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Report on the international comparison of activity measurements of a solution of ^{139}Ce (March 1976)

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CONTENTS

ABSTRACT

Pursuing the programme of international comparisons of radionuclides organised by the Bureau International des Poids et Mesures, 23 national and international laboratories have measured the radioactivity concentration of samples of a solution of ^{139}Ce. The active material was prepared by the National Physical Research Laboratory (Pretoria) and purity-checked by the Institut d'Electrochimie et de Radiochimie de l'Ecole Polytechnique Fédérale (Lausanne) and the Laboratoire de Métrologie des Rayonnements Ionisants (Saclay). The solution was bottled and distributed by the Agence Internationale de l'Energie Atomique (Vienna) in March 1976.

In most cases a 4π proportional gas flow counter (with atmospheric or higher pressure) or a liquid scintillation counter in coincidence with a NaI scintillation counter was used. The results obtained are presented and discussed.

Details on source preparation, counting equipment and data analysing are reported in tabular or graphical form. Special attention is given to the formulae used, the corrections applied and the uncertainties assessed.

1. INTRODUCTION

In 1970 it was decided [1] to suspend the programme of international comparisons organised under the aegis of the Bureau International des Poids et Mesures (BIPM) for a period. As the spread of the results obtained had not decreased as much as expected, Section II (Mesure des Radionucléides) of the Comité Consultatif pour les Etalons de Mesure des Rayonnements Ionisants (CCEMRI) expressed the view that the expenditure of effort on full scale comparisons was too large considering their low efficiency. In the meantime several Working Parties had been formed in order to study some special problems related to the measurement of activity. The results of some of these studies have been published (see e.g. [2, 3]); others are still in progress. It may be expected that these publications will help to avoid some experimental pitfalls and to improve the accuracy of future results. In addition, a special working party was charged with the choice of radionuclides and the preparation of future comparisons. Five proposals made by this group were submitted to the members of Section II whose answers led to the following in decreasing order of preference: ^{139}Ce, ^{134}Cs, ^{57}Co, ^{241}Am, ^{35}S. Preliminary comparisons with a reduced number of participants [4, 5, 6] were organised for the first three of these radionuclides; they permitted to clarify certain problems and to work out an appropriate reporting form. Previous international comparisons had already well demonstrated how important such details can be for the success of the whole enterprise.

The forms to be used for the present comparison were distributed to the participants in December 1975 (see specimen, Fig. 1) along with some instructions and remarks. The reference date had been fixed as 1976-03-15, 00 h UT.

In a preliminary report [7] the results and the most important data submitted by twenty participants have been circulated (1976-07-15). Three more results arrived a few weeks later. The list of the participants is reproduced in Table 1.

2. DESCRIPTION OF THE SOLUTION AND OF THE PURITY TESTS

The primary ^{139}Ce was produced [8] by bombardment of a lanthanum target with 16 MeV deuterons, at the NPRL*, according to the reaction

$$^{139}La(d, 2n)^{139}Ce.$$

After bombardment the active surface layer was shaven off and ^{139}Ce separated by a solvent extraction method [9]. The cerium was then reduced to the trivalent state, back extracted into an aqueous phase and evaporated to dryness. After destruction of the remaining organic material by evaporation with concentrated HNO_3, the cerium was taken up in diluted hydrochloric acid (1 mol HCl in 1 dm^3 of H_2O).

Much attention was paid to extensive purity tests and to the choice of the composition of the solution to be distributed.

Samples from a test run were sent to IER and LMRI in May 1975. Each sample contained about 10^8 Bq of ^{139}Ce, carrier free in about 1 cm^3 of diluted HCl. These two laboratories carried out purity tests by γ-ray spectrometry (Ge(Li) and superpure Ge detectors), β-ray spectroscopy, half-life measurement and determination of the slope/intercept ratio by 4π(PC)-γ counting. Similar checks were made by the same two laboratories, eight months later, with samples from the main run. Each laboratory sent comprehensive reports on their measurements to the BIPM in October 1975 and February 1976.

The results of these purity tests were slightly different from one run to the other and from one laboratory to the other. Nevertheless, these were minor differences, and the radionuclidic purity of the ^{139}Ce could be considered as sufficient in each case.

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* The full names of the laboratories can be found in Table 1.

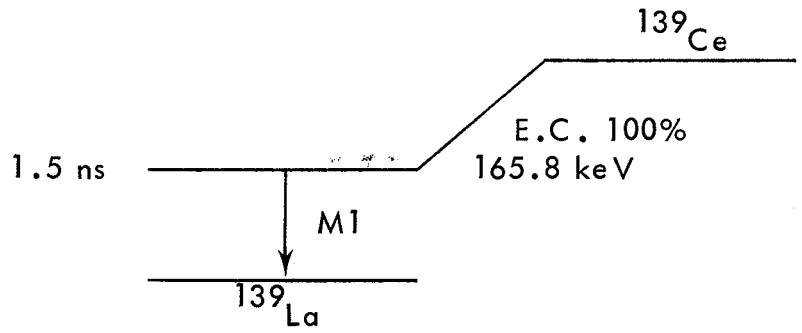
Table 2 gives a summary of the results obtained. By far the most important contamination is due to ^{141}Ce, not detected by IER because of too low a sensitivity of their particular equipment. However, the activity ratio, at the reference date of the comparison (1976-03-15), was $A_{141}/A_{139} = 1.6 \times 10^{-4}$, i.e. still negligible, at least for activity measurements.

The dilution of the main bulk to about 700 $kBq \cdot g^{-1}$, the addition of the carrier, the bottling of the solution and the dispatch of the samples were carried out by the AIEA at Seibersdorf (near Vienna).

In the first week of March 1976, each participant received two ampoules (type NBS) containing each about 3.5 g of solution. The chemical composition was 20 μg of $CeCl_3$ per gramme of an aqueous solution of 0.2 mol HCl in 1 000 cm^3. The mass of each sample had been determined by AIEA to $\pm$ 10 μg in order to make possible certain adsorption tests.

Decay scheme and nuclear data

The following data have been recommended for use to all the participants prior to the comparison.



$T_{1/2} = (137.65 \pm 0.07)$ d [10]

Energies of the radiations emitted: X rays mostly 33 and 38 keV,
Auger electrons $\leq$ 37 keV,
γ rays 165.8 keV,
conversion electrons $\geq$ 127 keV,
(total conversion coefficient $\alpha_{tot} \approx 0.25$).

3. ADSORPTION TESTS

Although the chemical composition of the solution distributed had been chosen so as to prevent adsorption at the walls of the glass ampoules, the following three procedures have been suggested by the Working Party in order to evaluate upper limits, using ion chamber measurements.

1. Comparison of the radioactivity concentration in the pycnometer before and after dispensing the sources.

2. Comparison of the solution in the original ampoule before breaking the seal to
   a) that part of the original solution which was left after source preparation and transfer to a new ampoule of the same type (for this purpose each participant had also received two empty ampoules),
   b) the small amount not extracted from the original ampoule.
   In 2a) and b) diluant had to be added in order to get the same volume of liquid as originally present.

Table 3 gives a summary of the results obtained. The methods 1) and 2a) being rather similar, they are not quoted separately. Method 2b) was not very sensitive, due to the low energy of the γ rays and the small amount of activity left in the original ampoules. As to method 2a), the precision seems to be quite good. However, the fact that NPL and BIPM found "negative adsorption" of nearly 0.1% suggests the presence of systematic errors of this order of magnitude. Thus, to this order of accuracy it seems unlikely that significant adsorption took place. The result obtained by ASMW deserves attention but does not seem to have influenced the final result of this laboratory.

4. SOURCE PREPARATION FOR MEASUREMENTS WITH PROPORTIONAL COUNTERS

All the information contained in the individual forms has been condensed in Table 4 which is self-explanatory.

The column headed "Range of N_c/N_γ" gives the range of efficiency of the proportional counter to Auger electrons and X rays. It deserves special attention, since high efficiency is always an advantage. As there is no clear correlation between the application of certain seeding or spreading agents and the highest efficiency obtained, the skill of the operator seems to be more important than the particular treatment applied.

The most frequently used backing material is metal-plated VYNS. One laboratory (SCK) dispensed sources on non-metallized VYNS films and compared, in a later experiment, ^{60}Co sources on metallized films with sources on non-metallized ones. Although practically no difference in radioactivity concentration was found, it is evident that the results obtained by SCK contain a large systematic error which may be due to charging-up of the sources. Therefore it was decided to withdraw them from Table 11 and Figures 4, 5 and 6.

5. LIQUID SCINTILLATION COUNTING

Only three laboratories have used 4π (LS)-γ counting in this comparison. Source preparation and equipment are described in Tables 5 and 6. Various procedures for varying N_c/N_γ were applied and the quality of the results obtained differed considerably from one laboratory to the other.

The three different gates in the γ channel used by IBJ (Table 9 and [7]) gave widely different and incompatible results. However, further experiments carried out by this laboratory showed that the highest and the lowest results are in error due to an excess and a loss, respectively, of coincidences. Therefore, only the intermediate result has been maintained.

As the result reported first by NPRL [7] was high by 1.0 to 1.5 %, this laboratory further investigated the performance of its equipment. It appeared that, despite the use in coincidence of two photomultiplier tubes in the 4π (LS) detector, spurious pulses had not been eliminated sufficiently. Therefore, two variations of the so-called gating technique [3] were applied for determining the probability for the production of spurious pulses. It could be shown [11] that a correction of $(1.3 \pm 0.2)\%$ has to be applied. This brings the result of NPRL down to a value in close agreement with others.

The special equipment used at NPL allowed a more sophisticated extrapolation procedure, which is described in [12], to be applied. A single γ window over the photopeak or a wide γ window from 50 keV to above the photopeak did not yield a satisfactory polynomial fit of the efficiency function. However, the simultaneous use of two windows, one over the photopeak, the other below and excluding the photopeak, gave a good fit. The correlation of the data points has been accounted for in a special fitting procedure. However, a relatively high systematic uncertainty (1.5%) subsisted. Correlation counting was used in order to estimate the upper limit of the spurious pulse rate (cf. [3]).

6. EQUIPMENT, BLOCK DIAGRAMMES, COINCIDENCE-COUNTING DATA

Details of the counting equipment as reported by the participants are described in Tables 6 and 7. Table 8 lists all the data concerning dead times, resolving times and the methods used for their determination.

Many participants have supplied block diagrammes of their electronic system (see Fig. 2). Others have quoted the corresponding references which may be found in Tables 7 and 8.

As is well known, a relative delay between β and γ channels can change the coincidence rate ("Gandy effect"). Such delays have been measured, but were found to be certainly less than 0.5 μs and often much shorter. The corresponding corrections applied to the final results are given in Table 9. The methods used for determining or eliminating the delay are described in [13] and [14].

7. EXPERIMENTAL DETAILS OF SPECIAL COINCIDENCE-COUNTING METHODS

Two laboratories have, in addition, made use of three methods, the description of which did not fit into the preceding tables. Therefore, they are summarized separately hereafter.

a) $\underline{X_K}$(NaI)-γ counting (IMM)

	X channel	γ channel
NaI(Tl) crystal (mm)	d = 30, h = 2	d = 30, h = 20
Background rate (s^{-1})	0.16	1.1 ($B_c = 0$)
Dead time (μs)	2.34 ± 0.05	2.34 ± 0.05
Resolving time (μs)	0.915 ± 0.007	
	1.475 ± 0.010	

Thirteen sources were measured during 1 000 s for each data point. The activity A was calculated as follows:

$$A = N_o \frac{P_K}{P_K + \alpha_K/(1+\alpha_K)} = N_o/1.232\,8$$ (from Nucl. Data Sheets 12, 2 (1974)),

where

N_o = activity calculated by means of Campion's formula,

P_K = K capture probability,

α_K = K shell conversion coefficient.

Components of systematic uncertainty:

weighing	0.05 %
dead-time correction	0.005
resolving time	0.02
afterpulses	0.05
constants (decay scheme)	0.7
total	0.7 %

b) $2\pi X_L$ (PC)-γ counting (IMM)

- L X-ray detector: semi-cylindrical 2π proportional counter made of aluminium, d = 180 mm, l = 80 mm; anode: constantan, d = 0.1 mm, l = 125 mm, distance from source 35 mm; 2.3 kV; Ar/CH_4 at atmospheric pressure, discrimination level 100 eV.
- γ-ray detector: one NaI(Tl) crystal, d = 40 mm, h = 30 mm.
- Dead times (μs): $\tau_x = \tau_\gamma = 2.34 \pm 0.05$
- Resolving time (μs): $\tau_r = 1.475 \pm 0.010$
  2.08 ± 0.01.
- Background rates (s^{-1}): $B_x = 2$, $B_\gamma = 5$, $B_c = 0$.

$$A = N_o\left(1 + \frac{n_K \cdot n_{KL} + n_L}{P_K \cdot n_K + P_L}\right) = N_o/1.216$$ (see Nucl. Data Sheets),

where

N_o = calculated activity,

$n_K (n_L)$ = relative number of K(L)-shell vacancies resulting from internal conversion of γ quanta,

n_{KL} = relative number of L-shell vacancies resulting from the formation of a K-shell vacancy,

$P_K (P_L)$ = K(L) capture probability.

Thirteen sources were measured during 1 000 s for each data point.

Components of systematic uncertainty:

weighing	0.05 %
dead-time correction	0.005
resolving time	0.02
background	0.005
afterpulses	0.05
constants (decay scheme)	0.8
total	0.8 %

c) 4πSi(Li)-γ counting (NPL)

- 4π detector: Li-drifted Si detector at 77 K, thickness (each half) 3 mm, radius 8 mm, active area 200 mm^2, depletion depth 3.0 mm, window thickness $\geqslant$ 0.2 μm (50 $\mu g \cdot cm^{-2}$), detector-source separation 0.13 mm, pressure 1.3 mPa, voltage 700 V.
- γ-ray detector: three NaI crystals (one well type), diameter 76 (well 102) mm, height 76 (well 152) mm.

Dead time and resolving time (μs): $\tau_\beta = 8.00 \pm 0.01$, $\tau_\gamma = 6.01 \pm 0.02$, $\tau_r = 2.95 \pm 0.05$.

Background rates (s^{-1}): $B_\beta = 1.3$, $B_\gamma = 1.0$ and 1.9, $B_c = 0.03$.

Ten sources (16 to 21 mg) on Al backings (200 $\mu g \cdot cm^{-2}$) were prepared; 23 data points (1 000 s each) from seven sources were used for extrapolation. N_c/N_γ was varied from 48 to 2% by computer discrimination. Data points were calculated using the same formulae as for 4π(LS)-γ counting. Uncertainties in N_c/N_γ have been considered.

The residuals (Fig. 3) showed a large trend of unknown origin.

8. 4πγ MEASUREMENTS WITH A LARGE CALIBRATED SCINTILLATION DETECTOR (IRK)

The results obtained by this laboratory are not considered to be "absolute" in the same way as is generally accepted for coincidence measurements. Nevertheless, the method summarized here is about midway between absolute and relative.

Source preparation

The total content of ampoule n° 3 was diluted with a diluent of the same composition (dilution factor = 28.076). A new (1975) Mettler balance type P 163 was used for determining the mass of the diluent.

Drops of 14 to 35 mg of the dilution were dispensed onto each of seven backings of diameter 18 mm and thickness 0.1 mm by means of a pycnometer. The source mass was determined simultaneously by differential weighing of the pycnometer and by the evaporation method. For the first method a Mettler balance type H 16 was used which had previously been recalibrated and checked for scale linearity. The samples were placed on the pan of a fast electronic microbalance (Perkin Elmer AD-2) with digital display. Several tens of readings were taken from 15 s until 8 min after dispensing, and extrapolated to zero time. Only those four sources were counted for which both weighings agreed within 40 μg.

The detector was a NaI(Tl) well crystal, d = h = 127 mm, well diameter 27 mm, depth 74 mm, mounted on a RCA 8055 photomultiplier ([15]). Integral counting above 13 keV was carried out with a dead time of (10.3 ± 0.2) μs. The measurements took place from April 26 to April 28; each sample was counted twice and the total counting time was 8 000 s. The count rates ranged from 340 to 740 s^{-1}. The background rate (of about 52 s^{-1}) was measured with a sample prepared from a drop of diluent, before and after each source was counted. The calculated efficiency [16] was 0.952 ± 0.005.

9. COINCIDENCE FORMULAE FOR CALCULATING THE ACTIVITY

Since a rigorous method for computing the sample activity from the observed count rates is still lacking, various approximate forms have been employed. A recent article by Cox and Isham [23] gives the solution which is exact under certain conditions*. The formulae used by the participants are mostly derived from those developed by Campion [17] or by Bryant [18] . It can be seen that the differences between the various expressions are often due to second-order terms or to the way in which background is corrected for. Moreover, some participants have taken into account delay offset or unequal pulse lengths in the two channels. In what follows the decay-scheme-dependent correction will not be considered.

