy calibration, mainly because of the experimental effort required to correct coincidence summing effects (Dolev et al., 1969; Debertin and Schötzig, 1979; Arnold and Shima, 2000; De Felice et al., 2000 and Piton et al., 2000). However, secondary phenomena, such as cascade transition and self-absorption effects are the main reasons for using a radioactive ore diluted with typical sand matrix (quartz) for spectrometric dosimetry of natural samples.

Fig.1A (CLOR) shows the spectrum of uranium ore diluted in quartz sand measured by our gamma spectrometer. This source (here called CLOR source) has previously been used as a reference standard for calibration of the uranium window (1.764 MeV, ²¹⁴Bi) of the old fourchannel gamma spectrometer (Azar Polon). It was calibrated by the Central Laboratory for Radiological Protection (Biernacka, 1999) and the specific activities measured by CLOR are quoted here (see **Table 3**). For comparison, **Fig.1A** (Nat) shows the spectrum of a natural quartz sample. The spectra measured using this uranium pitchblende ore and any natural sample are similar.

The CLOR source for which the specific activities of basic nuclides are known is used as the reference source. When the geometry and matrix are identical, so that there is only a difference in the activity of the two materials, the spectrum permits one to perform direct efficiency calibration. With regard to the problem of coincidence summing, no corrections have to be applied if a natural sample is measured relative to a standard of the same radionuclide, in the same geometry. However, one of the characteristic features of Sampo90 (and also other programming tools in this field) is the use of efficiency calibration curve for nuclide activity calculation.

2. EXPERIMENTAL SET-UP AND BASIC PROCEDURES

The gamma spectrometer Canberra System 100 used in our laboratory consists of a HPGe detector XtRa -GX1520 with 0.5 mm Al window. The resolution and relative efficiency of the detector for 1332 keV (⁶⁰Co) are 1.82 keV and 19.4 %, respectively. The detector and preamplifier are placed inside a low-background lead shield (model 747) and cooled by liquid nitrogen from vertical dipstick cryostat (7500SL). The integrated signal processor (model 1510) consists of a pulse height analysis system to transform pulses, which are collected and stored by a computer-based MCA. Standard Marinelli beakers (0.5 1) are used as sample containers. The Signal Processor contains high-resolution spectroscopy amplifier with a pile-up rejector and a live-time corrector, which allows the spectrum analysis nearly independent of a system count rate. In our measurements the input pulse shaping is set to 4 μ s. The dead time of a Wilkinson type AD converter with a clock rate of 100 MHz equals a fixed time of 1.5 μ s (for peak detection) plus 0.01 μ s multiplied by the channel number. Data are stored in 4096 sequential channels. Automatic correction for the dead time is obtained by collecting data for a given live time. In general, the gross count rates for our samples are low, therefore the random summing corrections are neglected.

Operating parameters of the system are governed and controlled by the computer program – Canberra System MCA 100. The information has been established from calibration measurements performed for Marinelli geometry and standard source: the relationship between gamma energy and channel, the absolute efficiency calibration and the dependence of peak shape parameters on gamma energy. Additionally, the spectrum of laboratory background activity has been established from prolonged measurements. The spectrum analysis is performed in the range up to about 2 MeV.

In the routine analysis of a sample spectrum the fundamental algorithms of Sampo90 (i.e. peak search and fitting, multiplet deconvolution, and background subtraction) are supported by the calibration data (Routti and Prussin, 1969; Koskelo et al., 1981; Aarnio et al., 1988 and Sampo90, 1993). The resulting table of calculated peak areas allows us to perform nuclide identification and activity analysis, by comparison with the information located in a nuclide library file. For TL dating purposes two nuclide libraries were edited. These libraries contain data concerning natural nuclides (NAT.ilf) and subgroups of nuclides (MUR.ilf) emitting gamma radiation (Oczkowski, to be published). The most important section of the final report is the quantitative nuclide identification list and the specific activity of isotopes (in Bq/kg) that is the base for the annual dose rate calculations (Oczkowski and Przegiętka, 1998, Oczkowski et al., 2000).

3. CALIBRATION SOURCE FOR USE IN TL DATING

The term absolute efficiency, $\boldsymbol{\epsilon},$ will be used here to mean the following ratio

= N / (A k t)	3.1)

where N is the full energy peak net count corresponding to the gamma photons with energy E and gamma emission probability k, A is the activity of the source and t – the counting time.

