

Activity measurements of a solution of ^{137}Cs
An international comparison on efficiency tracing by ^{134}Cs

A. Rytz

1. Introduction

At its 1977 meeting, Section II of Comité Consultatif pour les Etalons de Mesure des Rayonnements Ionisants (CCEMRI) decided that the object of the next full scale comparison to be organized by the Bureau International des Poids et Mesures (BIPM) should be ^{134}Cs . Simultaneous measurements of ^{137}Cs , using the former as efficiency tracer, were also proposed. Other tracers (^{82}Br , ^{60}Co , ...) could be considered as well. As it was felt that unexpected difficulties might arise, only a restricted number of participants should be involved in these tracer measurements. Eventually the results would help to decide whether or not a full scale comparison should be considered later.

Although the method of efficiency tracing has been described by several authors [1, 2, 3], no detailed recipe was available which could have served as a guide for setting up a useful reporting form. However, the fact that several participants used instead the same form as for the ^{134}Cs comparison showed that an incomplete form would have been better than none at all.

There were some doubts about the expediency of activity measurements, rather than γ -emission rate, in the case of ^{137}Cs , where the results depend on several nuclear parameters. However, as these are now sufficiently well known and affect each result equally, expressing the results in terms of activity seemed adequate. The participating laboratories and the names of the persons involved are listed in Table 1. Fig. 1 shows the decay schemes of the two cesium isotopes considered. The decay data were taken from the literature [4, 5, 6].

Table 1

List of participants

		Names of the persons who carried out the measurements
AECL	Atomic Energy of Canada Limited Chalk River, Canada	J. S. Merritt A. R. Rutledge L. V. Smith
AIEA	Agence Internationale de l'Energie Atomique Vienna, Austria	H. Houtermans
BCMNI	Bureau Central de Mesures Nucléaires Euratom, Geel, Belgique	A. Nylandsted Larsen E. Celen R. Vaninbrouckx G. Grosse W. Zehner
BIPM	Bureau International des Poids et Mesures Sèvres, France	C. Veyradier C. Colas J. W. Müller P. Bréonce
LMRI	Laboratoire de Métrologie des Rayonnements Ionisants, Saclay, France	J. Bouchard R. Vatin
NBS	National Bureau of Standards Washington, D. C., USA	A. T. Hirshfeld D. D. Hoppes
NPL	National Physical Laboratory Teddington, United Kingdom	S. Brown D. Smith M. J. Woods
NRC	National Research Council of Canada Ottawa, Canada	K. Munzenmayer G. C. Bowes A. P. Baerg
OMH	Országos Mérésügyi Hivatal Budapest, Hungary	A. Szörényi
PTB	Physikalisch-Technische Bundesanstalt Braunschweig, Federal Republic of Germany	K. F. Walz E. Funck