About one half of the participants have used a formula stemming from the well-known paper by Campion [17] and which may also be found in [19] and [20] :

$$N_o = \frac{N_\beta N_\gamma \left[1 - \tau_r (N'_\beta + N'_\gamma)\right]}{(N_c - 2\tau_r N'_\beta N'_\gamma)(1 - \tau' N'_c)} , \qquad \tau_c < \tau' .$$

The symbols used here and later on have the following meaning:

N_o	sample activity,
N'_β, N'_γ, N'_c	observed (uncorrected) count rates in the β, γ and coincidence channels, respectively,
N_β, N_γ, N_c	count rates corrected for background,
τ_β, τ_γ, τ_c	dead times of the respective channels,
τ'	the shorter of τ_β or τ_γ ,
τ_r	coincidence resolving time.

This formula was used by AAEC, BARC, BIPM, ETL, IEA, IER, IMM, OMH and SCK, while PTB added a delay-offset term according to Gandy's theory [21] .

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* For practical implementation, see D. Smith, Improved correction formulae for coincidence counting (Nucl. Instr. and Meth., in press).

Slightly different forms also used were

$$\text{ASMW}: \qquad N_o = \frac{N_\beta N_\gamma}{N_c} \cdot \frac{1 + \tau_r \left[2 \frac{N_\beta N_\gamma}{N_c} - (N_\beta + N_\gamma) \right]}{1 - \tau N_c}, \quad \tau_\beta = \tau_\gamma = \tau,$$

$$\text{NPRL}: \qquad N_o = \frac{N_\beta N_\gamma}{N_c} \left[1 + \tau' N_c + 2 \tau_r \frac{N_\beta N_\gamma}{N_c} - \tau_r (N_\beta + N_\gamma) \right],$$

$$\text{UVVVR}: \qquad N_o = \frac{N_\beta N_\gamma}{N_c} \left[1 + 2 \tau_r \frac{N'_\beta N'_\gamma}{N'_c} - \tau_r (N'_\beta - N'_\gamma) \right] (1 + \tau_\gamma N'_c) .$$

Seven participants have reported formulae derived from equations (4) or (6) given in [18]. As there are some small differences, mainly in the way the background is corrected for, we reproduce here all the expressions indicated by the participants concerned. Additional symbols appearing are

θ_β , θ_γ — duration of pulses from β and γ channels, respectively, arriving at the coincidence mixer; one has always $\theta_\beta + \theta_\gamma = 2 \tau_r$,

δ — delay between β and γ channels, positive when the β channel is delayed,

N^*_β , N^*_γ , N^*_c — count rates corrected for dead time and background rate of genuine coincidences,

B'_β , B'_γ , B'_c — observed background count rates,

Q' — ratio of coincidence to γ-channel count rates, both rates being corrected for dead time and resolving time,

Q — ratio of coincidences to γ-channel count rates, both rates being corrected for dead time, resolving time and background.

- following eq. (6) in [18] :

AIEA: $$N_c^* = \frac{N_c' - 2\,\tau_r N_\beta' N_\gamma'}{\left[1 - \tau_r (N_\beta' + N_\gamma')\right]\left[1 - \tau_\gamma (N_\gamma' - N_c')\right](1 - \tau_\beta N_\beta')} - B_c' \quad , \quad N_o = \frac{N_\beta N_\gamma}{N_c^*} \; .$$

BCMN: $$N_o = \frac{N_\beta N_\gamma (1 - \tau_\beta N_\beta' - \tau_\gamma N_\gamma' + \tau' N_c')\left[1 - \tau_r (N_\beta' + N_\gamma')\right]}{(N_c' - 2\,\tau_r N_\beta' N_\gamma')(1 - \tau_\beta N_\beta')(1 - \tau_\gamma N_\gamma')} \quad , \qquad \text{(see [22])} \; .$$

NPL: 4π(PC)-γ $$N_o = \frac{N_\beta N_\gamma (1 - \tau_\beta N_\beta' - \tau_\gamma N_\gamma' + \tau' N_c')\left[1 - \tau_r (N_\beta' + N_\gamma')\right]}{(N_c - 2\,\tau_r N_\beta' N_\gamma')(1 - \tau_\beta N_\beta')(1 - \tau_\gamma N_\gamma')} \; .$$

- following eq. (4) in [18] :

AECL:
$$N_\beta^* = \frac{N_\beta'}{1 - \tau_\beta N_\beta'} - B_\beta' \; ; \qquad N_\gamma^* = \frac{N_\gamma'}{1 - \tau_\gamma N_\gamma'} - B_\gamma' \; ; \qquad N_o = \frac{N_\beta^* N_\gamma^*}{N_c^*}$$
$$N_c^* = \frac{\left[N_c' - (\theta_\beta + \theta_\gamma) N_\beta' N_\gamma'\right](2 - \tau_\beta N_\beta' - \tau_\gamma N_\gamma')}{\left[2 - \tau_\beta N_\beta' - \tau_\gamma N_\gamma' + 2\,\tau' N_c' - 2(\theta_\gamma N_\beta' + \theta_\beta N_\gamma') + 2\,\delta (N_\beta' - N_\gamma')\right](1 - \tau_\beta N_\beta')(1 - \tau_\gamma N_\gamma')} - B_c' \; .$$

LMRI: $$N_o = \frac{N_\beta N_\gamma}{N_c - 2\,\tau_r N_\beta' N_\gamma'}\left[1 + \frac{2\tau N_c' - 2\,\tau_r (N_\beta' + N_\gamma')}{2 - \tau (N_\beta' + N_\gamma')}\right] \quad , \quad \text{with } \tau_\beta = \tau_\gamma = \tau \; , \; \tau_r < \tau/2 \; .$$

NBS and NRC:
$$Q' = \frac{(N_c' - 2\,\tau_r N_\beta' N_\gamma')\left[1 - \frac{\tau}{2}(N_\beta' + N_\gamma')\right]}{N_\gamma'(1 - \tau N_\beta')\left[1 - \frac{\tau}{2}(N_\beta' + N_\gamma' - 2\,N_c') - \tau_r (N_\beta' + N_\gamma')\right]} \quad , \quad \tau_\beta = \tau_\gamma = \tau$$
$$Q = \frac{Q' - B_c'/N_\gamma'}{1 - B_\gamma'/N_\gamma'} \quad , \qquad N_o = \frac{\left[N_\beta'/(1 - \tau N_\beta')\right] - B_\beta'}{Q} \; .$$

Special formulae

IPA used a formula which contains also higher-order terms in τ and τ_r, with $\tau = \tau_\beta = \tau_\gamma$:

$$N_c^* = \frac{(N'_c - B'_c)\left[1 + \tau\left(\frac{N'_\beta}{1-\tau N'_\beta} + \frac{N'_\gamma}{1-\tau N'_\gamma}\right) - \tau^2 \frac{N'_\beta}{1-\tau N'_\beta}\cdot\frac{N'_\gamma}{1-\tau N'_\gamma}\right] - 2\tau_r \frac{N'_\beta}{1-\tau N'_\beta}\cdot\frac{N'_\gamma}{1-\tau N'_\gamma}}{1 + (N'_c - B'_c)\tau\left[1 + \frac{\tau}{2}\left(\frac{N'_\beta}{1-\tau N'_\beta} + \frac{N'_\gamma}{1-\tau N'_\gamma}\right)\right] - \tau_r\left(\frac{N'_\beta}{1-\tau N'_\beta} + \frac{N'_\gamma}{1-\tau N'_\gamma}\right) + \frac{\tau_r^2}{2}\left(\frac{N'_\beta}{1-\tau N'_\beta} - \frac{N'_\gamma}{1-\tau N'_\gamma}\right)^2} ;$$

$$N_\beta^* = \frac{N'_\beta}{1-\tau N'_\beta} - B'_\beta , \qquad N_\gamma^* = \frac{N'_\gamma}{1-\tau N'_\gamma} - B'_\gamma .$$

For its set-up with three photomultipliers (see Fig. 2), IBJ developed the following expression

$$N_\beta = N_{K1} = \frac{N'_{K1}}{1 - \tau_{\beta 1} N'_{\beta 1} - \tau_{\beta 2} N'_{\beta 2} + \tau_{\beta \min} N'_{K1}} - B'_{K1} , \qquad N_\gamma = \frac{N'_\gamma}{1 - \tau_\gamma N'_\gamma} - B'_\gamma ,$$

$$N_c^* = N_{K2} = \frac{\dfrac{N'_{K2}}{1 - \tau_{\beta 1} N'_{\beta 1} - \tau_{\beta 2} N'_{\beta 2} - \tau_\gamma N'_\gamma + \tau_{\beta \min} N'_{K1} + \tau_{\min} N'_{K2}} - 2\tau_r N'_\gamma N'_{K1}}{1 - (N'_\gamma + N'_{K1})\tau_r} - B'_{K2} ,$$

where

$\tau_{\beta\,\min}$ is the shorter of $\tau_{\beta 1}$ or $\tau_{\beta 2}$,

$\tau_{\min}$ is the shorter of $\tau_{\beta 1}$, $\tau_{\beta 2}$ or τ_γ .

With both methods, 4π(LS)-γ and 4πSi(Li)-γ, NPL used the same formula:

$$N_o = \frac{N_\beta N_\gamma}{N_c - 2\tau_r N'_\beta N'_\gamma} \cdot \frac{\left[1 - \tau_r (N'_\gamma + N'_{\beta tot})\right] (1 - \tau_\beta N'_{\beta tot} - \tau_\gamma N'_\gamma + \tau_\gamma N'_{ctot})}{(1 - \tau_\beta N'_{\beta tot})(1 - \tau_\gamma N'_\gamma)} .$$

Here

N'_β and N'_c refer to the count rates above a chosen β energy,

$N'_{\beta tot}$ and N'_{ctot} are the rates over all β energies [12]. The equation refers to one γ window and becomes more complicated where two windows are employed.

Finally, we mention the formulae developed by NRC for the anti-coincidence method [24]:

$$N_\beta = \frac{C_\beta - A}{t'} - B'_\beta , \qquad N_\gamma = \frac{C_\gamma}{t - C_\gamma \tau_\gamma} - B'_\gamma ,$$

$$A = \frac{C_\beta C_Y \delta}{t' + \delta (C_\beta + C_Y)} , \qquad N_y = \frac{C_Y - A}{t'} - B'_Y .$$

Here

C_β , C_γ , C_Y are the accumulated counts in the β, γ and Y channels, respectively (see Fig. 2),

A is the number of accidental events in the β or Y channels,

t = real time,

t' = live time,

δ = delay gap ($\approx$ 13 ns).

10. EFFICIENCY FUNCTIONS, POLYNOMIAL FITTING

The simplicity of the ^{139}Ce decay scheme makes this radionuclide especially well suited for coincidence counting with efficiency extrapolation. This fact and the absence of particular difficulties (as e.g. impurities, insoluble salts) have already been realized on the occasion of the preliminary comparison organized by the BIPM [5] in 1974. Experimental and

theoretical justification of the extrapolation method has been discussed in detail [25] and systematic errors are likely to be rather small.

The efficiency function has been approximated by a polynomial $y = a_o + a_1 x + a_2 x^2 + \ldots$, when expressed in the coordinates $y = \frac{N_\beta N_\gamma}{m N_c}$ and $x = \frac{1 - N_c/N_\gamma}{N_c/N_\gamma}$, where the count rates are the same as in the preceding section and m is the source mass.

The methods used for varying N_c/N_γ, the efficiency of the 4π counter, are indicated in Table 4, where it may be seen that, besides counting gas variation, self-absorption, foil absorption and threshold level variation were applied. Self-absorption may be varied by redissolving a source and adding inactive carrier or by using several sources of different mass. The latter procedure is simpler but less reliable, since individual efficiency functions may differ from each other. However, no such difference has been reported. The coefficients a_o, a_1, a_2, ..., have been least-squares fitted to the data points (x, y) in the well-known manner.

The graphical representation of the residuals of the data points obtained from the various participants are reproduced in Fig. 3.

As one could expect, the efficiency functions turned out to be very nearly linear in most cases. The majority of the participants have also calculated second-order fits. However, the ratio a_2/a_1 seldom exceeded 0.002 5; in a single case (LMRI) it was as high as 0.012. Third and higher orders could always be neglected.

The order of the "best fit" can be determined by comparing the results of χ^2 tests from adjustments of different orders. Such tests have been reported by the participants and are listed in Table 10.

In the graphical representation of the efficiency function, the variables x and y are in general correlated and this should be accounted for in the calculation of the variance and the statistical weights.

In fact, if for ^{139}Ce the measurement of a data point corresponding to a fairly low efficiency, N_c/N_γ, is repeated a large number of times and plotted in a y(x) diagram, the correlation becomes obvious [26].

The estimation of the residual and its variance has been treated by Adams and Baerg and by Baerg [27]. Recently, Merritt et al. [29] have applied this approach to the case of a linear efficiency function $y' = y_o (1 + Kx)$, where the intercept y_o and the slope K are estimated from a preliminary fit (using e.g. equal weights). A better estimate of the weight will then be given by the inverse variance of the residual r'. One has, with m = source mass and $x = \frac{1 - N_c/N_\gamma}{N_c/N_\gamma}$:

$$r' = N_\beta N_\gamma/(m N_c) - y_o (1 + Kx) = \frac{1}{m}\left[N_c + X + Y + (X Y/N_c) - m y_o - (m y_o K Y/N_c)\right].$$

Here $X = N_\beta - N_c$ and $Y = N_\gamma - N_c$ are the non-coincident count rates, as in [28]. The variance is then

$$\sigma^2_{r'} = (\partial r'/\partial N_c)^2 \sigma^2_{N_c} + (\partial r'/\partial X)^2 \sigma^2_X + (\partial r'/\partial Y)^2 \sigma^2_Y .$$

Remembering that $\sigma^2_{N_c} = N_c/t$, $\sigma^2_X = X/t$, $\sigma^2_Y = Y/t$ (t = count duration) and putting $W = N_\beta - N_c - (m y_o K/D)$, where D is the decay correction to the reference time, one gets finally

$$\sigma^2_y = D^2 \sigma^2_{r'} = \left\{\left[1 - (Y W/N_c^2)\right]^2 N_c + \left[1 + (Y/N_c)\right]^2 X + \left[1 + (W/N_c)\right]^2 Y\right\} D^2/(m^2 t) .$$

For a well-designed experiment (i.e. with a reasonable distribution of points having comparable statistical accuracies), the computed value of the slope and intercept will not be sensitive to the choice of weights. The proper choice of weights may, however, yield significantly better estimates for the variances of the parameters and is essential if a meaningful value of χ^2 is required (cf. A.P. Baerg, references 6 and 7 in [27]). A second iteration using the new values of the parameters to recalculate $\sigma^2_{r'}$ for each data point is unlikely to be worthwhile because the variance estimates for the individual points are relatively insensitive to small changes in the parameters.

11. FINAL RESULTS, UNCERTAINTIES

The extrapolation to 100% efficiency N_c/N_γ leads to an intercept a_o with the ordinate y which represents the radioactivity concentration of the solution considered. The fitting procedure also includes the calculation of the standard error of the mean. The number of degrees of freedom (in general the number of data points used minus the number of coefficients a_o, a_1, ... fitted) helps then to define the random uncertainty of the final result. Eight participants have also taken into account the uncertainty in x (Table 9); the effect on the final result was always smaller than one part in 10^3.

The final results, as well as the random and total systematic uncertainties, are listed in Table 11. A graphical representation is given in Fig. 4 and a histogramme of the distribution in Fig. 5.

The various contributions to the estimated systematic uncertainties are listed in Table 12, where it is also explained how they were obtained. The way of combining them is not of great importance in the present case, since one of them is almost always by far the largest. Usually the linear sum was taken.

The largest contribution to systematic uncertainty is the one which is due to the extrapolation procedure. Most participants put it equal to the difference in a_o between first- and second-order fits. This difference and the corresponding estimate by the participant are listed separately.

12. SLOPE-TO-INTERCEPT RATIO

The slope a_1 of the efficiency function near to the intercept divided by a_o is an important parameter, since it depends in a simple way on the total internal conversion coefficient of the daughter atom and is, at least approximately, independent of the counting apparatus. However,

the histogramme of Fig. 6 shows a considerably larger spread than that of Fig. 5. As is evidenced by Fig. 7, the intercept and the slope are correlated, although not in a very clear way.

The slope-to-intercept ratio a_1/a_o is related to the total internal conversion coefficient α according to [30]

$$a_1/a_o = \frac{\varepsilon_{\beta\gamma} - p_c}{1+\alpha} + \frac{\alpha}{1+\alpha}\left[\varepsilon_e + (1-\varepsilon_e)\,\varepsilon_X\right],$$

where

$\varepsilon_{\beta\gamma}$ is the efficiency of the "β" counter to 166 keV γ rays,
ε_e " " " ≥127 keV electrons,
ε_X " " " X rays or Auger electrons,
p_c is the probability for registering a coincidence event when the electron capture event is not registered in the "β" counter.

