Report on a comparison of activity measurements of a solution of ^{133}Ba

by A. Rytz

Bureau International des Poids et Mesures, F-92310 Sèvres

This comparison was carried through by the Working Group for advising on future comparisons on behalf of Section II (Mesure des Radionucléides) of the Comité Consultatif pour les Etalons de Mesure des Rayonnements Ionisants.

Among a dozen possible candidates for such a minicomparison, ^{133}Ba was chosen for metrological and practical reasons. The original plan to use the six remaining ampoules of a solution from the 1979 comparison of γ-ray emission probability measurements (organized by LMRI on behalf of the International Committee for Radionuclide Metrology) had to be dropped due to insufficient reliability. Instead, LMRI made available a fresh solution of similar chemical composition, namely: $BaCl_2$ in an aqueous solution of hydrochloric acid (1 ml per dm^3) with a carrier concentration of 15 µg in 1 g of solution. Each participant (see Table 1) received in the first week of December 1980 a flame-sealed ampoule containing 1 ml of solution. Further ampoules were sent on request to BCMN and OMH in February 1981. In addition, BIPM received two ampoules, containing 3.6 g each, for the international reference system. No radionuclidic impurities had been observed, the detection limit being lower than 10^{-4} of the ^{133}Ba activity. A special reporting form had been set up in order to collect the information necessary for interpreting the results. The most important part of this information is reproduced in Tables 2 and 3.

Table 1

List of the participants

AECL	Atomic Energy of Canada Limited, Chalk River, Canada
BCMN	Bureau Central de Mesures Nucléaires, Geel, Belgium
BIPM	Bureau International des Poids et Mesures, Sèvres, France
LMRI	Laboratoire de Métrologie des Rayonnements Ionisants, Saclay, France
OMH	Orszagos Mérésügyi Hivatal, Budapest, Hungary
PTB	Physikalisch-Technische Bundesanstalt, Braunschweig, Federal Republic of Germany

Well-established techniques for source preparation and activity measurements were used and did, except for LMRI, not seem to present any particular difficulties. Nevertheless, unusually large discrepancies in the final results appeared which one is tempted to ascribe to the relatively high proportion of activity adsorbed on the ampoule walls as was observed by several participants. However, since this effect has not yet been studied systematically, it is not clear to what extent it should be corrected for. Therefore, no adsorption corrections have been applied.

Additional measurements were carried out by the Centre d'études nucléaires (SCK) at Mol, Belgium, using a large NaI well crystal detector. Four sources prepared by BCMN gave radioactivity concentrations of (1 119.2 ± 9.8) and (1 101.0 ± 9.8) Bq mg^{-1} at the reference date, for the ampoule numbers 7 and 5, respectively. These results and the one which LMRI obtained by the same technique depend to a certain extent on the values used for the decay parameters.

Finally, the international reference system for activity measurements of γ-ray emitting nuclides enables us to compare the present results with earlier ones obtained at other laboratories (see Fig. 1).

(14 April 1981)