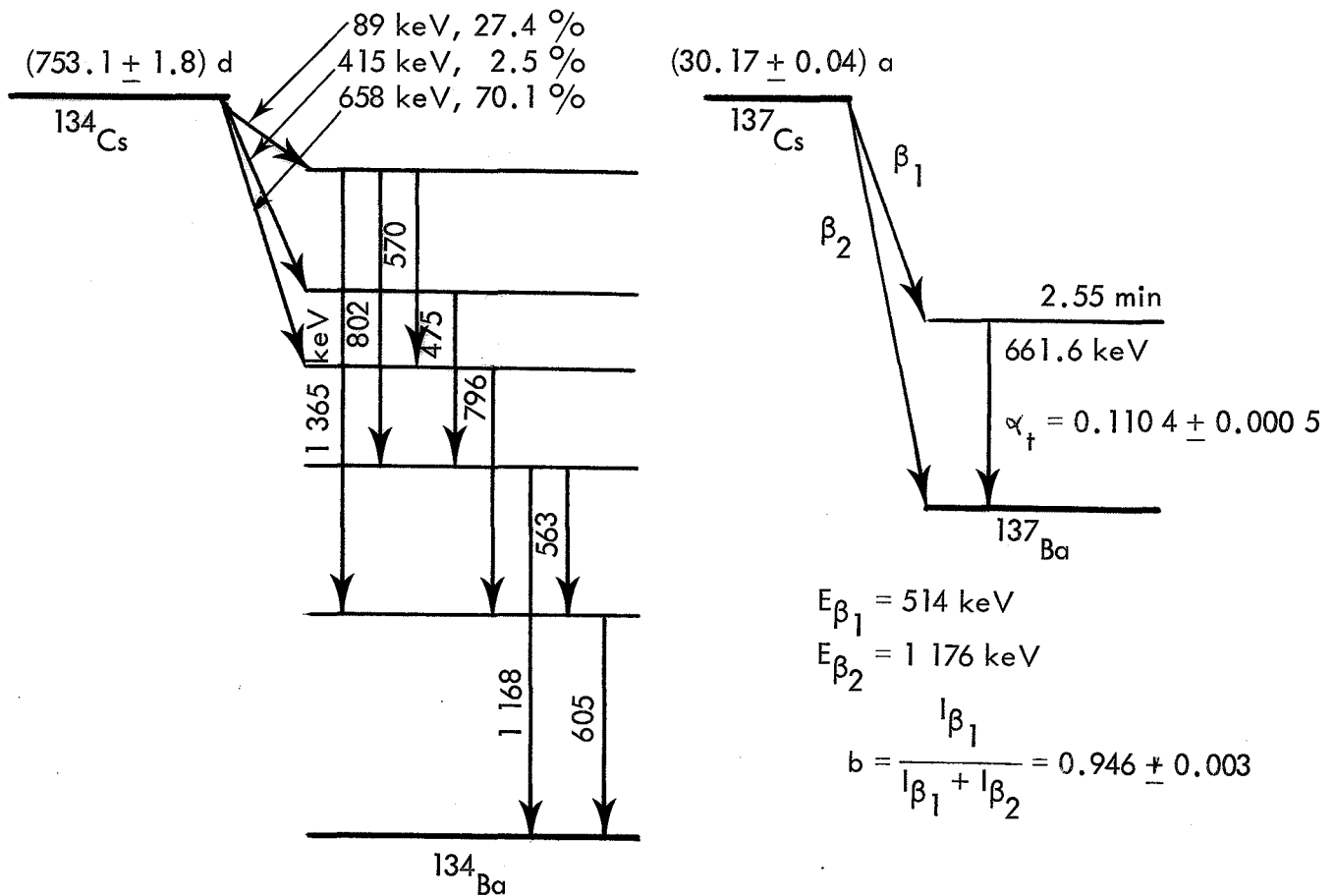


Figure 1 - Decay schemes of ^{134}Cs and ^{137}Cs (Data taken from [4,5,6])

2. Description of the solution and of source preparation

By circular letter of October 5, 1978 the participants were informed on the organization of the comparison and on the solution to be distributed. The NBS had generously offered to make available part of an old and very pure ^{137}Cs solution, to adjust its concentration and chemical composition and to do the bottling.

The activity concentration was about $800 \text{ Bq} \cdot \text{mg}^{-1}$ in an aqueous solution of HCl (0.2 mol per dm^3) with about $20 \mu\text{g}$ of CsCl per gram of solution. This composition was the same as that of the ^{134}Cs solution for the preceding full-scale comparison and which was to be used as efficiency tracer. No radioactive impurities had been found and the ^{134}Cs activity was estimated to be less than 10^{-4} that of ^{137}Cs .

Each participant, except AIEA, received 5 g of this solution in a flame-sealed glass ampoule. Two NBS ampoules were filled with 3.6 g only and sent to AIEA and BIPM for ionization chamber measurements.

The air parcels were despatched at the end of November, the reference date being 1978-12-01, 00 h UT.

Table 2
Source preparation

Laboratory	$^{134}\text{Cs}/^{137}\text{Cs}$ mixing ratio (mass)	Source backing Mass per cm^2 ($\mu\text{g} \cdot \text{cm}^{-2}$)	Numbers of sources prepared used		Mass of solution dispensed (mg)	Wetting agent, etc.
AECL	1/1	VYNS (Au-Pd) 28	15	13		Catanac, Ludox
AIEA	1/1 2/3	VYNS (Au + Pd) 35		12	10 - 32	Teepol + Ludox (4 sources) dried in open air. Dispensed on ion (7 sources) exch. resin, dried, redissolved in H_2O .
BCMNI	1/1	VYNS (Au) 50	11	9	10 - 28	Catanac
BIPM	2/1 1/1 1/2	VYNS (Au) 120	43	15	15 - 55	Ludox SM 10^{-4} one drop per source
LMRI	1/1 1/1	Cellulose (Au) 45	12 12	10	20	Insulin some drops of carrier
NBS	1/1 1/0.7	Collodion 25	10 10	9	16 - 50	Ludox or electrosprayed resin
NPL	0.77/1	VYNS (Au) 60	15	13	20 - 30	Catanac + 15 mg of $50 \mu\text{g}/\text{ml Al}^{3+}$
NRC	0.7/1	VYNS (Au + Pd) 40		10	31 - 34	Catanac
OMH	1/1 1/2	VYNS (Al) 30	40	36	12 - 29	Teepol + Ludox
PTB	1/0.9	VYNS (Au + Pd) 45	20	18 3 6	10 - 12	Ludox

AIEA decided to keep the ampoule unopened and to use instead an ampoule containing a similar solution which was compared with the NBS ampoule by means of a $4\pi\gamma$ ionization chamber.

Details of the sources prepared by the various participants are summarized in Table 2.