It is generally assumed that ε_e is close to unity and p_c negligible. Thus

$$a_1/a_o \approx \frac{\alpha + \varepsilon_{\beta\gamma}}{1+\alpha}.$$

The efficiency $\varepsilon_{\beta\gamma}$ is difficult to measure accurately. Merritt and Taylor [31] found that it is small and not strongly dependent on the size, shape or material of the cathode. From the work of Urquhart [32] or of Williams and Campion [33] not much information concerning 166 keV γ rays can be gathered. Plch et al. [30] estimate, for their Ar/CH_4-filled pressurized (0.5 MPa) counter, a value of $\varepsilon_{\beta\gamma} \approx (0.7 \pm 0.2)\%$. On the other hand, in the context of the preliminary comparison [5], PTB estimated, for CH_4 and atmospheric pressure, $\varepsilon_{\beta\gamma} \approx (5.0 \pm 1.5) \times 10^{-4}$; Taylor [34] indicated $\varepsilon_{\beta\gamma} \approx (4 \pm 1) \times 10^{-4}$. Therefore, $\varepsilon_{\beta\gamma}$ may be neglected in most of the results obtained in this comparison. From Figs. 6 and 7 it can be seen that the results cluster around a slope-to-intercept ratio of 0.200 5,

corresponding to a value of the total internal conversion coefficient of $\alpha \approx 0.250\,2$ with an uncertainty of not more than $0.002\,5$. Various measurements* of this coefficient have been published in the past:

First author	Reference	α	
Taylor	[34]	$0.250\,8 \pm 0.001\,4$	
Aristov	[35]	0.254 ± 0.006	
Legrand	[36]	$0.244\,6 \pm 0.001\,2$	
Plch	[30]	0.251 ± 0.002	
Hansen	[37]	0.252 ± 0.005	(deduced)
present		$0.250\,2 \pm 0.002\,5$	

The result suggested by this comparison agrees with most of the previous measurements, but does not confirm the value by Legrand et al.

13. CONCLUSION

Seventy percent of all the national and international radionuclide metrology laboratories which had been asked by the BIPM to participate accepted to do so. With one exception, all the results were obtained by coincidence counting with efficiency extrapolation. Although many different detector types and coincidence set-ups were used, a general systematic deviation of the results cannot be excluded completely. Therefore, no attempt has been made at deriving a mean value of the radioactivity concentration of the solution distributed. Nevertheless, the fact that the results of twenty-two participants give a total spread of only 1.1% (compared to 0.6% with five selected participants in 1974 [4]) is very gratifying. It may in addition be interpreted as expressing the ability of the participants to dispense sources correctly.

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* Note added in proof: E. Schönfeld and R. Brust report a value of $\alpha = 0.251\,9 \pm 0.000\,6$ (Isotopenpraxis 13, 311 (1977)).

This intercomparison has shown that the standardization of ^{139}Ce by conventional coincidence techniques is relatively easy; linear efficiency extrapolation is often sufficient, although the total systematic uncertainty seems mostly due to the extrapolation procedure. As the decay scheme of ^{139}Ce has much in common with that of ^{203}Hg, the difficulties previously encountered in standardizing the latter are not likely to be due to the measuring method but rather to some chemical properties of mercury compounds. Liquid scintillation counting has been applied successfully by three participants. However, in each case some particular difficulty (spurious pulses, loss or excess of coincidence events, special fitting procedure) called for a more elaborate treatment. On the other hand, proportional counting seems to be much less affected by these difficulties. Moreover, gas pressure and composition had no incidence on the quality of the results. Finally, the correlation between the extrapolated activity value and the slope-to-intercept ratio of the efficiency function may be due to incomplete knowledge of the various detector efficiencies or to insufficiently precise counting corrections.

Table 1

List of the 23 participants

		Names of the persons who carried out the measurements
AAEC	Australian Atomic Energy Commission, Lucas Heights, Australia	G.C. Lowenthal
AECL	Atomic Energy of Canada Limited, Chalk River, Canada	J.S. Merritt, F.H. Gibson, J.G.V. Taylor
AIEA	Agence Internationale de l'Energie Atomique, Vienna, Austria	H. Houtermans, E. Wehrstein
ASMW	Amt für Standardisierung, Messwesen und Warenprüfung, Berlin, German Democratic Republic	E. Schönfeld
BARC	Bhabha Atomic Research Centre, Trombay, India	S. Nagpal, P.K. Srivastava
BCMN	Bureau Central de Mesures Nucléaires d'Euratom, Geel, Belgium	I. Goodier, E. Celen, W. Zehner
BIPM	Bureau International des Poids et Mesures, Sèvres, France	C. Colas, C. Veyradier
ETL	Electrotechnical Laboratory, Tokyo, Japan	O. Yura, Y. Kawada
IBJ	Instytut Badán Jadrowych, Świerk, Poland	P. Zelazny
IEA	Instituto de Energia Atômica, Pinheiros-São Paulo, Brazil	Cl. Renner
IER	Institut d'Electrochimie et de Radiochimie de l'Ecole Polytechnique Fédérale, Lausanne, Switzerland	J.-J. Gostely
IMM	Institut de Métrologie D.I. Mendéléev, Leningrad, USSR	A.A. Konstantinov, T.E. Sazonova, S.V. Sepman
IPA	Institut de Physique Atomique, Bucarest, Romania	L. Grigorescu, M. Sahagia, G. Lates
IRK	Institut für Radiumforschung und Kernphysik, Vienna, Austria	H. Friedmann, F. Hernegger, G. Winkler

Table 1 (cont'd)

		Names of the persons who carried out the measurements
LMRI	Laboratoire de Métrologie des Rayonnements Ionisants, Saclay, France	J. Bouchard, R. Vatin
NBS	National Bureau of Standards, Washington, D.C., USA	R.L. Ayres, A.T. Hirshfeld, D.D. Hoppes, L.M. Cavallo
NPL	National Physical Laboratory, Teddington, United Kingdom	M.J. Woods, Armstrong, Brown, Lucas, D. Smith, A. Parr
NPRL	National Physical Research Laboratory, Pretoria, South Africa	J. Steyn, S.M. Botha
NRC	National Research Council of Canada, Ottawa, Canada	G.C. Bowes, A.P. Baerg
OMH	Országos Mérésügyi Hivatal, Budapest, Hungary	A. Szörényi
PTB	Physikalisch-Technische Bundesanstalt, Braunschweig, German Federal Republic	K.F. Walz
SCK	Studiecentrum voor Kernenergie, Mol, Belgium	C. Ballaux, P. Willeborts
UVVVR	Ústav pro výzkum, výrobu a využití radioisotopů, Prague, ČSSR	J. Plch, J. Zderadička

Table 2

Summary of impurity determinations carried out by IER and LMRI

Identified radionuclide	Half-life (d)	IER				LMRI		
		Activity relative to ^{139}Ce		Standard error (%)	Systematic uncertainty (%)	Activity relative to ^{139}Ce	Standard error (%)	Systematic uncertainty (%)
		(for comparison with LMRI)				a) Test run		
		1975-06-01	1975-10-01			1975-10-01		
^{22}Na	950	-	-	-	-	1×10^{-8}	30	14
^{52}Mn	5.6	1.43×10^{-6}	7.3×10^{-13}	1.2	10	-	-	-
^{54}Mn	312	2.84×10^{-6}	4.00×10^{-6}	0.9	5	4.9×10^{-6}	0.3	14
^{65}Zn	244	-	-	-	-	2.6×10^{-7}	5	14
$^{114}In^{m}$	50	1.78×10^{-6}	6.1×10^{-7}	9.6	10	9.2×10^{-7}	28	14
		$T_{1/2}$ = (137.8 ± 0.2) d; three sources measured during 2.5 months.				$T_{1/2}$ = (137.59 ± 0.12) d; uncertainty at the 99.7% confidence level. Three sources were followed during one year.		
						b) Main run		
		1975-02-01	1976-02-14			1976-02-14		
^{52}Mn	5.6	1.57×10^{-6}	3.34×10^{-7}	2.2	10	3.3×10^{-7}	2.1	10
^{54}Mn	312	4.68×10^{-7}	4.85×10^{-7}	2.7	5	4.5×10^{-7}	1.6	10
^{65}Zn	244	-	-	-	-	3×10^{-8}	30	10
^{140}La	1.68	3.85×10^{-6}	1.9×10^{-8}	7.5	10	-	-	-
^{141}Ce	32.53	-	-	-	-	3.5×10^{-4}	14	3
^{143}Ce	1.38	6.48×10^{-6}	1.01×10^{-8}	5.1	15	-	-	-

Table 3

Results of adsorption tests

Laboratory	Methods 1) or 2a)	Method 2b	Further checks
AAEC	agreement within $(0.013 \pm 0.090)\%$	-	-
AECL	agreement within 0.15%	adsorption $< 0.01\%$	-
AIEA	-	adsorption = $(0.00 \pm 0.02)\%$	-
ASMW	adsorption ($\approx 0.5\%$) at pycnometer walls, after $\geqslant 6$ hours	-	-
BIPM	1st amp.: $-(0.22 \pm 0.14)\%$ 2nd " : $-(0.20 \pm 0.14)\%$	$+(0.005 \pm 0.002)\%$ $-(0.103 \pm 0.002)\%$	- -
ETL	agreement within 0.1%	-	-
IBJ	agreement within 0.013%	-	-
IER	-	$(0.33 \pm 0.39)\%$	NaI detector
IPA	$< 0.000\ 5\%$ (during dispensing)	$\leqslant 0.05\%$	-
LMRI	check carried out (no result given)	-	-
NBS	agreement within 0.08%	no detectable adsorption	-
NPL	1st amp. : + 0.11% 2nd " : - 0.07%	-	-
NPRL	agreement very good	insignificant adsorption	-
OMH	-	-	calibrated Ge(Li) detector: $< 0.000\ 4\%$
SCK	-	-	NaI detector: less than standard error

Table 4

Source preparation for 4π(PC)-γ and 4π(PPC)-γ counting

Laboratory	Source mount 1. Nature 2. Out. diam. 3. Inn. " 4. Thickness (mm)	Source backing 1. Nature 2. Number of films 3. " of met. layers 4. Total mass ($\mu g/cm^2$)	Wetting or seeding agent	1. Number of sources 2. Range of source mass (mg)	Range of N_c/N_γ (%)	Method used for varying N_c/N_γ	Balance(s) used 1. Type 2. Year of purchase 3. Date of last calibration 4. Linearity check
AAEC	1. brass 2. 35 3. 25 4. 0.05	1. VYNS, Mylar 2. 1 or 2 3. 1; 2; 4 (Au-Pd) 4. 30 to 2 050	Electrosprayed ion exchange resin; Catanac	1. 10 2. 22 to 55	16 to 59	a) thickness of backing b) counting gas c) anode voltage	1. Mettler M5, H16 2. 1963, 1962 3. Feb. 1976 4. yes, $\pm 3\ \mu g$
AECL	1. Al 2. 38 3. 25 4. 0.5	1. VYNS 2. 2 3. 2 (Au-Pd) 4. ≈ 16	Catanac SN for some	1. 16 + 15 2. 16 to 53	8 to 57	Sources prepared by precipitation (NH_3) and with or without wetting agent	1. 2 Mettler M5 2. 1958, 1971 3. Apr. 1976 for one 4. yes
AIEA	1. Al 2. 31.5 3. 19.5 4. 0.1	1. VYNS 2. 1 3. 2 4. 40	Teepol + Ludox	1. 20 weighed, 10 el. plated 2. 9 to 20	23 to 41 9 to 79	a) dissolv. of el. pl. sources in HCl b) add wett. agent and carrier, for some	1. Mettler ME22 2. 1976 3. Feb. 1976 4. -
ASMW	1. st'l. steel 2. 38 3. 16 4. 0.05	1. VYNS 2. 1 3. 1 (Au-Pd) 4. 30	Insulin, exch. resin, H_2O, NH_3 in H_2O	1. 10 2. 11 to 33	8 to 55	Superposition of gold-coated VYNS films see [38]	1. Sartorius 2405 2. 1975 3. Apr. 1976 4. yes
BARC	1. Al 2. 38 3. 28.5 4. 0.8	1. VYNS 2. 1 3. 1 4. ≈ 35	Teflon suspension	1. 30 2. 10 to 60	7 to 29	By adding solid to the source	1. Mettler M5 2. 1962 3. - 4. yes

Table 4 (cont'd)

Laboratory	Source mount 1. Nature 2. Out.diam. 3. Inn. " 4. Thickness (mm)	Source backing 1. Nature 2. Number of films 3. " of met.layers 4. Total mass ($\mu g/cm^2$)	Wetting or seeding agent	1. Number of sources 2. Range of source mass (mg)	Range of N_c/N_γ (%)	Method used for varying N_c/N_γ	Balance(s) used 1. Type 2. Year of purchase 3. Date of last calibration 4. Linearity check
BCMN	1. steel 2. 34 3. 16 4. 0.1	1. VYNS 2. 1 3. 2 (Au) 4. 50	Catanac	1. 8 2. 15	10 to 40	a) wetting agent b) VYNS c) Al	1. Mettler M5 2. 1971 3. 1972 4. -
BIPM	1. st'l.steel 2. 40 3. 16 4. 0.1	1. VYNS 2. 1 3. 1 or 2 (Au) 4. 20 to 400	Ludox SM 10^{-4}	1. 20 + 15 2. 13 to 184	15 to 47	Superposition of gold plated VYNS films	1. Mettler M5 2. 1961 3. Mar. 1976 4. yes
ETL	1. brass, Au coated 2. 30 3. 16 4. 0.3	1. VYNS 2. 1 3. 2 (Au) 4. 30	Ludox SM	1. 20 2. 8 to 20	14 to 48	a) counting gas b) gold-coated VYNS c) anode voltage	1. Mettler M5 2. 1976 3. Feb. 1976 4. yes
IEA	1. st'l.steel 2. 40 3. 20 4. 0.1	1. VYNS 2. 1 3. 1 4. $\approx$ 35	Ludox	1. 50 2. 30 to 40	14 to 44	Superposition of absorber films	1. Mettler M5 SA 2. 1967 3. Feb. 1976 4. yes
IER	1. st'l.steel 2. 40 3. 16 or 20 4. 0.1	1. VYNS 2. 1 3. 2 4. 50	Ludox SM 10^{-4}	1. 62 2. 13 to 86	14 to 45	Superposition of one gold-coated film on selected sources	1. Mettler M5 SA 2. 1964 3. Mar. 1976 4. yes

Table 4 (cont'd)

Laboratory	Source mount 1. Nature 2. Out.diam. 3. Inn. " 4. Thickness (mm)	Source backing 1. Nature 2. Number of films 3. " of met.layers 4. Total mass ($\mu g/cm^2$)	Wetting or seeding agent	1. Number of sources 2. Range of source mass (mg)	Range of N_c/N_γ (%)	Method used for varying N_c/N_γ	Balance(s) used 1. Type 2. Year of purchase 3. Date of last calibration 4. Linearity check
IMM	1. Al 2. 40 3. 22 4. 0.1	1. X-ray film Agfa 2. 2 3. 2 (Au) 4. 30 to 40	Insulin + Ludox	1. 5 2. 40 to 80	18 to 40	Discrimination	1. CMD-1000 2. 1971 3. Oct. 1975 4. -
IPA	1. Al 2. 30 3. 16 4. 0.1	1. VYNS 2. 1 3. 2 4. 50 to 150	Ludox	1. 15 2. 28 to 49	5 to 29	foil absorption	1. Mettler M5 2. 1975 3. 1976 4. yes
LMRI	1. Al 2. 38 3. 22 4. 3	1. cellulose 2. 1 3. 2 4. 100 to 160	Insulin	1. 25 2. 15 to 25	24 to 52	anode voltage + self absorption	1. Mettler ME22 2. 1975 3. - 4. yes
NBS	1. Al 2. 38 3. 17 4. 0.04	1. non-flexible collodion 2. 2 3. 1 4. 20 to 30	Ludox SM 10^4 some sources dried in H_2S atmosphere	1. 26 2. 16 to 48 dilution(DF≈5) for sources used in 4π(PPC)-γ	14 to 74 13 to 60	sandwich with 25 $\mu g/cm^2$ absorbers; discrimination	1. Mettler M5 2. 1963/4 3. 1975 4. yes
NPL	1. Al 2. 38 3. 25 4. 0.5	1. VYNS 2. 1 3. 2 (Au) 4. 50	Johnsons W.A. 0.03%	1. 10 + 10 2. 28 to 49	14 to 37	addition of carrier	1. Mettler M5 2. 1964 3. 1976 4. yes

Table 4 (cont'd)