As it was mentioned, in routine analysis performed by Sampo90 three distinctive tables of calibration parameters are used: the peak energy versus channel relationships, the dependence of absolute efficiency on peak energy (efficiency curve) and the table which contains peak shape parameters. The calibration library contains the energy and intensity data used for the energy-channel and efficiency calibrations. In principle, an automatic calibration procedure for establishing the efficiency curve is possible. Each reference peak can be associated with the closest

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L	ine No.		Nuclide	Energy	[keV]	γ Eff	Acti	vity
Е	D	S	/Multiplet	Line	Unc.	[%]	[Bq]	Unc. [%]
1	1	1	ma	12.900	0.2	100.00	196.0	15.0
2	2		mb	15.200	0.2	100.00	30.0	25.0
3	3	2	mc	16.200	0.2	100.00	100.0	10.0
4	4	3	md	19.500	0.2	100.00	13.0	30.0
5	5		Th-231Ac	25.642	0.1	14.60	37.7	10.0
6	6	4	Pa-231Ac	27.396	0.1	9.33	37.7	10.0
7	0	5	Pb-2100	46.520	0.1	4.05	574.0	2.0
ø	8	07	I II-22/AC	50.144 53.172	0.1	8.53	3/./	10.0
0	10	/ Q	PD-214U	55.1/2	0.1		055.0 813.0	2.4
10	10	0	Ph-214U	03.200 74 814	0.1	6.52	633.0	3.4 2.4
10	12	,	Pb-214U	77.107	0.1	11.00	633.0	2.4
12	13		Bi-214U	79.290	0.1	0.97	574.0	2.0
13			Ra-223Ac	81.067	0.1	15.24	37.7	10.0
14	14		Pb-214U	87.190	0.1	3.88	633.0	2.4
	15		mE	90.100	0.2	100.00	11.0	10.0
15	16		Ra-223Ac	94.677	0.1	8.90	37.7	10.0
16	17	10	Th-234U	112.800	0.1	0.24	813.0	3.4
17	18	11	mI	143.800	0.2	100.00	5.2	11.5
18	19	12	U-235Ac	163.379	0.1	4.74	37.7	8.2
10	20	13	mK	185.900	0.2	100.00	42.9	8.4
19	21	14	U-235AC Db 214U	205.334	0.1	4.74	3/./	8.2
	22	15	Pb-214U	241.920	0.1	0.55	633.0	2.4
	23	16	Pb-214U	238.940	0.1	19 24	633.0	2.4
20	25	17	Pb-214U	351 870	0.1	37 10	633.0	2.4
20	26	1/	Bi-214U	386.834	0.1	0.36	574.0	2.4
22	27		Bi-214U	388.950	0.1	0.41	574.0	2.0
23	28	18	Bi-214U	454.832	0.1	0.32	574.0	2.0
	29	19	Pb-214U	480.313	0.1	0.34	633.0	2.4
24		20	Pb-214U	487.130	0.1	0.44	633.0	2.4
25	30	21	Pb-214U	580.060	0.1	0.36	633.0	2.4
26	31	22	Bi-214U	609.311	0.1	46.10	574.0	2.0
27	32	23	Bi-214U	665.442	0.1	1.57	574.0	2.0
28	33	24	Bi-214U	703.064	0.1	0.47	574.0	2.0
29	34	25	BI-214U	719.856	0.1		574.0	2.0
30	35	26	DI-214U mP	785.000	0.1	4.00	5/4.0	2.0
31	36	20 27		785.900 806 155	0.2	1 23	0.7 574 0	3.2 2.0
32	37	28	Pb-214U	838 999	0.1	0.59	633.0	2.0
33	57	29	Ac-228Th	911.160	0.1	29.30	2.9	13.6
34	38		Bi-214U	934.039	0.1	3.17	574.0	2.0
35	39	30	Pa-234mU	1001.003	0.1	0.65	812.0	3.4
36	40	31	Bi-214U	1120.273	0.1	15.03	574.0	2.0
		32	Bi-214U	1133.650	0.1	0.26	574.0	2.0
37	41	33	Bi-214U	1155.183	0.1	1.69	574.0	2.0
38		34	Bi-214U	1207.674	0.1	0.46	574.0	2.0
39	42	35	Bi-214U	1238.107	0.1	5.92	574.0	2.0
40	43	36	Bi-214U	1280.952	0.1	1.48	574.0	2.0
41	44	37	Bi-214U	1377.659	0.1	4.03	574.0	2.0
40		38	BI-214U B; 214U	1385.295	0.1	0.78	574.0	2.0
42	15	30	DI-214U Bi-21/II	1401.480 1500 222	0.1	2 20	5/4.U 574.0	2.0
43 44	43	39	Bi-214U Bi-214U	1509.225	0.1		574.U 574.D	2.0
	46	40	Bi-214U Bi-214U	1583 220	0.1	0.41	574.0	2.0
45	47	70	Bi-214U	1599.300	0.1	0.72	574.0	2.0
46		41	Bi-214U	1661.258	0.1	1.15	574.0	2.0
47		42	Bi-214U	1729.580	0.1	3.06	574.0	2.0
48	48	43	Bi-214U	1764.490	0.1	15.93	574.0	2.0
49	49	44	Bi-214U	1847.413	0.1	2.13	574.0	2.0
		45	Bi-214U	1873.112	0.1	0.23	574.0	2.0
50	50	46	Bi-214 U	2118 533	01	1 21	574 0	2.0