3. Counting equipment

All the participants made use of the efficiency-tracing method by ^{134}Cs , in combination with a coincidence counting technique. In addition, some other methods were applied, as shown in Table 3.

Table 3

Laboratory	Methods used
AECL	a) 4π (PC)- γ b) calibrated $4\pi\gamma$ ionization chamber c) relative γ counting
AIEA	4π (PC)- γ using an ampoule of a different stock solution, comparison with NBS ampoule by means of a pressurized ionization chamber
BCMNI	a) 4π (PC)- γ b) efficiency calibrated NaI(Tl) detector
BIPM	4π (PC)- γ
LMRI	4π (PC)- γ with one Ge(Li) detector in the γ channel
NBS	4π (PPC)- γ shared-dead-time anticoincidence system, with NaI(Tl) as γ detector.
NPL	4π (PC)- γ
NRC	4π (PPC)- γ anticoincidence counting with 2 NaI(Tl) or 2 Ge(Li) detectors [13]
OMH	4π (PC)- γ
PTB	a) 4π (PC)- γ } with one ordinary NaI(Tl) detector b) 4π (PPC ₁)- γ } c) 4π (PPC ₂)- γ with a large well-type NaI(Tl) detector, diameter = height = 152 mm, well diameter = 50 mm, well depth = 100 mm, resolution = 9.2 %.

PC = proportional counter used at atmospheric pressure

PPC = pressurized proportional counter.

Several participants had previously replaced their old scintillation detectors by new ones in order to improve the separation of the 662 keV peak from $^{137}\text{Ba}^m$ and the (796 + 802) keV peaks from the tracer.

In Table 4 typical count rates, γ -channel settings and resolutions of γ -detectors (for the 662 keV line) are given.

Table 4

Some data of the counting equipments

Laboratory	Typical count rates (s^{-1})			γ -channel setting (keV)	resolution of γ detector (%)
	N'_β	N'_γ	N'_c		
AECL	25 000	2 000	1 700	around (796+802) and 730-1 500	6.7
AIEA	3 000-10 000	80-240 100-320	65-190 90-275	755-870 755- ∞	6.5
BCMNI	9 500	630	540	720-850 720-1 600	6.4
BIPM	15 000	150	100	700-900	7.6
LMRI	16 000	40		796	0.23 ($^{60}\text{Co}, \text{Ge(Li)}$)
NBS	2 000-13 600	80-500	50-420 5-80 (AC)*	730-830	6.5
NPL	8 000	300-400	270-375	710-1 500 710-880	7.7
NRC	24 400	116 88	10-20 (NaI) (AC)* 7-14 (Ge(Li)) (AC)*	795-910 780-810	8.5 0.45
OMH	10 000-20 000	200-400	160-320	750-930 750-1 540	8.0
PTB	9 000	180 130 3 400	160 110 3 000	760-880 760-880 800-1 400	6.8 8.5 9.2

* For the AC results the entries under N'_c are the anti-coincident gamma count rates.

4. Efficiency of the β detector, ϵ_β

The conditions for the applicability of the efficiency-tracing technique, namely similar β spectra and intimate mixture of the two nuclides, are seldom fulfilled. Yet many examples are known where this method has been successfully used for standardizing pure β -emitters. As the present measurements involved two isotopes of an element with simple chemistry, intimate mixing could be taken as granted. However, the difference in the β spectra gave rise to some doubts. The comparison was undertaken in order to clarify the situation and to find out unsuspected sources of error.

As stated in [1] the total β -count rate of each source plotted versus the efficiency ϵ_β to the tracer may be extrapolated to $\epsilon_\beta = 1$ to give the sum of the disintegration rates of the two nuclides. The extension by Baerg et al. [2] offers a considerable variety of procedures to alter the efficiency. However, it must be emphasized that this quantity, although clearly defined theoretically, is not directly measurable. For the purpose of an extrapolation to 100 %, ϵ_β may be assimilated to the quotient N_c/N_γ , where N_γ is the γ -count rate corrected for background and dead time, N_c is the coincidence count rate corrected for background and accidental coincidences.

The γ -background rate depends on the resolution of the detector. NPL found that the 662 keV radiation did not contribute to the count rate in the γ channel, the lower limit of which was at 710 keV. AECL observed no change in background when a ^{137}Cs -only source was counted. BIPM made a correction of 0.54 s^{-1} per mg of ^{137}Cs solution. The other participants did not mention this influence. NRC found background rates at least 50 % higher than normal, due to pile-up effects. NBS reported a small contribution from the 662 keV peak in the Y_1 channel. A correction of 0.031 s^{-1} per mg of original ^{137}Cs solution was applied. On the grounds of daily checks of the count rate of 662 keV photons in the γ channel, PTB estimated this rate to be less than 4×10^{-4} .