Laboratory	Source mount 1. Nature 2. Out.diam. 3. Inn. " 4. Thickness (mm)	Source backing 1. Nature 2. Number of films 3. " of met.layers 4. Total mass ($\mu g/cm^2$)	Wetting or seeding agent	1. Number of sources 2. Range of source mass (mg)	Range of N_c/N_γ (%)	Method used for varying N_c/N_γ	Balance(s) used 1. Type 2. Year of purchase 3. Date of last calibration 4. Linearity check
NRC	1. Al 2. 38 3. 25 4. 0.8	1. VYNS 2. 1 3. 2 4. ≈ 40	Catanac SN	1. 10 + 10 2. 20 to 25	9 to 50	pulse height discrimination	1. Mettler M5 2. 1965 3. Feb. 1976 4. yes
OMH	1. Al 2. 38 3. 16 4. 0.3	1. VYNS-3 2. 1 3. 1 (Au) 4. 20	Teepol + Ludox SM	1. 24 2. 10 to 46	15 to 51	sandwich with gold-coated absorber films	1. Mettler M5 SA 2. 1968 3. 1971 4. yes
PTB	1. Al 2. 40 3. 15 4. 0.1	1. VYNS 2. 1 3. 2 (Au-Pd) 4. 45	Ludox	1. 15 2. 17 to 19	4 to 23	addition of carrier and of conductive VYNS films	1. Mettler M5 2. 1966 3. Feb. 1976 4. yes
SCK	1. Al 2. 50 3. 10 4. 0.06	1. VYNS 2. 2 3. no metal layer 4. 10	Tween 20 for some sources	1. 28 2. 4.5 to 20	11 to 32 16 to 36	sources with different amounts of carrier	1. Mettler ME22 2. 1976 3. Apr. 1976 4. yes
UVVVR	1. Al 2. 30 3. 18 4. 0.15	1. ∀YNS 2. 1 3. - 4. ≈ 40	Insulin + Ludox	1. 20 2. 19 to 25	12 to 48	variation of discrimination level	1. Sartorius 1801 2. 1972 3. Jan. 1976 4. no

Table 5

Source preparation for 4π(LS)-γ counting

Laboratory	Composition of the liquid scintillator	Intermediate solvent	1. Volume of scint. vessel (cm^3) 2. Check of adsorp. or precipitation	Range of source mass (mg)	Method used for varying N_c/N_γ	Balance used 1. Type 2. Year of purchase 3. Date of last calibration 4. Linearity check
IBJ	4 g/dm^3 PPO + 0.8 g/dm^3 bis MSB	Toluen + Triton x - 100 2:1	1. 22 2 -	19 to 28 32 to 66	variation of high voltage in β channel	1. Sartorius 1801 2. 1968 3. Jan. 1976 4. no
NPL	Unisolve 1 (Koch Light Ltd.) 6 cm^3 + saturated $Pb(NO_3)_2$ solution 0.65 cm^3	-	1. 10 2. yes $\leq$0.2%	15 to 30	computer see [12]	1. Mettler M5 2. 1964 3. 1976 4. yes
NPRL	Xylene-based scintillator "Instagel" (Packard-Corp.)	-	1. 20 2. yes	32 to 59	pulse height selection	1. Mettler ME22 2. 1975 3. - 4. yes correction: + 20 μg

Table 6

Equipment for 4π(LS)-γ counting

Laboratory	Number of phototubes for viewing the counting cell	1. Material of counting cell 2. Type of photomultiplier	1. Precautions taken against counting of spurious pulses 2. Upper limit of sp. pulses	Gamma-ray counter 1. Number of NaI(Tl)crystals 2. Diameter (mm) 3. Height (mm)
IBJ	2 in coincidence [39]	1. low potassium glass 2. EMI 9634 QR	1. - 2. -	1. one 2. 45 3. 50
NPL	one	1. glass 2. RCA 31000 D	1. correlation counting 2. 0.1%	1. one (well-type) 2. 100 3. 100
NPRL	two, in coincidence	1. glass 2. EMI 9635 QB	1. separate determination 2. (0 $\pm$ 0.1) %	1. one 2. 76 3. 76

Table 7

Equipment for 4π(PC)-γ and 4π(PPC)-γ counting

Laboratory	4 pi proportional counter								Gamma-ray counter		
	Wall material	Height of each half (mm)	Anode 1. Nature 2. Wire diam. (μm) 3. " length (mm)		4. Distance from the source (mm) 5. Voltage (kV)		Gas 1. Nature* 2. Discr. level (eV) 3. Pressure (kPa)		1. Number of NaI(Tl) crystals 2. Diameter (mm) 3. Height (mm)		
AAEC	Al	27	1. Pt 2. 50	3. 48 4. 25	5. 2.750 to 2.900 1.750 to 1.925		1. CH_4 Ar/CH_4	2. 300 3. atm.	1. 1	2. 76	3. 25
AECL	stainless steel	21	1. st'l. steel 2. 15	3. 36 4. 10	5. 2.4		1. CH_4 2. $\approx$ 100	3. atm.	1. 2	2. 76	3. 76
	2 independent sets of electronics following the linear stages; circuitry to set and continuously monitor interchannel delays; 36-sample automatic changer [41]										
AIEA	stainless steel	12	1. W 2. 25	3. 35 4. 8	5. 1.9 to 2.1		1. CH_4 2. 170 or 700	3. atm.	1. 1	2. 76	3. 76
ASMW	brass (+ Al) [40]	20	1. Mo(Au-coated) 2. 40	3. 55 4. 10	5. 3.9		1. C_3H_8 2. 250	3. atm.	1. 2	2. 102	3. 76
BARC	Al	26	1. st'l. steel 2. 13	3. 38 4. 13	5. 1.5		1. Ar/CH_4 2. 200	3. atm.	1. 1	2. 76	3. 76
BCMN	plexiglas + Al	14	1. steel 2. 50	3. 75.5 4. 10	5. 2.1		1. Ar/CH_4 2. -	3. atm.	1. 2	2. 76	3. 51
BIPM	brass Au-plated	20	1. st'l. steel 2. 50	3. 47 4. 11	5. 2.3 [20]		1. Ar/CH_4 2. $\approx$ 75	3. atm.	1. 1	2. 76	3. 51

* Ar/CH_4 stands for 90% Ar + 10% CH_4

Table 7 (cont'd)

Laboratory	4 pi proportional counter									Gamma-ray counter
	Wall material	Height of each half (mm)	Anode				Gas			1. Number of NaI(Tl) crystals 2. Diameter (mm) 3. Height (mm)
			1. Nature 2. Wire diam. (μm) 3. " length (mm)		4. Distance from the source (mm) 5. Voltage (kV)		1. Nature* 2. Discr. level (eV) 3. Pressure (kPa)			
ETL	brass Au-coated	20	1. st'l. steel 2. 50	3. 80 4. 10	5. 2.3 3.6		1. Ar/CH_4 CH_4	2. 200 3. atm.		1. 2 2. 76 3. 76
IEA	brass	22.5	1. st'l. steel 2. 20	3. 120 4. 13	5. 1.6		1. Ar/CH_4 2. 50	3. atm.		1. 2 2. 76 3. 76
IER	Al	25	1. Au 2. 100	3. 34 4. 12.5	5. 3.5 to 3.6		1. CH_4 2. $\approx$ 1 000	3. atm.		1. 1 2. 76 3. 76
IMM	brass	30	1. Constantan 2. 30	3. 50 4. 15	5. $\approx$ 2		1. Ar/CH_4 2. 200	3. atm.		1. 1 2. 40 3. 30
IPA	brass	24	1. W 2. 20	3. 40 4. 11	5. 3.2		1. CH_4 2. 1 000	3. atm.		1. 1 2. 76 3. 76
LMRI	perspex	22	1. W + Au 2. 20	3. 80 4. 10	5. 1.8		1. Ar/CH_4 2. $\approx$ 100	3. atm.		1. 1, with Be window Ø 48.5 x 0.24 mm 2. 44 3. 3
NBS	stainless steel	27	1. st'l.steel 2. 25.4	3. 38.1 4. 18.4	5. 2.05		1. Ar/CH_4 2. 750	3. atm.		1. 2, 180° opposed 2. 76 3. 76
	Al-6061	28.3	1. st'l. steel 2. 51	3. $\approx$ 53 4. 14	5. 8.0		1. Ar/CH_4 2. 1 000 to 20 000	3. 1 430		

* Ar/CH_4 stands for 90% Ar + 10% CH_4

Table 7 (cont'd)

Laboratory	4 pi proportional counter						Gamma-ray counter
	Wall material	Height of each half (mm)	Anode 1. Nature 2. Wire diam. (μm) 3. " length (mm) 4. Distance from the source (mm) 5. Voltage (kV)			Gas 1. Nature* 2. Discr. level (eV) 3. Pressure (kPa)	1. Number of NaI(Tl) crystals 2. Diameter (mm) 3. Height (mm)
NPL	Cu and perspex (Ag-coated)	14	1. P-bronze 2. 76	3. 75 4. 8	5. 2.1	1. Ar/CH_4 3. atm. 2. 300	1. 2 2. 102 3. 76
NRC	Al	25	1. st'l. steel 2. 25	3. 38 4. 12.7	5. ≈ 4.8	1. Ar/CH_4 3. 1 584 2. 600 to 6 000	1. 2 2. 76 3. 76
	2 independent coincidence counting systems used alternatively [25, 42]; live-timed anti-coincidence counting system [24]						
OMH	plexiglas Au-coated	24	1. W 2. 11	3. 45 4. 12	5. 1.55	1. Ar/CH_4 3. atm. 2. 180	1. 1 2. 76 3. 76
PTB	Al	22.5	1. st'l. steel 2. 50	3. 30 4. 12	5. 3.7	1. CH_4 3. atm. 2. 500	1. 1 2. 76 3. 76
SCK	perspex Au-coated	17	1. Ni, 5 wires per half-counter 2. 50 3. 50-64 4. 9 5. 2.0			1. Ar/CH_4 3. atm. 2. ≈ 40	1. 1 2. 76 3. 76
	stainless steel	50	1. Ni 2. 50	3. 50 4. 25	5. 5.7	1. Ar/CH_4 3. 540 2. -	
UVVVR	stainless steel	96 diameter	1. Mo (Au-coated) 2. 50	3. 140 4. 24	5. 4.05	1. Ar/CH_4 3. ≈ 500 2. 200	1. 2 2. 76 3. 51 [43]

* Ar/CH_4 stands for 90% Ar + 10% CH_4

Table 8

Dead times and coincidence resolving times (in parenthesis: uncertainty in units of last decimal)

Laboratory	Dead times τ_β (μs)	τ_γ (μs)	Method of measurement*	Resolving time** τ_r (μs)	Method of measurement*	Remarks and references
AAEC	9.00 (5)	20.0 (2)	DP	1.175 (1)	RC (TS, SP)	
AECL	2.049 (6) 2.020 (6)	2.043 (6) 2.016 (6)	SP	0.643 0 (3) 0.664 5 (3)	SP	checked with calibrated oscilloscope
AIEA	4.087 (12)	4.085 (12)	TO	0.950 7 (42)	TO	[44]
ASMW	4.008 (18)	4.000 (18)	DP, TO, TS	1.046 (6)	RC [45]	τ_β, τ_γ checked before and after each counting process
BARC	10 (1)	10 (1)	TS	1.8 (2)	RC (^{137}Cs)	
BCMN	≈7	≈7	TO	0.99 (1)	TO	[46]
BIPM	4.43 (1)	4.48 (1)	TO	1.05 (1)	TO	
ETL	4.35 (5)	2.11 (5)	DP	0.688 7 (28)	RC	β source, γ pulser
IBJ	7.500 (25) 7.820 (25)	7.800 (25)	DP	0.270 (10)	DP	calibrated by synchronoscope
IEA	3.01 (2)	3.01 (2)	SP, TO	1.04 (1)	RC	[47]

Abbreviations: DP = double-pulse generator, RC = random coincidences, SP = source-pulser method, TO = two-oscillator method, TS = two-source method

* For general information on recent measuring methods see a) for SP: [47, 49], b) for TO: [50, 51, 52]

** $\tau_r = \frac{1}{2}(\theta_\beta + \theta_\gamma)$, see p. 12

Table 8 (cont'd)

Laboratory	Dead times τ_β (μs)		τ_γ (μs)		Method of measurement*	Resolving time** τ_r (μs)		Method of measurement*	Remarks and references
IER	2.201	(1)	2.196	(1)	TO	0.775	(2)	TO	[48]
IMM	1.36	(2)	1.36	(2)	SP	1.475 2.080	(10) (10)	TS, TO	
IPA	10.0	(5)	10.0	(5)	TS	1.095	(5)	RC	
LMRI	5.200	(25)	5.200	(25)	-	0.973	(2)	SP	variable delays
NBS	5.12	(49)	5.24	(20)	SP	0.47	(2)	SP	4π(PC)-γ
						set with calibr. oscilloscope			
	20.0	(12)	20.0	(12)	SP	0.60	(5)	SP	4π (PPC)-γ
NPL	1.527	(5)	3.0 oscilloscope	(5)	τ_β: DP	0.716	(5)	TO	4π (PC)-γ
	24.6	(1)	1.96	(5)	τ_β: DP	0.25	(1)		4π (LS)-γ
	τ_γ: add to real γ spectrum, at preamplifier, pulses of correct size, observe fraction lost					pulse widths into AND gate			
	8.00	(1)	6.01	(2)		2.95	(5)		4πSi(Li)-γ
	using time interval averager								
NPRL	1.25	(2)	3.23	(1)	TS	0.502 0.510 0.510	(3) (3) (3)	RC	three units in parallel

Abbreviations: DP = double-pulse generator, RC = random coincidences, SP = source-pulser method, TO = two-oscillator method, TS = two-source method

* For general information on recent measuring methods see a) for SP: [47, 49], b) for TO: [50, 51, 52]

** $\tau_r = \frac{1}{2}(\theta_\beta + \theta_\gamma)$, see p. 12

Table 8 (cont'd)

Laboratory	Dead times τ_β (μs)		τ_γ (μs)		Method of measurement*	Resolving time** τ_r (μs)		Method of measurement*	Remarks and references
NRC	2.03	(2)	2.04	(2)	SP	0.982	(1)	SP	system 1
	2.12	(2)	2.10	(2)	SP	0.976	(1)	SP	system 2
	5.4	($\approx$1)	2.07	(2)	DP				anticoincidence
OMH	4.99	(10)	4.93	(10)	TO	1.032	(15)	TO	
PTB	2.98	(3)	3.00	(3)	TO	2.87	(3)	TO	
SCK	15.0	(5)	2.2	(1)	116_1m1	2.0	(1)	N_c/N_γ vs. delay	4π(PC)-γ, 4π(PPC)-γ
UVVVR	6.57	(5)	4.12	(5)	[43]	1.980	(8)	TS	
	study of time-interval distribution								

Abbreviations: DP = double-pulse generator, RC = random coincidences, SP = source-pulser method, TO = two-oscillator method, TS = two-source method

* For general information on recent measuring methods see a) for SP: [47, 49], b) for TO: [50, 51, 52]

** $\tau_r = \frac{1}{2}(\theta_\beta + \theta_\gamma)$, see p. 12

Table 9

Coincidence counting data

Laboratory	γ channel (keV) from	to	Background rates (s^{-1}) β	γ	c	Number of sources measured with variation of N_c/N_γ	without variation of N_c/N_γ	Number of data points used in slope det.	Mean time for one data point (s)	Correction for Gandy effect (%)	Time of the measurements from	to	Uncertainty in $\frac{1 - N_c/N_\gamma}{N_c/N_\gamma}$ accounted for ?	% change in a_o
AAEC	161	171	1.2 to 1.8	0.24	0.001	4	6	15 see [53]	11 000	0	Apr. 8	Apr.30	yes	0.003 3
AECL	≈125	200	0.3	2.0	0.006	16	-	210	1 000	0 to 0.080	Apr. 5	Apr. 9	no	
	120	200	0.3	2.0	0.006	15	-	180	860	0 to 0.005	Apr.26	Apr.29	no	
AIEA	60	240	1	9	0.05	10	20	62	2 000	0	Mar.20	May 15	no	
ASMW	100	200	1.9	7.6	0.008	9	-	24	2 000	0	Apr.19	Apr.23	yes	0.045
BARC	-	-	0.5	1.7	0.002	4	29	58 see [54]	4 000	-	Mar. 5	May 26	no	
BCMN	-	-	0.4	4	0.01	8	-	64	1 000	-	Mar.	Mar.	-	
BIPM	140	190	0.5	1.4	0.007	6	4	27	3 000	0	Mar.24	Mar.30	no	
ETL	130	200	0.8	3.6	0.001 2	10	10	39	1 500	0	Mar.14	Mar.23	-	
IBJ	* 65	∞	3.7	9.7	0.35	10	-	12	600	0	Mar.15	Mar.16	no	
	95	235	3.8	4.8	0.07	6	-	9	300	0	Jun.30	Jul. 2	no	
	*125	235	3.6	3.8	0.06	9	-	9	300	0	Jun.29	Jun.30	no	

* The results obtained with these γ-channel settings were found to be low and high, respectively, by about 2%, due to incorrect coincidence count rates. They were therefore discarded.