Table 1. Calibration library for CLOR source.

library line within a given tolerance. In particular, for the absolute detector efficiency ε the calibration library must contain nuclide activity; fraction of decays yielding gamma rays corresponding to each reference peak and relative uncertainty of the activity.

A Marinelli beaker (0.51) with uranium ore uniformly distributed in quartz matrix (m = 0.77 kg) was adapted in our laboratory as a calibration standard. The Central Laboratory for Radiological Protection determined the specific activity of several basic nuclides (CLOR, Warsaw; Biernacka, 1999). The results reported here refer to calibration experiments with this source. **Table 1** contains the list of gamma ray energy (keV), the fraction of gamma



Fig 1. A) Pulse-height spectrum measured for 0.5I Marinelli beaker by our gamma spectrometer. CLOR (solid line): from uranium pitchblende ore diluted in quartz sand (specific activities for selected nuclide are given in Table 3, column 2). Vertical lines indicate the localization of peaks used for efficiency calibration. (see Table I). Nat (dashed line): from the natural sample (aeolian quartz from Kępa Kujawska dune).
B) Variation of absolute efficiency with photon energy (keV). Eff [%] (solid line): calculated by a least squares fit of a polynomial function (3.2) to the efficiency calibration, which illustrates the effectiveness of photon energy absorption.

ray emission per decay (γ Eff, %) and the number of disintegrations per second for a particular peak and nuclide (activity in Bq). The data concerning energy and gamma emission probability of natural isotopes were taken from the Table of Radioactive Isotopes (Browne and Firestone, 1986). The first three columns show the peak numbers used for energy (E), detector efficiency (D) and shape calibration (S). The recommended number of points depends on the accuracy required. Usually one point for every hundred keV is needed. However, there should be more points in the low energy part of the spectrum and near the maximum of the efficiency curve. The size of each calibration list (E, D and S) is limited to 50 rows. In this table multiplets are also listed. They are denoted as a, b, c and d for the lowest energy region and E, I, K and R for the remainder.

Table 2 shows the results of numerical and experimental analysis performed for overlapping peaks observed from the CLOR sample (Oczkowski, 2001). These multiplets were selected to cover gaps existing for single peaks in the low energy region of our calibration. The calculated gamma intensities (Calc Gps) are obtained from numerical simulation of the partial intensities corresponding to the interfering nuclides. This calculation is based on the nuclide activity report provided by the CLOR laboratory (see Table 3). In the case of overlapping peaks, the estimated intensity (Calc Gps from Table 2 quoted also in Table 1 as Activity in Bq) corresponds to 100 % efficiency of gamma emission (γ Eff in **Table 1**). The multiplet deconvolution is based on the data concerning overlapping peaks. Since this procedure is reliant on the accuracy of several parameters (set of nuclides, nuclide activity, gamma emission probability), an arbitrary large error is assumed in the calibration library (Table 1) for the respective activity of multiplets. The experimental gamma intensity (Table 2: Experimental, gps) is a result of division of the peak area (cps) by the absolute detector efficiency (DE). The last column shows the peak usage value (in per cent) taken from the nuclide identification and activity analysis report.