Accidental coincidences were taken into account by means of Campion's formula [7] (OMH, BIPM, PTB low-activity sources) or by the formula derived from the work of Cox and Isham (NPL, PTB high-activity sources), see [8]. These corrections are small and of minor importance, and no such corrections are required for the anti-coincidence counting results [13].

The methods used for varying ϵ_β are indicated in Table 5.

5. Extrapolation procedures

The principle of the method of efficiency tracing, measuring ϵ_β for the tracer in a mixed source, variation of ϵ_β and extrapolation of the count rate to $\epsilon_\beta = 1$, was the same for each laboratory, but the experimental procedure varied considerably from participant to participant. The important details are presented in Table 6.

With few exceptions, the quantity ϵ_{β} designates the quotient N_c/N_{γ} . Corrections for photons from $^{137}\text{Ba}^m$ and for accidental coincidences were mentioned by some laboratories. It may be assumed that the others considered these corrections as negligible.

Table 5

Methods for varying the beta efficiency

Laboratory	Method used	Range of ϵ_{β} (%)
AECL	a) wetting agent b) cation exchange resin c) foil absorption d) carrier	93-76
AIEA	Sandwich and addition of aluminized mylar films (0.9 or 1.25 mg · cm ⁻²)	92-74
BCMNI	Foil absorption; VYNS film, AL foils	91-75
BIPM	Foil absorption: VYNS films, Al foils	91-69
LMRI	From 10 to 30 mg of carrier solution (100 g per l)	95-52
NBS	Threshold variation	94-77
NPL	VYNS foils	94-70
NRC	Threshold variation	92.5-82
OMH	Different amounts of carrier	92-79
PTB	Foil absorption: VYNS films (4 sources only)	I 93.5-72
	Threshold variation	II 89-67
	Threshold variation	III 92-65

Table 6

 $\epsilon_{\beta} \rightarrow 1$, efficiency extrapolation; signification of $y(x)$ and x

Laboratory	$y(x)$	x	Remarks
AECL	N_{β}	$1 - \epsilon_{\beta}$	N_{β}^{tot} was corrected for dead time, background and decay to mid-time of run, which typically lasted ≈ 10 h. Four runs with 2 γ -channel gates and 15 to 31 degrees of freedom. First order fits no less suitable than 2nd order. Final value is mean of 8 individual results.
AIEA	1. $\frac{N_{\beta} - m_{134} \epsilon_{\beta} A_{134}}{m_{137}}$	ϵ_{β}	4 values of ϵ_{β} for each source, 48 data points, 24 different plots (separately for each source and γ discrimination), linear fits.
	2. $y(1) \rightarrow y_0(1)$	$m_{137} \rightarrow 0$	2 different plots (for the two γ discriminations) linear fit; $m_{137} \rightarrow 0$; mean of the two results.
BCMNI	a) $\frac{N_{\beta} N_{\gamma}}{m N_c}$	$\frac{1 - \epsilon_{\beta}}{\epsilon_{\beta}}$	Linear fits with 67 and 72 degrees of freedom for the two γ -channel settings, respectively. Results of a) were rejected later.
	b) N_{β}	$1 - \epsilon_{\beta}$	
BIPM	$\frac{N_{\beta} - m_{134} \epsilon_{\beta} A_{134}}{m_{137}}$	$1 - \epsilon_{\beta}$	As $\epsilon_{\beta} = N_c/N_{\gamma}$ did not lead to reasonable results, an alternative and independent method for determining ϵ_{β} (to be published later) was applied. First order simultaneous adjustments [9] with sets of data points from 3 dilutions (mixing ratios see Table 2). Higher orders were less suitable.
LMRI	$\frac{N_{\beta} N_{\gamma}}{m N_c}$	$1 - \epsilon_{\beta}$	Second order fit with 13 data points.

Table 6 (cont'd)