Table 9 (cont'd)

Laboratory	γ channel (keV)		Background rates. (s^{-1})			Number of sources measured with / without variation of N_c/N_γ		Number of data points used in slope det.	Mean time for one data point (s)	Correction for Gandy effect (%)	Time of the measurements		Uncertainty in $\frac{1 - N_c/N_\gamma}{N_c/N_\gamma}$ accounted for ? % change in a_o
	from	to	β	γ	c	with	without				from	to	
IEA	148	200	2.0	4.3	0.019	3	50	25	1 800	1	Apr.20	May 20	-
IER	142	193	≤2	≤9.8	≤0.009	8	46	16	2 036	0	Mar.25	Apr. 6	yes 0.005 6
IMM	a wide gate		2	18	0.2	5	-	17	1 000	0	Mar.16	Apr.23	-
IPA	130	200	2.1	1.9	0.029	4	11	19	(1 to 3.6) $\times 10^3$	0	May 20	Jun.18	no
LMRI	136	216	2.5	0.2	0.02	24	-	120	600	-	May 25	Jun. 2	yes 0.016
NBS	128	202	0.4	4.9	0.002	11	-	37	3 600	0	Apr. 2	Apr. 8	no
	113	218	≤1.9	5.5	0.02	4	-	42 see [55]	2 300	0	Mar.24	Apr. 8	no
NPL	100	300	2.2	4.8	0.13	PC 9	1	63	1 000	0	Mar.25	Apr. 1	no
	100	300 photopeak only, and				PC 8	2	40	1 000	0	Apr. 6	Apr. 9	no
	50	100	3.0	1.2	0.02	LS 9	-	30	(1 to 2)$\times 10^3$	0	Mar.25	Mar.26	yes -
	140 and 100	190 230	1.3	1.0	0.03	SiLi7	-	23	1 000	0	Mar.30	Apr. 9	yes -

Table 9 (cont'd)

Laboratory	γ channel (keV) from	to	Background rates (s^{-1}) β	γ	c	Number of sources measured with variation of N_c/N_γ	without variation of N_c/N_γ	Number of data points used in slope det.	Mean time for one data point (s)	Correction for Gandy effect (%)	Time of the measurements from	to	Uncertainty in $\frac{1 - N_c/N_\gamma}{N_c/N_\gamma}$ accounted for ?	% change in a_o
NPRL	photopeak		≤8	3.4	-	16	-	15	800	0	Mar.18	Mar.23	yes	0.01
NRC	130	200	0.8	1.7	0.004	10	-	15	500	0	Mar.15	Mar.20	yes	<0.1
						10	-	15	500	0	Mar.22	Mar.27		
	130	200	0.5	1.6	0.005	10	-	15	500	0	Mar.15	Mar.20		
	130	200	0.5	1.6	1.6	AC 10	-	15	≈530	0	Mar.20	Mar.27		
OMH	140	193	2.1	5.1	0.01	3	24	9	2 000	0	May 25	May 28	no	
PTB	130	∞	0.5	16	0.2	8	7	53	4 000	+ 0.01	May 3	May 15	no	
SCK	140	210	36	1.4	0.05	PC -	25	25	400	0	Apr.16	Apr.30	no	
			12	11	0.056	PPC-	20	18	800	0	Apr.17	Apr.22	no	
UVVVR	90	230	19	7.1	0.09	2	19	20	1 000	0	Apr. 4	Apr.15	no	

Table 10

Results of the χ^2 tests for the efficiency functions (ν is the number of degrees of freedom)

Laboratory	Counting method	χ^2/ν
AECL	4π(PC)-γ	1.8 1.8
BIPM	"	0.71 1.02
IER	"	0.95
IMM	"	-
IPA	"	1.2
LMRI	"	0.6
NPL	4π(PC)-γ	1.27 0.19
	4π(LS)-γ	1.2 —— 2.9
	4πSi(Li)-γ	1.5 —— 5.0
NPRL	4π(LS)-γ	0.85
NRC	4π(PPC)-γ coinc.	0.97
	" anticoinc.	1.13

Table 11

Final results and uncertainties

Laboratory	Method used*	Ampoule numbers	Slope-to-intercept ratio (and uncertainty)	Radioactive concentration 1976-03-15 0 h UT ($Bq \cdot mg^{-1}$)	Standard error of the mean ($Bq \cdot mg^{-1}$)	Standard error of the mean (%)	Number of degrees of freedom	Total systematic uncertainty (see also Table 12) ($Bq \cdot mg^{-1}$)	Total systematic uncertainty (see also Table 12) (%)
AAEC	4π(PC)-γ	2 42	0.199 18 (18)	710.24	0.82	0.12	13	1.78	0.25
AECL	4π(PC)-γ	11	0.200 32 (4)	710.55	0.09	0.013	208	0.34	0.048
		49	0.200 39 (7)	710.46	0.12	0.017	178	0.61	0.086
AIEA	4π(PC)-γ	33 72	0.198 72 (25)	711.59	0.33	0.05	-	0.71	0.1
ASMW	4π(PC)-γ	13 51	0.201 64 (7)	711.1	0.7	0.1	9	1.9	0.262
BARC	4π(PC)-γ	18 56	0.197 0 (4)	712.2	1.1	0.15	56	3.8	0.53
BCMN	4π(PC)-γ	7 46	0.201 4 (6)	709.98	0.21	0.03	>10	4.62	0.65
BIPM	4π(PC)-γ	15	0.203 3 (6)	710.48	1.35	0.19	25	8.31	1.17
		53	0.202 9 (2)	711.08	0.52	0.07	31	4.05	0.57
ETL	4π(PC)-γ	19 57	0.201 4 (6)	710.37	0.96	0.14	37	3.48	0.49
IBJ	4π(LS)-γ	22 60	0.191 8 (24)	712.6	2.5	0.35	5	5.0	0.70
IEA	4π(PC)-γ	8 47	0.206 7 (7)	708.5	0.3	0.04	44	1.5	0.21
IER	4π(PC)-γ	27 65	0.200 36 (16)	709.86	0.08	0.012	45	2.13	0.3

Table 11 (cont'd)

Laboratory	Method used*	Ampoule numbers	Slope-to-intercept ratio (and uncertainty)	Radioactive concentration 1976-03-15 0 h UT ($Bq \cdot mg^{-1}$)	Standard error of the mean ($Bq \cdot mg^{-1}$)	Standard error of the mean (%)	Number of degrees of freedom	Total systematic uncertainty (see also Table 12) ($Bq \cdot mg^{-1}$)	Total systematic uncertainty (see also Table 12) (%)
IMM	4π(PC)-γ	30 67	0.189 4 (6)	710.47	0.71	0.1	3	$^{+0.78}_{-1.07}$	$^{+0.11}_{-0.15}$
	XK(NaI)-γ		-	709.01	0.35	0.05	12	4.96	0.7
	2π XL(PC)-γ		-	708.05	0.35	0.05	12	5.67	0.8
IPA	4π(PC)-γ	23 61	0.198 72 (53)	714.47	0.27	0.038	17	2.63	0.368
IRK	4π(NaI)γ	3 43	-	715.0	0.4	0.06	7	4.0	0.56
LMRI	4π(PC)-γ	14 52	0.204 5 (40)	708.4	0.9	0.13	117	0.7	0.092
NBS	4π(PPC)-γ	28	0.202 8 (1)	713.73	0.16	0.02	41	3.07	0.43
	4π(PC)-γ	66	0.198 0 (2)	712.86	0.26	0.04	36	3.42	0.48
NPL	4π(PC)-γ	16	0.200 4 (7)	710.1	0.9	0.13	62	3.1	0.43
	4π(PC)-γ	54	0.200 2 (3)	711.6	0.4	0.06	38	3.5	0.49
	4π(LS)-γ	16	-	711.45	0.65	0.1	18	11.4	1.6
	4πSi(Li)-γ	16	-	709.58	2.31	0.32	6	9.3	1.3
NPRL	4π(LS)-γ	24 62	(0.248 3) (5)	(719.8)	0.4	0.06	15	(1.1)	(0.15)
			0.245 1	710.4				3.5	0.5
NRC	4π(PPC)-γ 1	48	0.203 0 (4)	711.9	0.2	0.03	9	<1.3	<0.18
	4π(PPC)-γ 1	9	0.202 9 (4)	711.7	0.2	0.03	9	<1.3	<0.18
	4π(PPC)-γ 2	48	0.203 9 (3)	711.4	0.2	0.03	9	<1.3	<0.18
	4π(PPC)-γ AC	48	0.202 9 (3)	711.5	0.2	0.03	9	<1.0	<0.14

Table 11 (cont'd)

Laboratory	Method used*	Ampoule numbers		Slope-to-intercept ratio (and uncertainty)		Radioactive concentration 1976-03-15 0 h UT ($Bq \cdot mg^{-1}$)	Standard error of the mean ($Bq \cdot mg^{-1}$)	(%)	Number of degrees of freedom	Total systematic uncertainty (see also Table 12) ($Bq \cdot mg^{-1}$)	(%)
OMH	$4\overline{\pi}$ (PC)-γ	17	55	0.199 7	(45)	710.7	0.4	0.05	17	5.2	0.73
PTB	$4\overline{\pi}$ (PC)-γ	4	44	0.200 04	(3)	710.42	0.24	0.05	51	1.1	0.15
UVVVR	4π (PPC)-γ	12	50	0.200	(2)	715.85	0.24	0.04	18	5.37	0.75

* PC : proportional counter
PPC : pressurized proportional counter
LS : liquid scintillator
AC : anti-coincidence

the hyphen indicates the use of a coincidence technique

Table 12

Systematic uncertainty of the final result

Laboratory	% uncertainty due to 1) weighing, 2) dead time, 3) resolving time, 4) background, 5) extrapolation, 6) others, 7) difference between intercepts for 1st and 2nd order fits	How was it obtained?
AAEC	1) ≲0.025	$\Delta m = 5\ \mu g$.
	2) ≲0.05	Uncertainty in time measurement.
	3) 0.005	
	4) -	
	5) 0.15, see [56]	Compar. with earlier measurements.
	6) 0.01	
	7) 0.17	
AECL	1) 0.005	Diff. 2 balances + uncert. of buoyancy corr.
	2) 0.007	By setting each parameter separately to its extreme value.
	3) 0.001	
	4) 0.003	From 5% background variation observed.
	5) 0.028 0.066	Diff. betw. intercepts for 1st and 2nd order fits.
	6) 0.004	Extreme delay mismatch = 10 ns.
	7) 0.028	
AIEA	1) 0.05	Δm
	2) -	large number of measurements; no bias expected.
	3) -	
	4) -	
	5) 0.05	-
	6) 0.05	General uneasiness.
	7) ≈0.01	
ASMW	1) 0.08	$\Delta m = \pm 2\ \mu g$, tested at diff. balances.
	2) 0.004	2% of correction.
	3) 0.06	
	4) 0.01	3% of corr. for γ background.
	5) 0.09	Estim. from slope and error in x.
	6) 0.018	$\Delta T_{1/2}$ and timing.
	7) 0	
BARC	1) 0.1	$\Delta m = 20\ \mu g$.
	2) 0.01	
	3) 0.08	Considering extreme values.
	4) 0.06	
	5) 0.27	Comparing different sets of data.
	6) 0.01	Timing.
	7) 0.58	

Table 12 (cont'd)

Laboratory	% uncertainty due to 1) weighing, 2) dead time, 3) resolving time, 4) background, 5) extrapolation, 6) others, 7) difference between intercepts for 1st and 2nd order fits	How was it obtained?
BCMN	1) 0.05	-
	2) 0.1	see [46].
	3) 0.1	
	4) -	-
	5) 0.4	see [57].
	6) -	-
	7) 0.55 for 4 sources out of 8.	
BIPM	1) 0.02	$\Delta m = 10\ \mu g$.
	2) 0.006	$\Delta\tau' \cdot N_c/(1 - \tau' N_c)$.
	3) 0.15	$(\Delta\tau_r/\tau_r) \cdot (N_{acc}/N_c)$.
	4) 0.001	$(\varepsilon_\gamma \cdot N_o)^{-1} \cdot (B_\gamma/t)^{1/2}$.
	5) 0.69	Diff. 1st and 2nd order fits.
	6) -	
	7) 0.69	
ETL	1) 0.05	$\Delta m = 10\ \mu g$.
	2) 0.04	$2\,\Delta\tau_\beta \cdot N_{\beta\,max}$.
	3) 0.015	$3\,\Delta\tau_r \cdot N_\beta N_\gamma/N_c$.
	4) 0.003	$\Delta B_\gamma/N_\gamma$.
	5) 0.3	Diff. 1st and 2nd order fits.
	6) 0.05	Delay variation.
	7) 0.3	
IBJ	1) <0.088	$\Delta m/m$
	2) <0.002	$\Delta\tau' N_c'/t$.
	3) <0.087	$(\Delta\tau_r/t)\,(N_\beta' + N_\gamma' - (2 N_\beta' N_\gamma'/N_c))$.
	4) <0.01	$\Delta B/N'$.
	5) -	
	6) $<0.000\ 1$	Timing.
	7) 0.69	
IEA	1) 0.04	(States a total syst. uncertainty of 0.21%.)
	2) 0.001	
	3) 0.01	
	4) 0.015	
	5) -	
	6) -	
	7) 0.59	

Table 12 (cont'd)

Laboratory	% uncertainty due to 1) weighing, 2) dead time, 3) resolving time, 4) background, 5) extrapolation, 6) others, 7) difference between intercepts for 1st and 2nd order fits	How was it obtained?
IER	1) 0.03	$\Delta m/\overline{m}$.
	2) 0.003	$\Delta\tau' N_\beta/(1-\tau' N_{\beta\,max})$.
	3) 0.02	$(\Delta\tau_r/\tau_r)(N_{acc}/N_{c\,max})$.
	4) 0.04	$\sigma_{z\gamma}/\overline{N}_\gamma$.
	5) 0.2	Using extreme slope and $\overline{x} = 1.5$.
	6) -	
	7) 0.023	
IMM	4πPC-γ / KX(NaI)-γ / 2πPC LX-γ	
	1) 0.05 / 0.05 / 0.05	
	2) 0.005 / 0.005 / 0.005	
	3) 0.01 / 0.02 / 0.02	
	4) <0.005 / / <0.005	
	5) 0.1	
	6) -0.1 / 0.05 / 0.05	Afterpulses.
	0.07 / 0.8	Constants.
	7) -	
IPA	1) 0.05	Δm, evaporation.
	2) 0.015	
	3) 0.02	Error propagation.
	4) 0.003	
	5) 0.11	
	6) 0.17	Spurious pulses, adsorption, $T_{1/2}$, timing.
	7) 0.20	
IRK	1) 0.082	
	2) 0.015	
	6) 0.041	Timing, $T_{1/2}$.
LMRI	1) 0.04	(States a total syst. uncertainty of 0.092%;
	2) 0.001	(we add 7) and use 0.95% in Fig. 4).)
	3) 0.02	
	4) 0.001	
	5) -	
	6) 0.03	Timing, $T_{1/2}$.
	7) 0.86	

Table 12 (cont'd)

Laboratory	% uncertainty due to 1) weighing, 2) dead time, 3) resolving time, 4) background, 5) extrapolation, 6) others, 7) difference between intercepts for 1st and 2nd order fits	How was it obtained?
NBS	4πPPC-γ / 4πPC-γ	
	1) 0.01 / 0.01	Estimated from past results.
	2) 0.05 / 0.06	From measured uncertainty at 99% confidence level.
	3) 0.10 / 0.04	
	4) 0.02 / 0.02	From variations observed.
	5) 0.10 / 0.20	Max. range 1st through 3rd order fits.
	6) 0.15 / 0.15	Gandy effect, $T_{1/2}$, dilution.
	7) 0.019 / 0.003	
NPL	4πPC-γ / 4πLS-γ / 4πSi(Li)-γ	
	1) 0.02 / 0.02 / 0.02	From uncert. of total mass of 10 sources.
	2) 0.006 / 0.04 / 0.002	$\Delta\tau'$.
	3) 0.02 / 0.02 / 0.05	$\Delta\tau_r$.
	4) 0.01 / / 0.01	Δ B.
	5) 0.42(0.35) / 1.5 / 1.2	Diff. between 1st and 2nd order fits.
	6) 0.02 / / 0.02	Contamination by ^{141}Ce.
	7) 0.42(0.35) / /	
NPRL	1) 0.01	Manufacturer's estimate.
	2) 0.002	Estimated from inaccuracy in the measurements of these parameters.
	3) 0.03	
	4) 0.003	
	5) 0.1	By using different channel settings.
	6) 0.3	Count rate dependence (+ spurious pulses).
	7) 0.05	
NRC	4πPPC-γ / AC	4πPPC-γ / AC
	1) <0.03 / <0.03	$\Delta m/m$ Balance calibration.
	2) <0.01 / -	$m\Delta\tau'(a_o - (a_1 N_c^2)/(N_\beta N_\gamma))$. / -
	3) <0.03 / -	$a_1 m (2 - N_c/N_\gamma - N_c/N_\beta)\cdot\Delta\tau_r$. / -
	4) <0.01 / <0.01	$m a_1 N_c \Delta B_\gamma/(N_\beta N_\gamma^2)$ / $m a_1 (N_\gamma - Y)\Delta B_\gamma/(N_\beta N_\gamma^2)$
	5) <0.1 / <0.1	Max. standard error on N_o.
	6) - / <0.005	- / live-time.
	7) -	
OMH	1) 0.04	By calculating the maximum values.
	2) 0.01	
	3) 0.05	
	4) 0.01	
	5) 0.60	
	6) 0.02	
	7) 0.47	