Table 2	AECL	BCMN	BIPM	LMRI	OMH	PTB
Dilution factor(s)	8.282 5	–	–	–	1.997 6	–
Source preparation		2 ampoules:				
Total backing mass per unit surface ($\mu g\ cm^{-2}$)	30	No.7: 50	100	30	30	60
Wetting agent	Catanac	No.7: Ludox 10^{-4}	Ludox SM 10^{-4}	Insulin	Ludox	Ludox SM 10^{-4}
Number of sources measured	22	No.7: 5	10	12	12	4
" " " used in final result	22	No.7: 5	10	11	12	4
Range of source mass	12 to 33	13 to 16	14 to 50	9 to 53	12 to 55	11 to 23
Activ. adsorbed on amp. walls(%)	0.34 ± 0.07 (after 1 month)	–	0.6	–		0.46 ± 0.04
4π proportional counter						PPC / PC
Gas, pressure (MPa)	CH_4; Ar + CH_4; 0.1	CH_4 0.1	CH_4 0.1	Ar + CH_4 0.91	Ar + CH_4 0.4	Ar/CH_4 2 / CH_4 0.1
Discrimination level (keV)	≈ 0.1	3.0	≈ 2		0.6	6 to 32 / 0.7
Gamma-ray detector(s) Number	1 NaI	1 NaI	1 NaI	1 Ge	1 NaI	1 NaI / 1
Diameter/height (mm)	76/76	76/76	76/76	–	76/76	76/76
Resolution at 662 keV (%)	6.7	6.3	7.6	2 (at 1 332 keV)	8.2	9 / 8
Dead times τ_β; τ_γ (μs)	2.01 ± 0.02	3.98; 4.08 ± 0.02 / 6.44; 6.19	4.42; 4.41 ± 0.01	5; 5	3.138; 3.011 ±0.005	5.10; 5.00 ± 0.05
Coincidence resolving time (μs)	0.698 ± 0.01	1.35 ± 0.08 / 0.98	1.06 ± 0.01	1	1.053 ± 0.010	1.00 ± 0.02
Counting data, effic. functions						
γ- channel window limits (keV)	200 to 435	250	250 to ∞ / 320 to 410	272 to 387(4peaks)	100–490 / 240–490	250 to ∞ / 250 to 500
Number of data points	99 **	40 / 60	32 / 34	28 / 28	12 / 12	48 / 32
" of sources measured	22	5 / 12	6 / 4	12 / 12	12 / 12	4 / 5 (+ 13)
Range of effic. param. N_c/N_γ (%)	74 to 60	60 to 30	63 to 8 / 60 to 15	81 to 14	82 to 45	67 to 45; 66 to 48
Method for varying N_c/N_γ		absorber foils	absorber foils	discrimination		
Mean time of one data point (s)	600	1 000	4 800 / 4 000	4 000	1 000 to 1 500	12 000
Order of fitted polynomial*	1	1 / 1	2 / 2	*** / 1	1 / 1	1 / 1
Number of degrees of freedom	98	3 to 9 / 7	29 / 31	23 / 26	11 / 11	46 / 30
Intercept, y (x → 0) ($Bq\ mg^{-1}$) at reference date	1 110.2 **	1 130.5 / 1 115.6	1 125.4 / 1 129.8	1 135 / 1 105	1 116.7 / 1 118.1	1 127.7 / 1 129.0 (1 128.6)
Standard deviation ($Bq\ mg^{-1}$)	1.3	0.8 / 0.6	1.1 – 4.1	11 / 0.41	0.17 / 0.15	2.2 / 0.4 (0.9)
Coincidence formula used	Bryant	Cox-Isham	Cox-Isham		Cox-Isham	Cox-Isham

* $y = a_0 + a_1 x + a_2 x^2 + \ldots = (N\beta\, N\gamma)/(m\, N_c)$, $x = (1 - N_c/N\gamma)/(N_c/N\gamma)$, ** Further data are quoted on the next page, *** Multiparametric adjustment

Table 3	AECL	BCMN		BIPM	LMRI		OMH	PTB	
Uncertainty components of the final result (approximations of the corresponding standard deviations, in %) due to counting statistics	0.06	0.2*		0.111	*		0.01	PPC 0.20	PC 0.08
weighing	0.01	0.07		0.033	0.003 3		0.01	0.02	0.02
dead time	0.01	0.05		0.002	10^{-3}		0.01	0.01	0.01
resolving time	0.01	0.05		0.034	10^{-3}		0.03	0.03	0.04
delay mismatch	0.01	0.06		0.020	0.005 (jitter)		0.05	0.01	0.01
pile-up	small	-		-	-		-	0.01	0.01
background	0.01	0.01		0.050	*		0.01	0.02	0.02
timing	negligible	-		0.001	0.003 3		0.01	0.01	0.01
fitting procedure	0.23	0.05		0.023	0.97	0.036	0.10	0.10	0.10
adsorption	≈ 0.3	0.08		0.10	-		-	0.05	0.05
others	0.02 (decay) 0.01 (impurity)	* included in fitting procedure		0.007 (decay)	-		0.02 (decay)	-	-
Combined uncertainty (square root of summed squares)	0.39	0.16	0.15	0.17	* included in fitting procedure 0.97	0.037	0.12	0.23	0.15
FINAL RESULT									
Radioactivity concentration at the reference date (1980-12-01, 0 h UT), combined uncertainty (Bq mg^{-1})	1 111.2 4.3	1 130.5 1.8	1 115.6 1.7	1 125.9 1.9	1 135 11	1 105.0 0.4	1 117.9 1.3	1 127.7 2.6	1 128.6 1.7
How was the final result calculated from the various extrapolations?	weighted mean	-		simultaneous 2nd order fitting			weighted mean	PPC: weighted mean PC: extrapolation of results from 18 sources, using slope obtained with 5 sources.	
Further data from AECL and LMRI									
Number of data points	66	66	43		$4\pi\gamma$ method				
" " sources measured	22	22	2		using a large NaI well crystal				
Range of efficiency parameter (%)	74 to 64	74 to 60	71 to 53						
Mean time for one data point (s)	600	600	1 000		Calculated overall efficiency = 0.991 3 ± 0.003 5				
Order of fitted polynomial	1	1	1						
Number of degrees of freedom	65	65	42						
Intercept (Bq mg^{-1})	1 112.0	1 110.3	1 111.4	1 113.7	Radioactivity concentration = (1 119.0 ± 1.3) Bq mg^{-1}				
Standard deviation (" ")	2.4	1.5	1.1	2.1					