Laboratory	y(x)	x	Remarks
NBS	$\frac{N_{\beta} - m_{134} A_{134} (1 - N_Y/N_Y)}{1 - N_Y/N_Y}$	$\frac{N_Y/N_Y}{1 - N_Y/N_Y}$	<p>From 8 to 18 data points and separate 2nd order fit for each source. Unweighted mean of intercepts decay corrected and divided by source mass. N_Y = anticoincidence rate.</p>
NPL	$\frac{N_{\beta} - m_{134} N_{134}}{m_{137}}$	$1 - \epsilon_{\beta}$	<p>Separate 2nd order fits for each source and for the two γ-channel settings. Final result is unweighted mean of the two individual means.</p> <p>$N_{\beta} = A_{134} \epsilon_{\beta} \left(1 + a_{11} \frac{1 - \epsilon_{\beta}}{\epsilon_{\beta}} \right)$ for the 710 - 880 keV gate,</p> <p>$N_{\beta} = A_{134} \epsilon_{\beta} \left[1 + a_{21} \frac{1 - \epsilon_{\beta}}{\epsilon_{\beta}} + a_{22} \frac{(1 - \epsilon_{\beta})^2}{\epsilon_{\beta}} \right]$ for the 710 - 1 550 keV gate.</p> <p>The a_{ij} are fitted parameters found in the separate standardization of ^{134}Cs which happened to be linear for the narrow gate:</p> <p>$N_{\beta}/\epsilon_{\beta} = A_{134} \left(1 + a_{11} \frac{1 - \epsilon_{\beta}}{\epsilon_{\beta}} \right)$ for ^{134}Cs, quadratic for the wider gate:</p> <p>$N_{\beta}/\epsilon_{\beta} = A_{134} \left[1 + a_{21} \frac{1 - \epsilon_{\beta}}{\epsilon_{\beta}} + a_{22} \frac{(1 - \epsilon_{\beta})^2}{\epsilon_{\beta}} \right]$ for ^{134}Cs.</p>
NRC	$\frac{N_{\beta} - m_{134} A_{134} (1 - N_Y/N_Y)}{m_{137}}$	$\frac{N_Y}{N_Y}$	<p>14 data points taken for each γ-detector type (one or two channels). Two-dimensional analysis, when 2 γ detectors added for one channel; 3-dim. analysis, when 2 separate γ channels. Linear fitting (order defined by χ^2 test). Weighted mean of 4 results.</p>

Table 6 (cont'd)

Laboratory	y(x)	x	Remarks
OMH	$\frac{N_{\beta} N_{\gamma} / N_c - m_{134} \epsilon_{\beta} A_{134}}{m_{137}}$	$\frac{1 - \epsilon_{\beta}}{\epsilon_{\beta}}$	Separate linear fits for two solutions and two γ -channel settings. Weighted mean of 4 results. About 18 data points for each fit.

(γ gates see Table 4) I 4 4th order fits, 37 data points, 4 sources
 II 5 " " " 154 " " 3 "
 III 1 " " fit 108 " " 6 "

The fitted polynomials were:

$$y = a_0 + a_1 x$$

$$y = a_0' + a_1' x + a_2' x^2$$

$$y = a_0'' + a_1'' x + a_3'' x^3$$

$$y = a_0''' + a_1''' x + a_4''' x^4$$

The fourth order was found to be the most suitable.

PTB	$\frac{N_{\beta}}{m}$	$1 - \epsilon_{\beta}$
-----	-----------------------	------------------------

Explanation of symbols

N_{β} = total count rate in the β channel
 N_{γ} = " " " " γ "
 N_c = " " " " c "
 N_Y = anticoincidence count rate

} corrected for dead time, background (and accidental coincidences)

A_{134} = activity concentration of the tracer solution (^{134}Cs)

m_{134} = mass of ^{134}Cs solution dispensed onto a mixed source

m_{137} = mass of ^{137}Cs solution dispensed onto a mixed source

m = $m_{134} + m_{137}$

ϵ_{β} = N_c / N_{γ} (with the necessary corrections).

6. Calculation of the ^{137}Cs -activity concentration

The information received being somewhat disparate and containing some more or less obvious errors, it is not clear if there are any significant differences in the formulae used by the various participants. For the sake of clarity, it was necessary to unify the presentation and the symbols. Decay corrections were considered sufficiently straightforward to be omitted. Table 7 presents expressions for $Z =$ numerator and $D =$ denominator of $A_{137} = \frac{Z}{D}$.

Table 7

Formulae used for calculating the activity concentration $A_{137} = \frac{Z}{D}$

Laboratory	Z	D
AECL BCMN LMRI PTB	$\frac{y(1) - m_{134} A_{134}}{m_{137}}$	$1 + \frac{b(\alpha + \epsilon_{\beta\gamma})}{1 + \alpha}$
AIEA	formula not indicated	
BIPM NRC OMH	$y(1)$	$1 + \frac{b(\alpha + \epsilon_{\beta\gamma})}{1 + \alpha}$
NBS	$\frac{y(1)}{m_{137}}$	$1 + \frac{b(\alpha + \epsilon_{\beta\gamma})}{1 + \alpha}$
NPL	$y(1)$	$1 + \frac{b}{1 + \alpha} (\alpha \epsilon_{ce} + \epsilon_{\beta\gamma})$

where

$y(1)$ is the value of $y(x)$ of the corresponding laboratory (see Table 6) for $\epsilon_{\beta} = 1$,

b = β branching ratio of ^{137}Cs ,

α = total internal conversion coefficient for the 662 keV radiation,

$\epsilon_{\beta\gamma}$ = γ efficiency of the β detector,

ϵ_{ce} = efficiency of the β detector to conversion electrons from $^{137}\text{Ba}^m$.

7. Results and uncertainties

The results obtained by the ten participants are summarized in Table 8 along with the values of $\epsilon_{\beta\gamma}$, the β -detector efficiency to 662 keV photons, and how they were determined. A graphical representation of the results is shown in Fig. 2.