Table 12 (cont'd)

Laboratory	% uncertainty due to 1) weighing, 2) dead time, 3) resolving time, 4) background, 5) extrapolation, 6) others, 7) difference between intercepts for 1st and 2nd order fits	How was it obtained?
PTB	1) 0.02	$2\Delta m/m$, $\Delta m = 2\,\mu g$.
	2) <0.01	-
	3) 0.03	$(3\Delta\tau_r/\tau_r)\cdot N_{acc}/N_c$.
	4) 0.02	$3\Delta B_\gamma/N_\gamma$.
	5) 0.05	Spread of individual slopes.
	6) 0.04	$\Delta T_{1/2} = 0.07$ d.
	7) 0.058	
SCK	4πPC-γ / 4πPPC-γ / 4π(CsI)γ	
	1) ≤0.2 / ≤0.2 / ≤0.2	4π(CsI)γ Experimental.
	2) 0.001 / 0.003 / <0.1	See [58].
	3) 0.04 / 0.06 / -	
	4) - / - / -	
	5) 0.5 / 0.5 / 0.5	See [25]. Experimental.
	6) - / - / 0.6	Statistics, geometry, decay-scheme corr.
	7) 0.73 / 0.57 / -	
UVVVR	1) 0.07	$\Delta m = 15\,\mu g$.
	2) 0.04	Max. 150 ns.
	3) 0.02	Max. 25 ns.
	4) 0.1	Mainly γ channel.
	5) 0.5	Max. $a_1 = 0.002$; $a_2 = 0.000\,1$
	6) 0.02	Max. $\pm$ 5 h.
	7) 0.6	

Symbols used in this table

a_o, a_1, a_2	See reporting Form, Fig. 1	t	Measuring time
B	Stands for B_β or B_γ	$T_{1/2}$	Half-Life of ^{139}Ce
B_β, B_γ	Background rates	x, y	See reporting Form
m	Source mass	Δ	Uncertainty of ...
N_o	Disintegration rate	ε_γ	Efficiency of γ detector
N_β, N_γ, N_c	Count rates, corrected for background	$\sigma_{z\gamma}$	Standard deviation of 21 measurements of B_γ
N'_β, N'_γ	Count rates, uncorrected	τ'	The shorter of τ_β or τ_γ
N'	Stands for N'_β or N'_γ	$\tau_\beta(\tau_\gamma)$	Dead time in $\beta(\gamma)$ channel
N_{acc}	Rate of accidental coinc.	τ_r	Coincidence resolving time

BUREAU INTERNATIONAL DES POIDS ET MESURES

International comparison of a solution of ^{139}Ce

Participating Laboratory: $T_{1/2} = (137.65 \pm 0.07)$ d

Source preparation

for 4π(PC)-γ counting

Source mount
- Nature . . . . . .
- Outer diameter . . . . . mm
- Inner diameter . . . . . mm
- Thickness . . . . . mm

Backing
- Nature . . . . . .
- Number of films per source . . . . . .
- Number of metal layers per source . . . . . .
- Total mass per cm^2 . . . . . $\mu g\ cm^{-2}$

Wetting or seeding agent . . . . . .

Range of source mass (. . . . . to . . . . .) mg

Number of sources prepared . . . . . .

for 4π(LS)-γ counting

Range of source mass: (. . . . . to . . . . .) m

Volume of scintillation vessel . . . cm^3

Intermediate solvent . . . . . .

Composition of liquid scintillator

. . . . . .

. . . . . .

. . . . . .

Was possible adsorption or precipitation of active material in the counting cells checked? . . . . . .

Were checks made to see that the actual liquid used in the source dispensing was representative of the solution, e.g. by preparing ampoules for ion-chamber measurement from the solution in the pycnometer before and after dispensing the sources ? . . . . . .

If so, how good was the agreement ? . . . . . .

Result of an eventual adsorption test . . . . . .

Balance used - Type . . . . . Has the optical scale been checked for linearity ? . . . . . .

Year of purchase . . . . . .

Date of last weight calibration . . . . . .

Was a buoyancy correction applied ? . . . . . If so, of how much ? . . . .

Counting equipment

4π proportional counter

Wall material . . . . . .

Height of each half . . . . . mm

Anode
- Nature . . . . . .
- Wire diameter . . . . . μm
- Wire length . . . . . mm
- Distance from source . . . . . mm
- Voltage . . . . . kV

Gas
- Nature . . . . . .
- Pressure . . . . . .
- Discrimination level . . . . . eV

Liquid scintillation counter

Material and volume of counting cell . . . . .

Number of photomultipliers used for viewing the cell . . . . . .

Type of photomultiplier . . . . . .

If two phototubes were used, were they used in coincidence ..., summation ..., or both?.

For one tube only, or two in summation, were precautions taken against counting of spurious pulses? . . . . . Describe how . . . . . .

Upper limit of percentage of sp. pulses . . . . . .

Gamma ray counter - Number and nature of crystals . . . . . .

Diameter . . . . . mm

Height . . . . . mm

Dead times and their uncertainties (standard errors) $\tau_\beta = (. . . . . \pm . . . . .) \mu s$

Explain how they were determined $\tau_\gamma = (. . . . . \pm . . . . .) \mu s$

. . . .

Please provide a block diagram of your counting set and eventual references as to published papers, internal reports, etc. (on a separate sheet). .../...

Figure 1 - Specimen of the reporting form

Coincidence resolving time τ_r ($\approx \frac{1}{2} \frac{n_{acc}}{n_\beta \cdot n_\gamma}$) = (..... ± .....) μs
Explain how it was measured
....
....
....

"Gandy effect":
Mean interval between arrival times of γ and β pulses at coincidence mixer ..... μs
(positive, if γ pulse arrives first). By how much did you correct the final result ? ..... %

Counting data

γ channel setting .....

Background rates: B_β = ... s^{-1}, B_γ = ... s^{-1}, B_c = ... s^{-1}

Number of sources measured { with variation of N_c/N_γ (for slope determination) ..... ; without variation of N_c/N_γ ..... }

Number of data points used in slope determination .....

Mean measurement time for one data point ..... s

Range of N_c/N_γ ..... to .....

Procedure used to vary N_c/N_γ .....
.....

Slope/intercept ratio (with standard error) ..... ± .....

Final result

Intercept for $N_c/N_\gamma \rightarrow 1$ at reference date (1976- - , 0 h 00 UT)	..... Bq mg^{-1}
Standard error of the mean	..... Bq mg^{-1}, ... %
Number of degrees of freedom	.....

Time of the measurements (year, month, day) ..... to .....

Let the fitted polynomial be of the form $y = a_o + a_1 x + a_2 x^2 + \ldots$, with $y = \frac{N_\beta N_\gamma}{m N_c}$,
$x = \frac{1 - N_c/N_\gamma}{N_c/N_\gamma}$, m = source mass. If the order exceeds one, indicate the values of the coefficients normalized to unit intercept:
a_1/a_o = ..., a_2/a_o = ..., a_3/a_o = ...
Has the uncertainty in the values of the abscissa x been accounted for ? .....
If so, by how much does this affect a_o ? .....
Goodness of fit: Give a graph of the residuals of the fitted efficiency function similar to the one enclosed. State result of χ^2 test (if carried out):

Formula used for calculating the data points

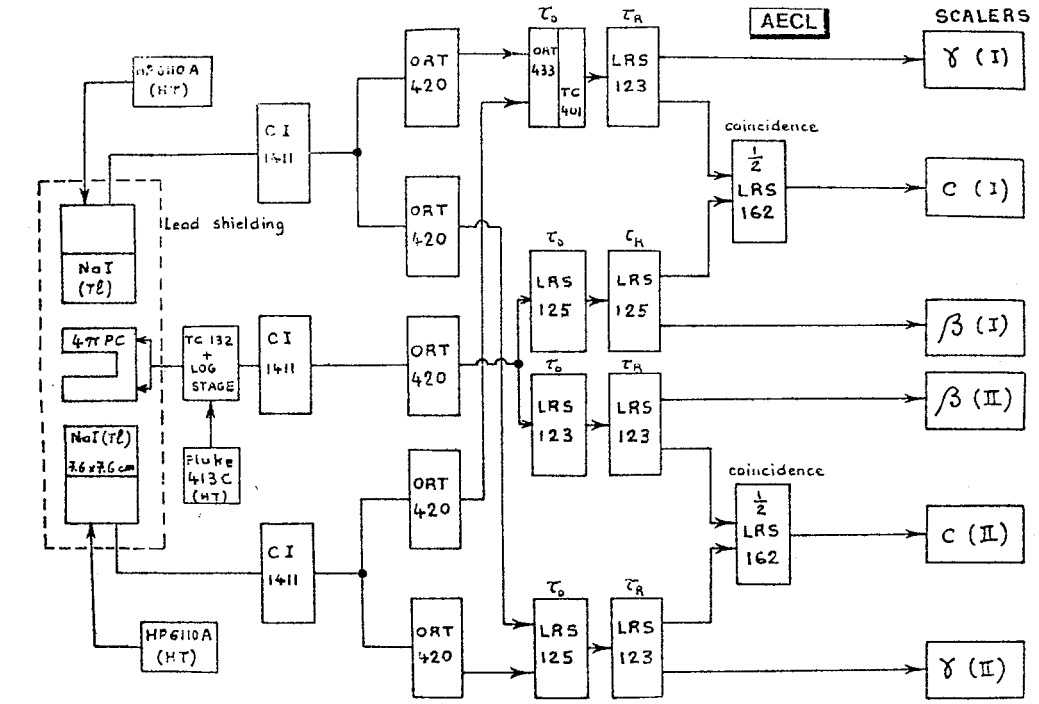
Systematic uncertainty of final result - Explain how it was determined

due to	weighing	..... %	.....
	dead time	..... %	.....
	resolving time	..... %	.....
	background	..... %	.....
	extrapolation	..... %	.....
	others (indicate)	..... %	.....
	total	..... %	.....

Laboratory:
....

Name(s) of person(s) who carried out the measurements
.....

Figure 2 - A selection of equipments used by the participants



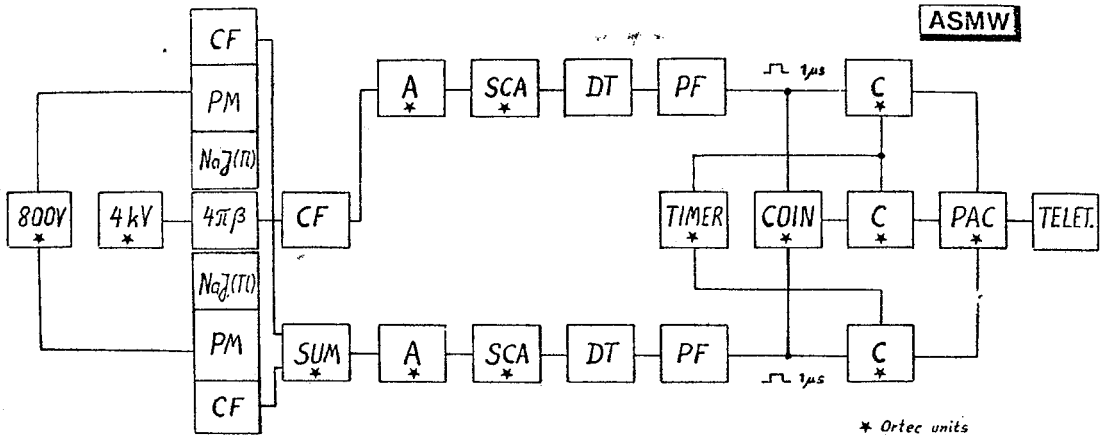
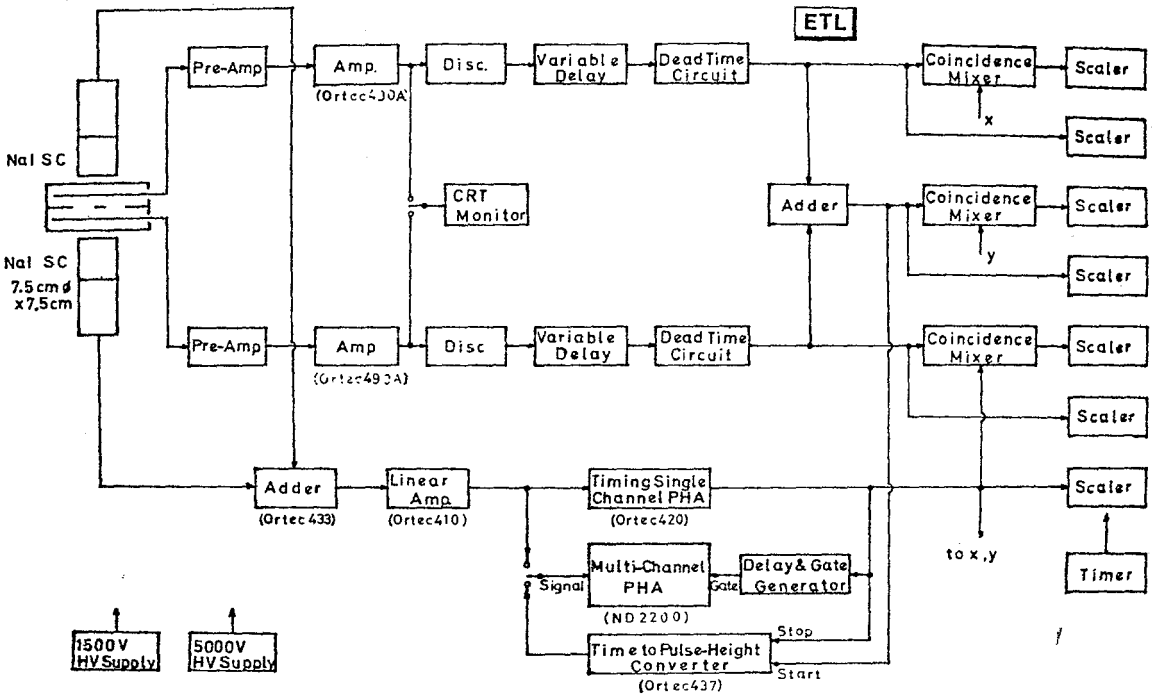
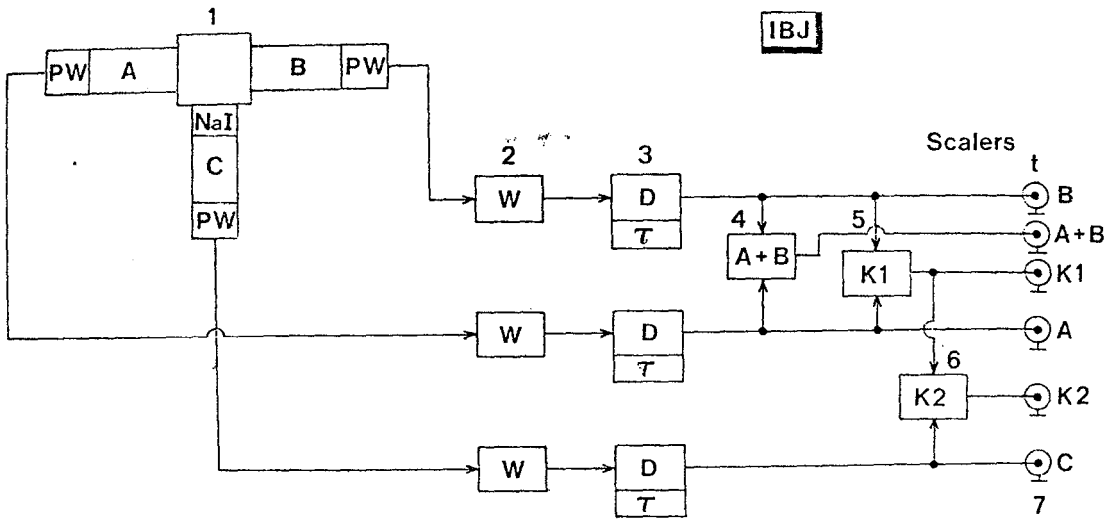