To carry out the absolute efficiency calibration, gamma lines from the calibration library were associated with the lines from the peak fit table for the CLOR spectrum. This association is performed automatically by Sampo90. In order to obtain the most accurate intermediate values the least squares fit of a polynomial function to the efficiency calibration points is performed and uncertainties are estimated. In Sampo90, for gamma energy E the following formula is used

$$\ln \varepsilon = \sum_{i=1}^{n} a_i \left[\ln (E / E_0) \right]^{i-1} , \qquad (3.2)$$

where a_i represents the fitted parameters and E_0 is constant (1keV). The total uncertainty of the calculated detector efficiency includes relative uncertainties of the gamma peak area, the calibration source activity and also co-variances introduced in the curve fitting. The following data for a_i were obtained: $a_1 = -69.23 \pm 1.616$, $a_2 = 44.46 \pm 1.215$, $a_3 = -10.86 \pm 0.356$, $a_4 = 1.146 \pm 0.0442$, $a_5 = -0.0449 \pm 0.0020$. The resulting values for the

No	Table	Nuclide	Calc.	Experimental					
Ln	[keV]		Gps	Ch	[keV]	[cps]	DE[%]	[gps]	[%] Us
1		ma	195.6	19.7	12.9	0.01	0.008	173.0	103.1
	12.325	Th-227Ac	6.43						3.3
	12.636	Pa-231Ac	8.45						5.2
	12.705	Ra-228Th	0.13						0.0
	12.952	U-238U	24.88						0.0
	12.952	U-235Ac	8.30						4.5
	12.952	Ac-228Th	0.43						0.2
	13.066	Pb-210U	59.72						41.3
	13.088	Pb-214U	36.39						21.0
	13.099	Pb-212Th	0.17						0.1
	13.274	Th-234U	32.04						20.3
	13.274	Th-231Ac	18.48						7.2
2		mb	30.0	24.0	15.1	0.01	0.027	32.0	99.9
	15.218	Th-227Ac	7.96						22.4
	15.230	Th-228Th	0.13						0.0
	15.376	Pb-212Th	0.03						0.1
	15.439	Pb-214U	7.35						23.0
	15.537	Pb-210U	14.51						54.4
3		mc	100.3	26.0	16.1	0.03	0.044	74.0	95.2
	15.659	Pa-231Ac	9.62						15.2
	16.119	U-235Ac	5.88						8.1
	16.154	Ac-228Th	0.58						0.6
	16.161	U-238U	33.99						0.0
	16.180	Ra-228Th	0.02						0.0
	16.559	Th-231Ac	13.99						13.7
	16.572	Th-234U	36.26						57.7
4		md	12.7	32.0	19.3	0.01	0.134	8.0	148.9
	19.113	Ac-228Th	0.13						1.1
	19.118	U-235Ac	1.01						0.0
	19.811	Th-231Ac	2.80						24.4
	19.839	Th-234U	8.80						123.4
19		mE	11.0	167.6	89.9	0.35	4.407	7.9	121.2
	89.639	Bi-214U	1.98						24.9
	89.955	U-235Ac	1.27						0.0
	89.955	Th-231Ac	0.36						3.1
	89.955	Ac-228Th	0.10						0.8
	90.128	Pb-214U	7.28						94.7
	90.128	Pb-212Th	0.05						0.7
27		mI	5.2	271.4	143.9	0.18	3.698	4.9	104.3
	143.786	U-235Ac	3.96						78.6
	144.183	Ra-223Ac	1.23						25.8
30		mK	42.9	352.3	186.0	1.35	3.067	44.1	99.9
	185.739	U-235Ac	20.26						44.6
	186.110	Ra-226U	22.65	1 - 2					55.3
68		mR	8.7	1504.3	785.9	0.06	0.854	6.7	131.3
	785.827	Pb-214U	6.92						105.1
	786.420	Bi-214U	1.78						26.2

Table 2. Analysis of selected multiplets. NoLn - line number from the peak Table for CLOR source, DE[%] - detector efficiency.

efficiency curve and experimental points are shown in **Fig. 1B**. For an ideal calibration source and procedure, the efficiency curve should be a smooth function of energy for this spectral range (from 15 to 2100 keV). Since in particular nuclear states of ²¹⁴Pb-U and ²¹⁴Bi-U

(Hellwege, 1961) are deexcited by cascade transition related to calibration peaks, shown in **Table 1**, it is fortunate that despite the cascade corrections the uranium pitchblende source gives calibration points positioned quite close to the smoothed efficiency curve.