Uncertainties are given at the 1σ level. Where no allowance for the uncertainty of the ^{134}Cs activity concentration was made, we added the value from the recent ^{134}Cs comparison (overall uncertainty of the corresponding laboratory). As may be seen from Table 7, the factor D is practically the same for all participants, since they used the same values for α and b . Therefore, the uncertainties in these constants must not be considered in assessing systematic errors from this comparison. The values recommended by the BIPM were $\alpha = 0.1104 \pm 0.0005$, $b = 0.946 \pm 0.003$ and represent weighted means as taken from Christmas and Cross [6]. The combined effect on the final result is

$$\frac{b\alpha}{1+\alpha} = 0.09406 \pm 0.00049.$$

It should further be noted that an uncertainty in $\epsilon_{\beta\gamma}$ of 0.008 is equivalent to the combined effect of the uncertainty in α and b as given above.

Mean values from international comparisons are often difficult to interpret and may be misleading. However, the request to "conserve" a representative result of the present comparison by means of the International Reference System for Activity Measurements of γ -ray emitters (SIR) made it necessary to take an average value. Since the weighted and unweighted means nearly coincided, an intermediate value and its (external) standard deviation were chosen:

$$A = (760.0 \pm 4.0) \text{ Bq} \cdot \text{mg}^{-1}.$$

A graphical comparison of all the SIR results is presented in Fig. 3.

Table 8

Results of the measurement

Laboratory	Method	^{137}Cs activity concentration 1978-12-01 ($\text{Bq}\cdot\text{mg}^{-1}$)	Uncertainties (1σ)				γ efficiency of the β detector	
			random		systematic		$\varepsilon_{\beta\gamma}$ (%)	how determined
			(Bq·mg ⁻¹)(%)		(Bq·mg ⁻¹)(%)			
AECL	4 π (PC)- γ	760.35	0.81	0.11	1.45	0.19	0.10 ± 0.03	estimated from [10]
	4 π γ (IC)	760.75	1.12	0.15	1.45	0.19		
	rel. γ count.	762.21	1.12	0.15	1.45	0.19		
AIEA	4 π (PC)- γ	765.2	0.7	0.1	3.8	0.5	-	neglected
BCMNI	4 π (PC)- γ	754.5	1.4	0.19	2.8	0.37	0.11	foil absorption
	calibr. NaI(Tl)	754.1	0.8	0.1	3.0	0.4	± 0.03	
BIPM	4 π (PC)- γ	764.2	1.0	0.13	4.8	0.63	0.3 ± 0.1	estimated from measurements with ^{54}Mn
LMRI	4 π (PC)- γ	758.7	1.2	0.16	2.0	0.26	0.11	after [11]
NBS	4 π (PPC)- γ (AC)	756.0	0.6	0.08	2.3	0.31	0.6 ± 0.1	interpolated between results from ^{51}Cr and ^{54}Mn
NPL	4 π (PC)- γ	757.17	0.45	0.06	+2.7 -0.9	+1.04 -0.35		after [12]
NRC	4 π (PPC)- γ (AC)	759.55	0.34	0.05	2.6	0.34	0.32 0.21	for NaI(Tl) in γ channel for Ge(Li) " (calculated or measured)
OMH	4 π (PC)- γ	766.65	0.63	0.08	3.1	0.41	0.10 ± 0.05	estimated [14]
	4 π (PC)- γ	757.6	0.3	0.04	1.6	0.21	0.11 ± 0.04	with sources of ^7Be and ^{54}Mn and interpolated
PTB	4 π (PPC ₁)- γ	760.1	0.5	0.07	2.7	0.35	0.25	
	4 π (PPC ₂)- γ (well)	757.2	0.5	0.07	2.7	0.35	± 0.05	

750 760 770
 ^{137}Cs - Activity concentration ($\text{Bq}\cdot\text{mg}^{-1}$; 1978-12-01)