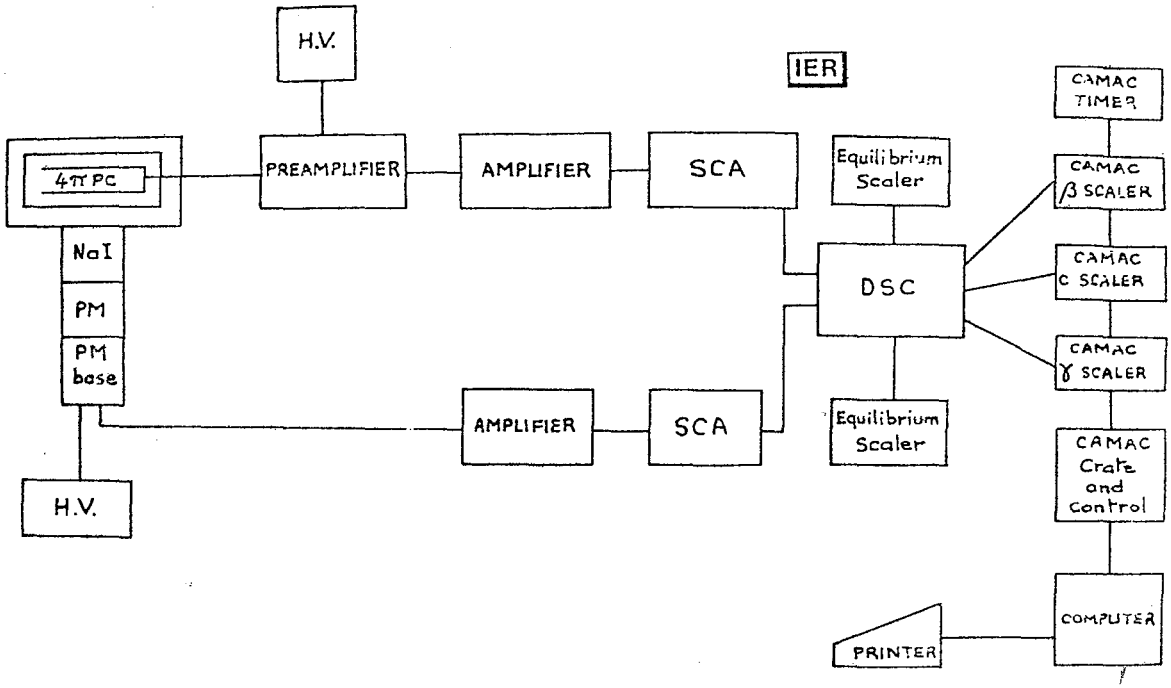
Figure 2 (cont'd)





1 - triple scintillation head, 2 - amplifiers, 3 - discriminators, 4 - sum unit, 5 - first stage coincidence unit, 6 - second stage coincidence unit, 7 - outputs to the scalers

Figure 2 (cont'd)



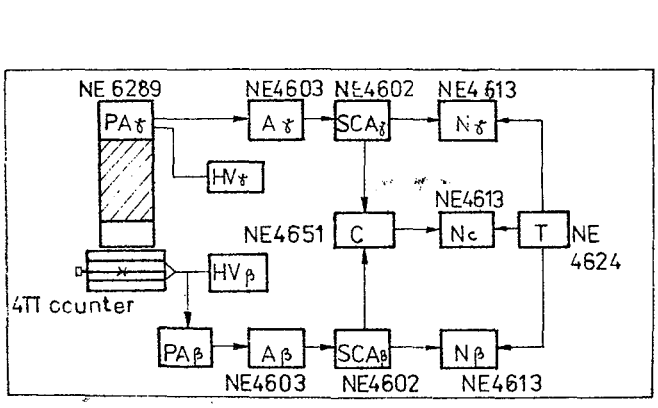
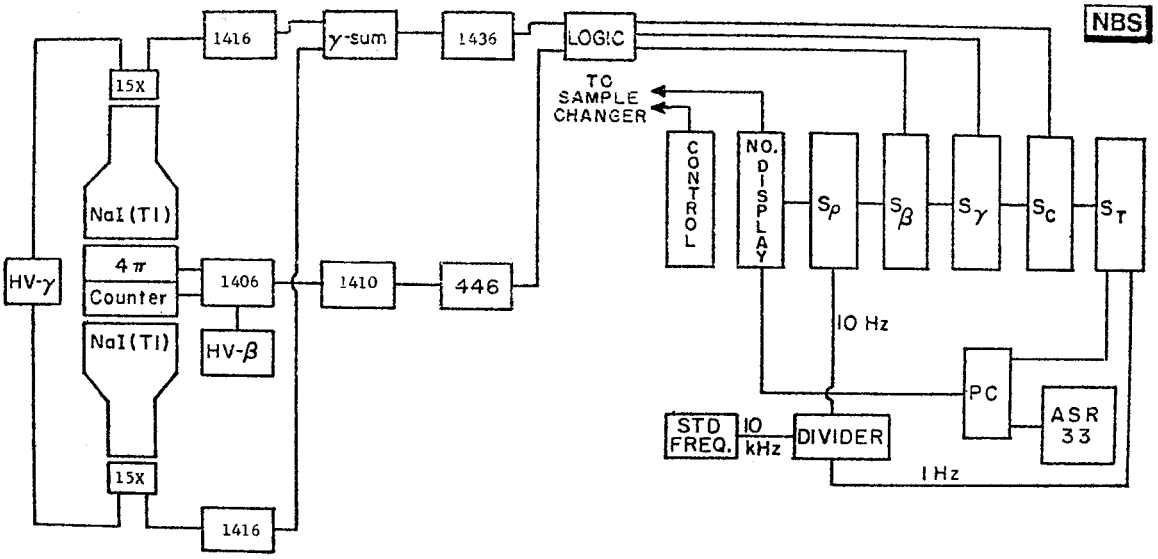


Figure 2 (cont'd)



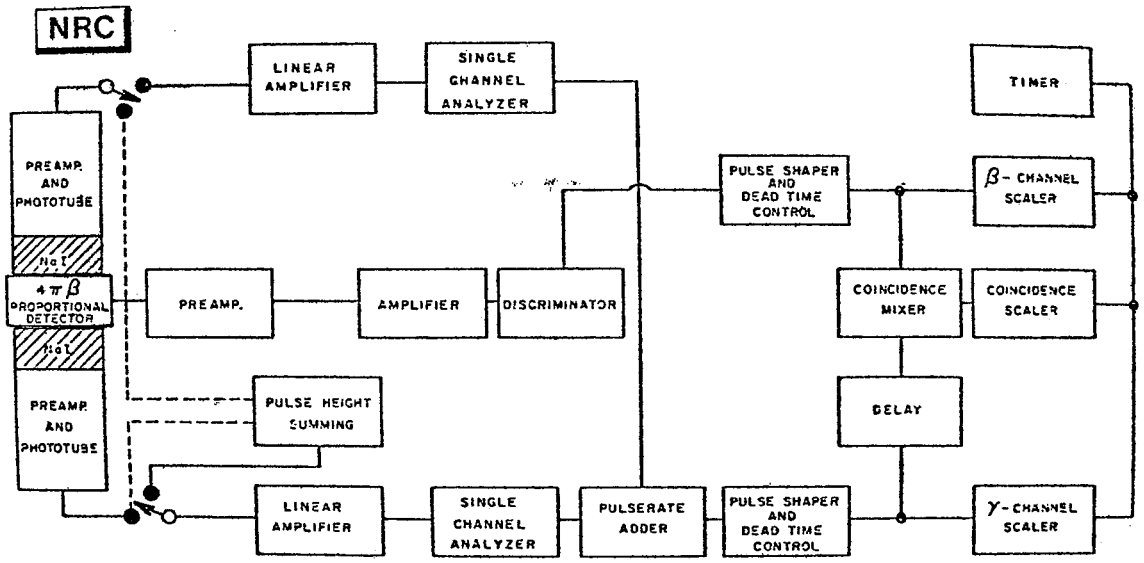
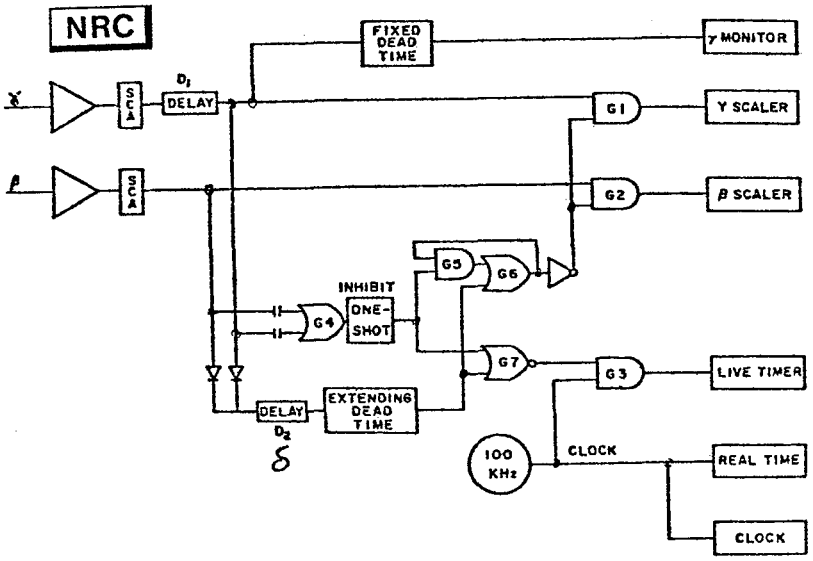


Figure 2 (cont'd)



Schematic of live-timed anti-coincidence circuitry

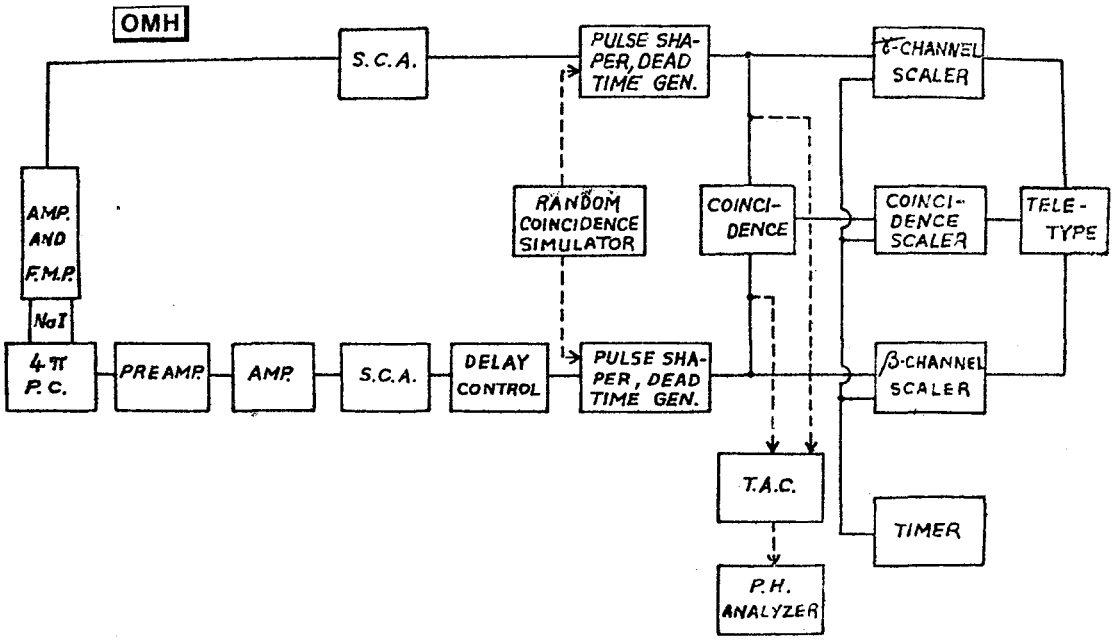
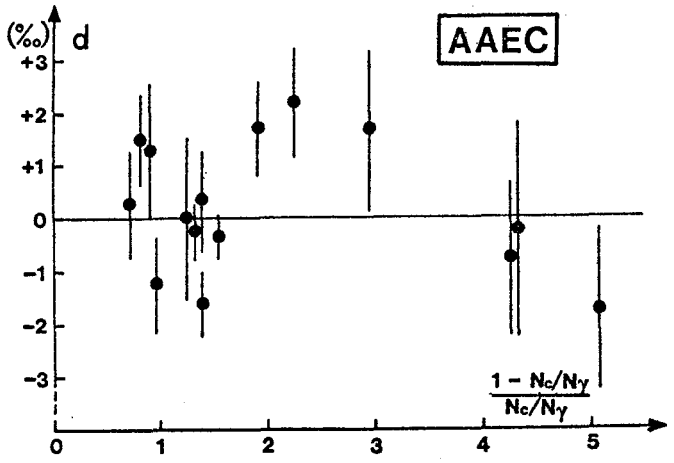


Figure 3 - Efficiency extrapolation, residuals.

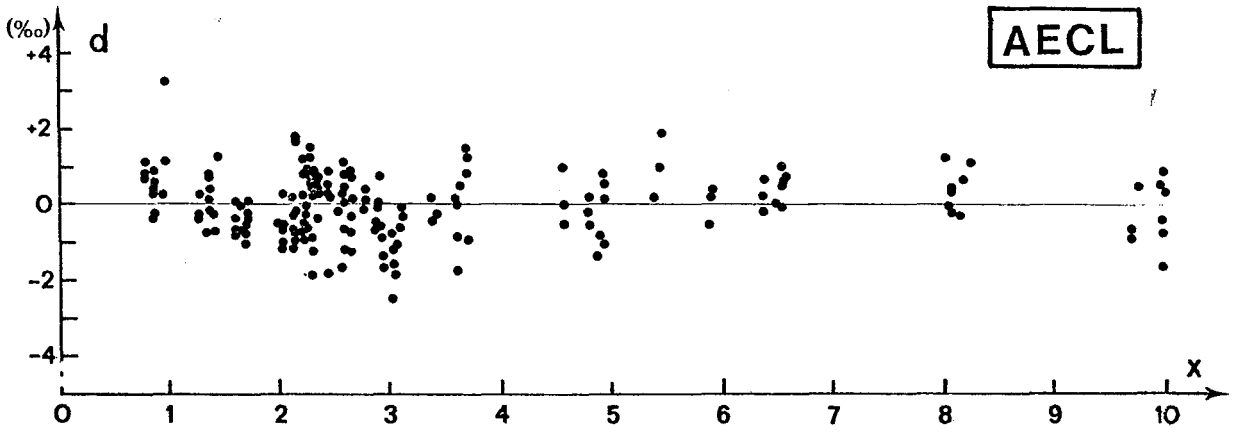
The bars of the data points represent one time the random uncertainty as estimated from

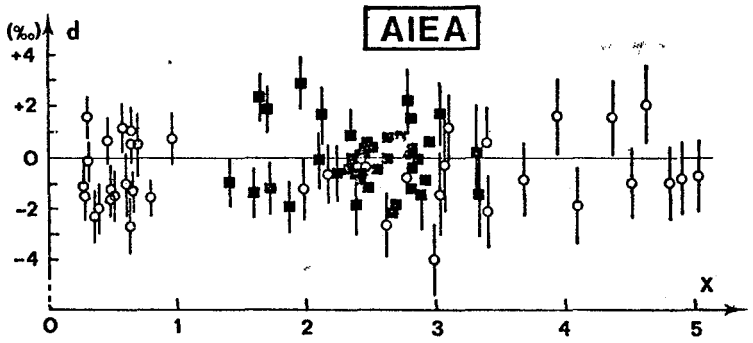
- the efficiencies ε_β and ε_γ in a single count (BIPM, ETL, LMRI, NPRL, OMH), see [28],
- the standard error of the mean of m counts, where m is 5 to 6 (AIEA), 4 (IEA), 20 (IER), 6 to 20 (IPA), 5 (NPL), 4 to 6 (PTB).

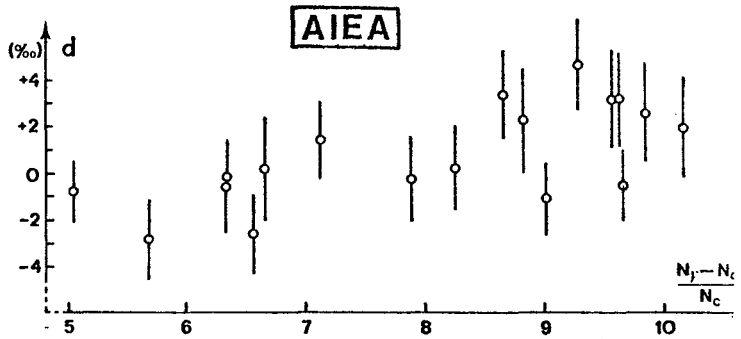


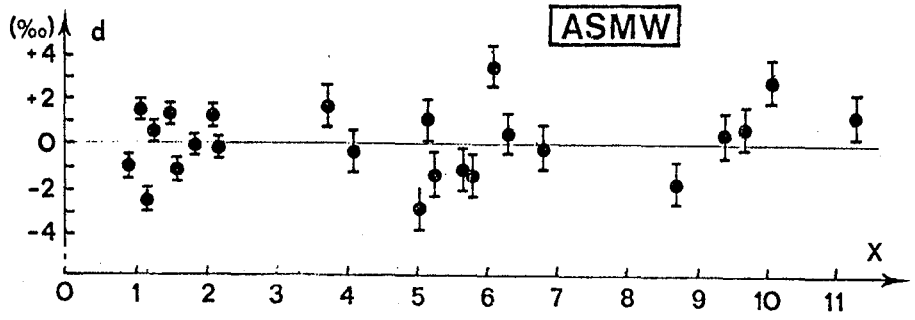
Other participants have used more elaborate procedures:
AECL (see p. 17)
BARC (by combining uncertainties of slope and intersection with the standard deviation according to [28])
NRC (see [27]).