	CL	OR		NAT20		MUR8			
Nuclide									
		TT	.		TT		D (TT
	Act.	Unc.	Lines	Act.	Unc.	Sub	Parent	Act.	Unc.
	[Bq/kg]	[Bd/kg]	Id/Lib	[Bq/kg]	[Bq/kg]			[Bq/kg]	[Bq/kg]
U-238U	1056	36	2/2	570	185		U-238	953	9
Th-234U			6/6	789	13				
Pa-234mU			2/2	1100	12				
Th-230U			1/1	1500	292	ш	Th- 230U	1500	292
Ra-226U	895	16	2/2	940	31	IV	Ra-	944	31
	022	20	10/10	000	4	X 7	226U	==>	
Pb-214U	822	20	18/18	822	4	v	Rn-	752	2
D: 01 411	= 4 =	1.5	20/40	=20	-		2220		
BI-214U	745	15	39/40	738	2	X 7 X	DI	0.52	20
Pb-210U			3/3	857	28	VI	Рb- 210U	853	28
Ac-228Th	3.7	0.5	12/30	2.3	0.2	VII	Th- 232Th	-	-
Pb-212Th			8/9	3.7	0.5	VIII	Rn- 220Th	2.6	0.1
Tl-208Th			2/7	2.5	0.1				
U-235Ac	49	4.0	6/6	48.0	1	IX	U- 235Ac	48	1
Th-231Ac			7/7	30.6	4				
Pa-231Ac			7/8	52.8	2	X	Pa- 231Ac	46	1
Th-227Ac			11/12	45.2	1				
Ra-223Ac			9/12	46.9	1	XI	Ra- 223A c	47	0.5
Rn-219Ac			2/2	42.6	1	1			
Pb-211Ac			4/4	48.3	1	1			
K-40	1.2	1.1	1/1	2.6	1	XII	K-40	2.6	1

Table 3. Specific activities of nuclides and subgroups for CLOR source. In column CLOR specific activities certified by CLOR for m = 0.77 kg are quoted. Id - number of identified peaks Lib - number of peaks in the nuclide library.

In **Table 3** the specific activities (in Bq/kg) corresponding to our reference source (m = 0.77 kg) are shown. In the first part of the table (column 2 and 3) data calibrated by CLOR are quoted. The following two parts of the table (NAT20 and MUR8) show respective values of our measurements obtained after calibration. Taking into account a noticeable number of assumptions, concerning in particular the multiplet deconvolution and cascade corrections, the agreement between the reference and experimental values seem to be satisfying.

4. DISCUSSION

Since natural ageing and radiation damage processes modify the detector sensitivity with time, each laboratory should have its own calibration source, even for modern detectors and low level of radiation. In the dating laboratory this standard source should be related to the method used to measure the annual dose, in this case gamma spectrometry.

The use of uranium pitchblende in equilibrium with its decay products for the calibration of gamma spectrometers was suggested many years ago (Waldort and Doust, 1968; Wallace and Coote, 1969; Zobel *et al.*, 1977 and Momeni, 1982). Shortly afterwards, such a method of calibration was criticized for its coincidence summing effects (Dolev *et al.*, 1969); the presence of cascade transitions leads to the necessity of calculating coincidence corrections. Correction of the gamma-ray spectrum for coincidence summing effect requires significant numerical and experimental effort. In general, however, the calibration procedure should be performed in as similar a manner to that used routinely as possible. In the context of luminescence dating, this means that the calibration source should be very similar to the samples being dated.

In a special case of, say, a "comparative" efficiency calibration, when a detector is used with the same size and type source both for the measurement and for efficiency calibration, secondary effects, like coincidence summing and attenuation, need not be considered. Of course, there may be differences between the "comparative" and absolute determinations of the efficiency curve. But such a relative efficiency, as a tool in dating, significantly reduces the experimental effort required to correct spectra of natural samples for summing effects. Finally, the cascade summing correction for each gamma peak has to be taken into account twice, during the absolute efficiency calculation as the denominator and next as the multiplier for peak intensity re-calculation. Moreover, for natural radionuclides emitting photons at several energies, the reliability of the activity analysis is improved by calculating it as the weighted average of the values obtained from each line.

It seems to be a separate challenge to estimate the contribution to the activity uncertainty due to omission of cascade summing corrections. This task needs extensive programs and libraries (e.g. KORSUM, GESPECOR, ETNA) in order to evaluate coincidence corrections for natural radionuclides and a particular spectrometer. Moreover, it should be done in the context of the uncertainty of an annual dose.

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