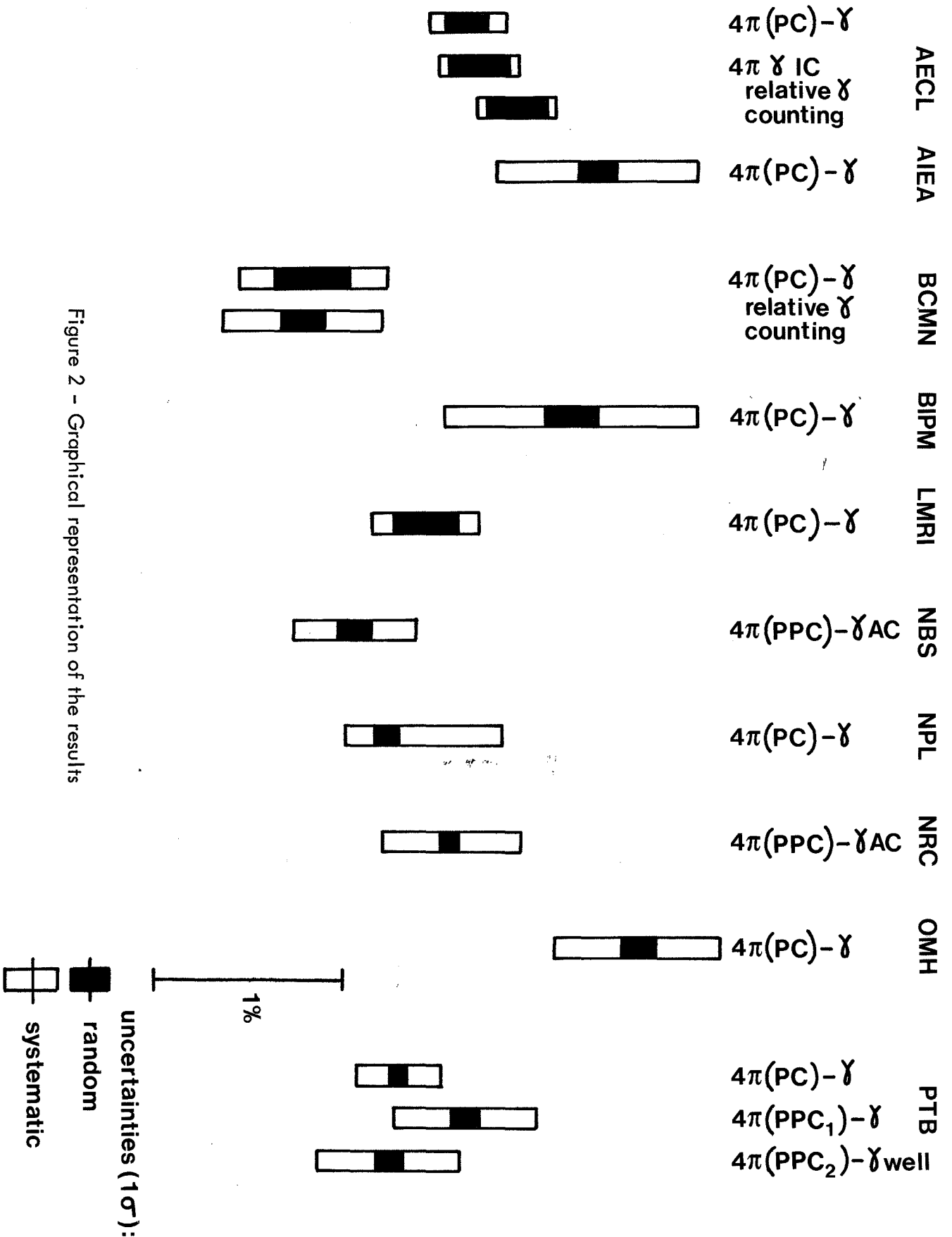


Figure 2 - Graphical representation of the results

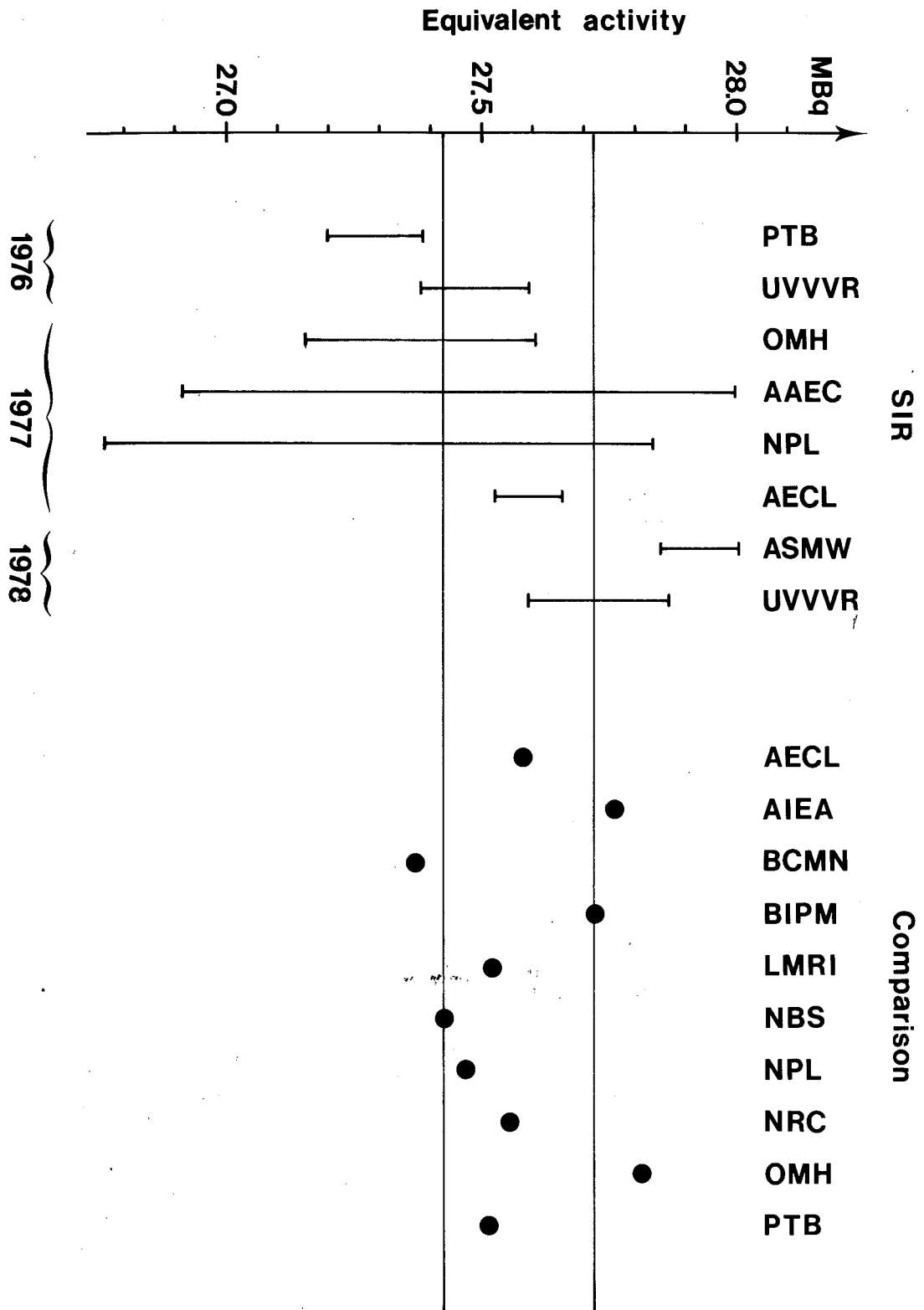


Figure 3 - The results of the international reference system (top) referred to the present comparison (bottom).

References

- [1] P.J. Campion, J.G.V. Taylor and J.S. Merritt, The efficiency tracing technique for eliminating self-absorption errors in $4\pi\beta$ -counting, *Int. J. Appl. Radiat. and Isotopes* 8, 8-19 (1960)
- [2] A.P. Baerg, S. Meghir and G.C. Bowes, Extension of the efficiency tracing method for the calibration of pure β -emitters, *ibid.* 15, 279-287 (1964)
- [3] J.S. Merritt and J.G.V. Taylor, Decay of Cesium-137 determined by absolute counting methods, *Anal. Chem.* 37, 351-354 (1965)
- [4] J.S. Merritt and F.H. Gibson, Standardization of ^{137}Cs by the $4\pi(\text{PC})-\gamma$ efficiency-tracing method with ^{134}Cs as tracer, *AECL-6203* (1979), 19 p.
- [5] R.L. Bunting, *Nuclear Data Sheets A = 137*, Academic Press, New York and London, Vol. 15, No. 3 (1975)
- [6] P. Christmas and P. Cross, The decay of ^{137}Cs determined by β -ray spectrometry, *Metrologia* 14, 157-166 (1978)