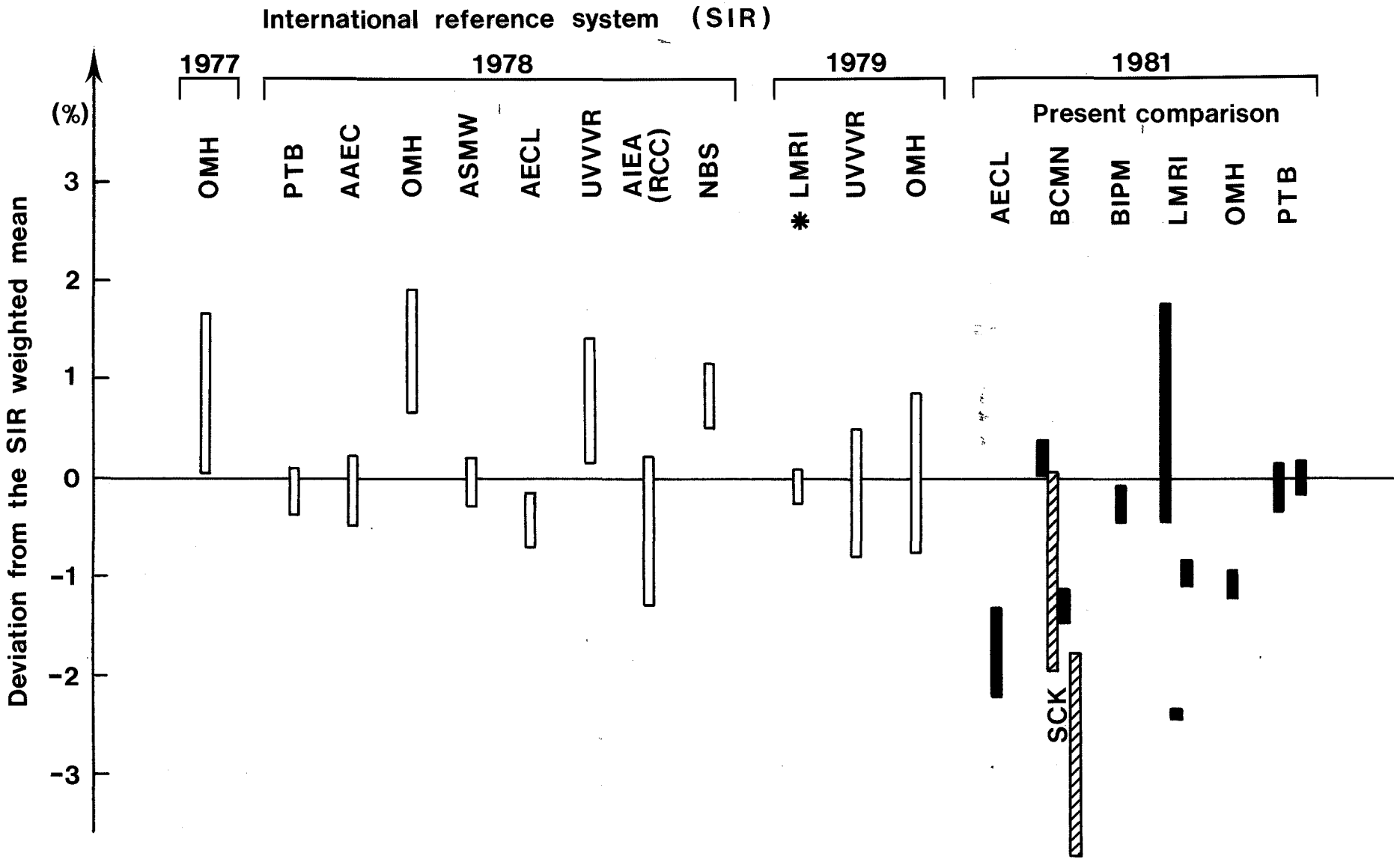

Fig: 1 ^{133}Ba activity measurements, results and (combined) uncertainties

* solution used in the ICRM comparison