- [7] P.J. Campion, The standardization of radioisotopes by the beta-gamma coincidence method using high efficiency detectors, *Int. J. Appl. Radiat. and Isotopes* 4, 232 (1959)
- [8] D. Smith, Improved correction formulae for coincidence counting, *Nucl. Instr. and Meth.* 152, 505-519 (1978)
- [9] J.W. Müller, Ajustements simultanés, *CIPM, Procès-Verbaux des séances, 2^e série, tome 47, 68^e session* (1979), to be published
- [10] J.S. Merritt and J.G.V. Taylor, Response of 4π proportional counters to γ -rays, in "Standardization of Radionuclides", IAEA, Vienna, 147-152 (1967)
- [11] M. Lainé, J.-P. Pérolat, J. Bouchard et Y. Le Gallic, Les coïncidences $4\pi\beta-\gamma$ au laboratoire de métrologie de la radioactivité, *Rapport CEA-R-4131, Saclay* (1971), 55 p.
- [12] A. Williams and P.J. Campion, Measurement of the γ -sensitivity of a $4\pi\beta$ -counter, *Int. J. Appl. Radiat. and Isotopes* 14, 533-540 (1963)
- [13] A.P. Baerg, K. Munzenmayer and G.C. Bowes, Live-timed anti-coincidence counting with extending dead-time circuitry, *Metrologia* 12, 77-80 (1976)
- [14] A.P. Baerg, Pressurized proportional counters for coincidence measurements, *Nucl. Instr. and Meth.* 112, 95-99 (1973)