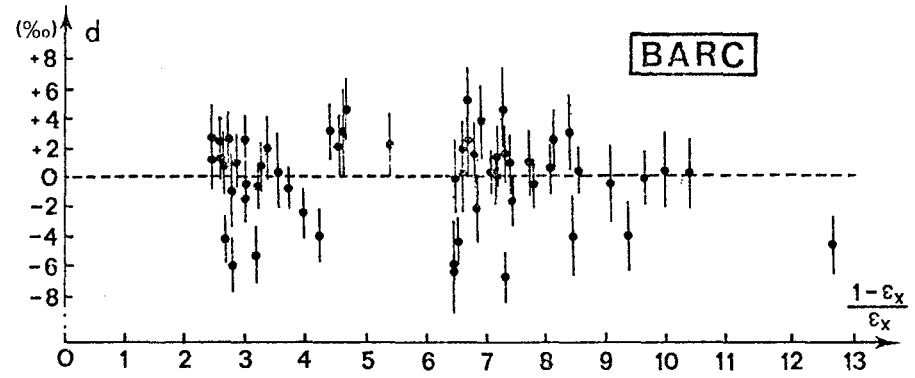
Additional information may be found in the last column of Table 9.

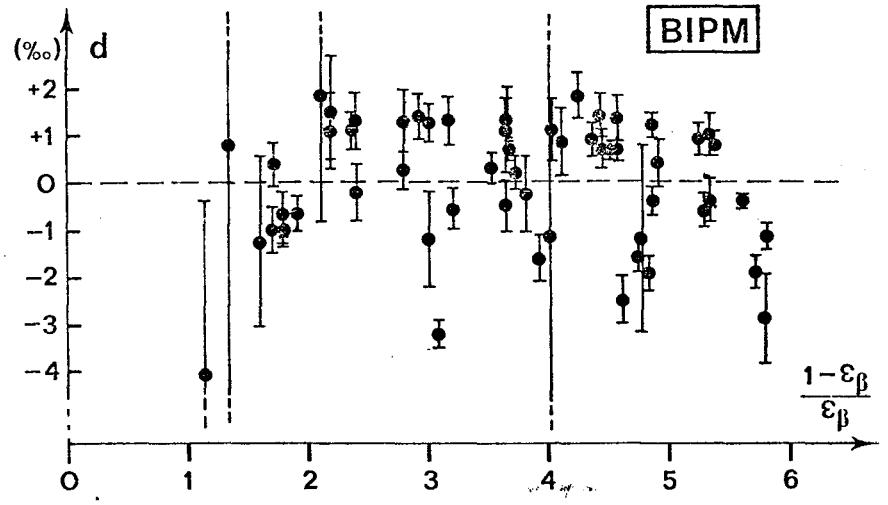


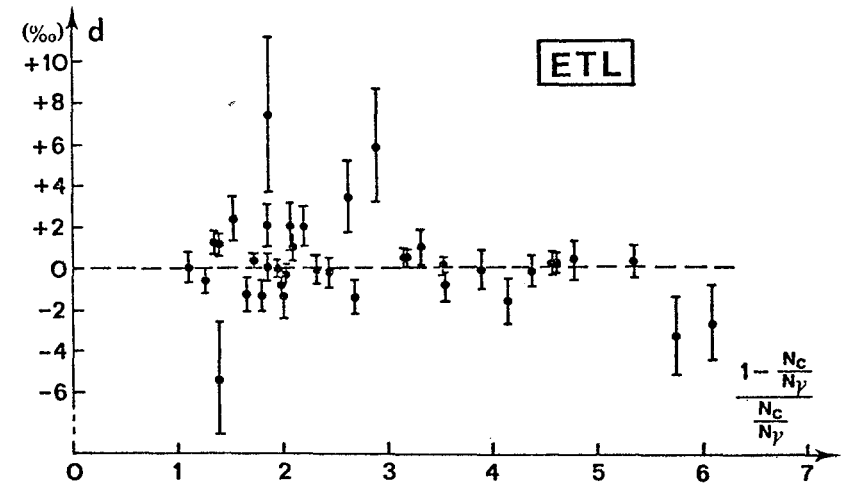


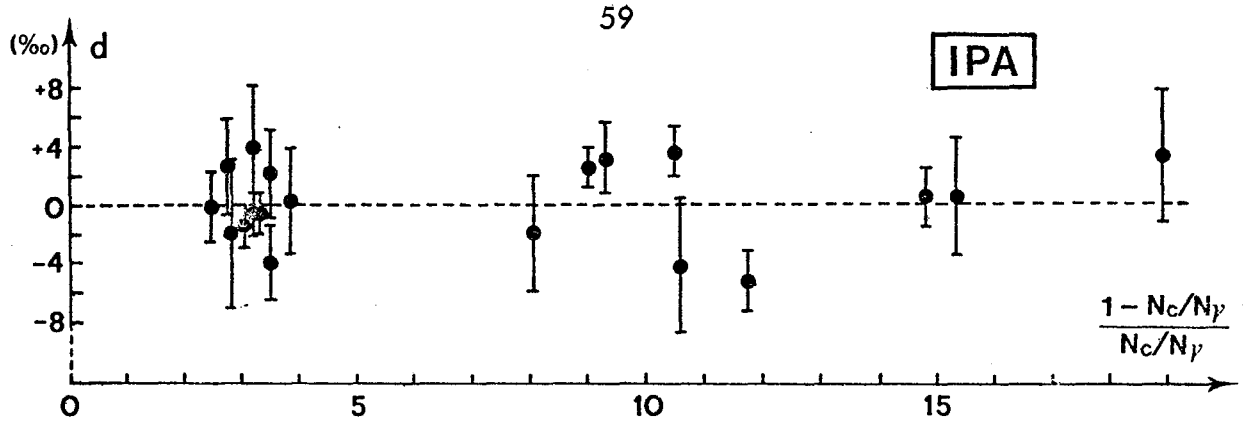


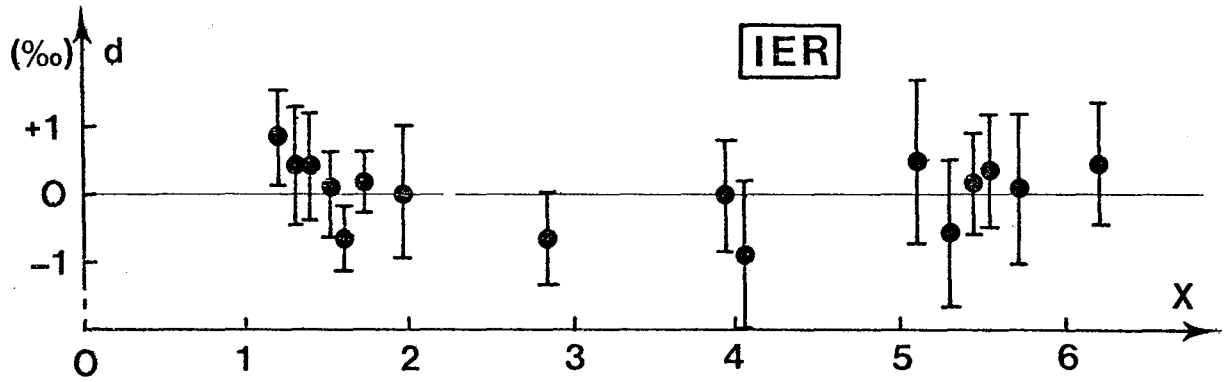

(‰) d
ASMW
+4
+2
0
-2
-4
X
0 1 2 3 4 5 6 7 8 9 10 11

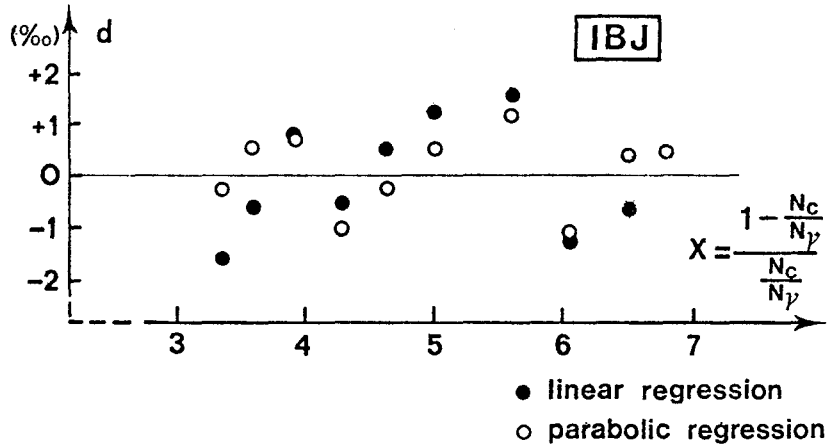

(‰) d
BARC
+8
+6
+4
+2
0
-2
-4
-6
-8
$\frac{1-\varepsilon_x}{\varepsilon_x}$
0 1 2 3 4 5 6 7 8 9 10 11 12 13

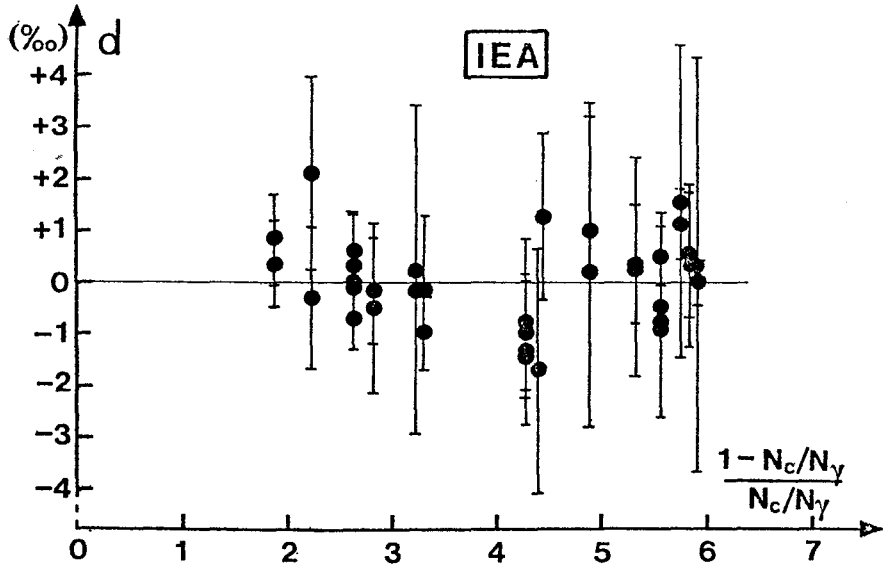

(‰) d
BIPM
+2
+1
0
-1
-2
-3
-4
$\frac{1-\varepsilon_\beta}{\varepsilon_\beta}$
0 1 2 3 4 5 6

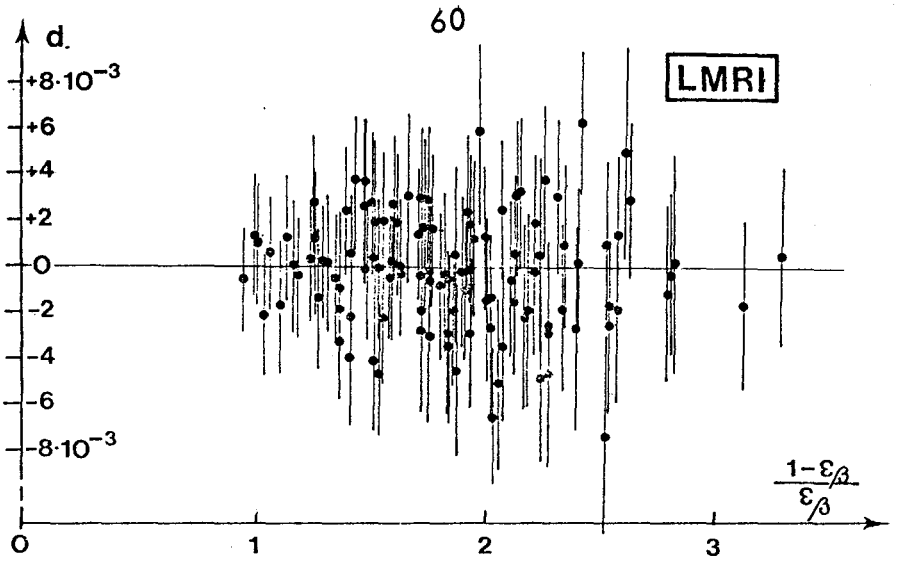

(‰) d
ETL
+10
+8
+6
+4
+2
0
-2
-4
-6
$\frac{1-\frac{N_c}{N_\gamma}}{\frac{N_c}{N_\gamma}}$
0 1 2 3 4 5 6 7

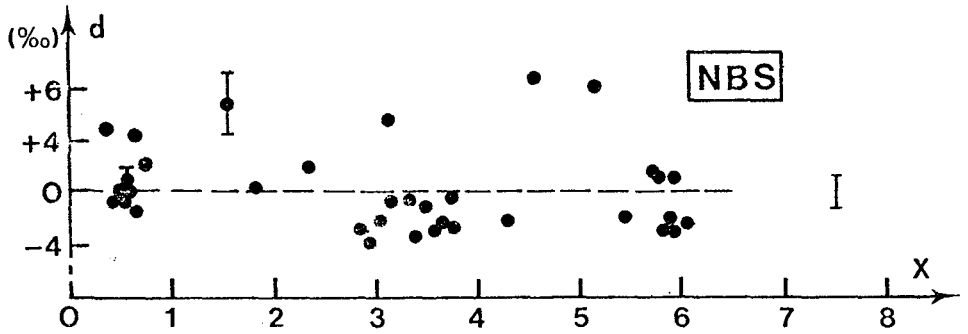

(‰) d
IPA
+8
+4
0
-4
-8
0
5
10
15
1 − Nc/Nγ
Nc/Nγ

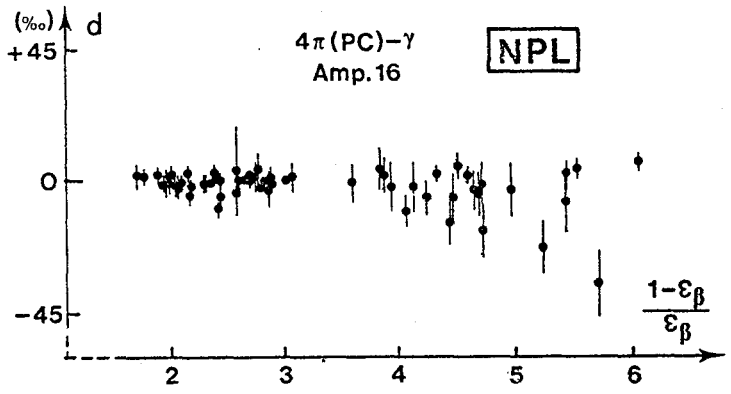

(‰) d
IER
+1
0
−1
0
1
2
3
4
5
6
X

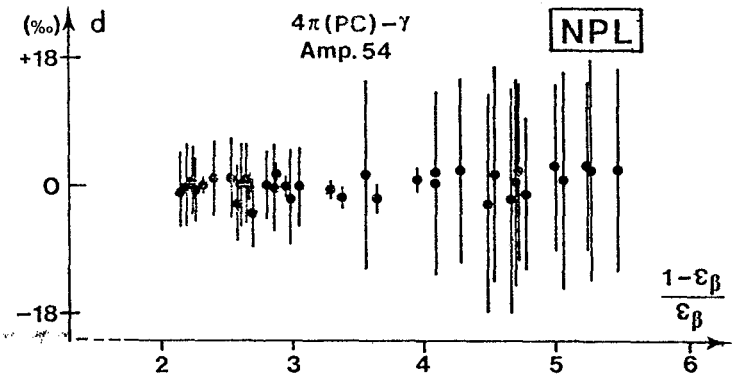

(‰) d
IBJ
+2
+1
0
−1
−2
3
4
5
6
7
X = (1 − Nc/Nγ) / (Nc/Nγ)
● linear regression
○ parabolic regression

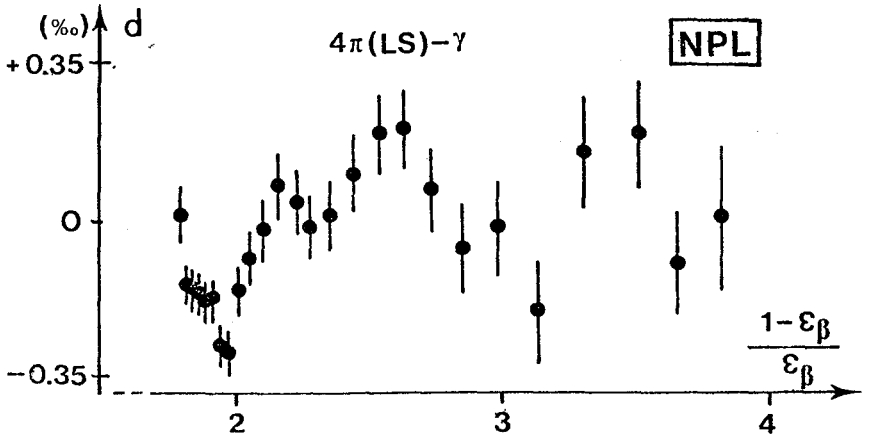

(‰) d
IEA
+4
+3
+2
+1
0
−1
−2
−3
−4
0
1
2
3
4
5
6
7
1 − Nc/Nγ
Nc/Nγ

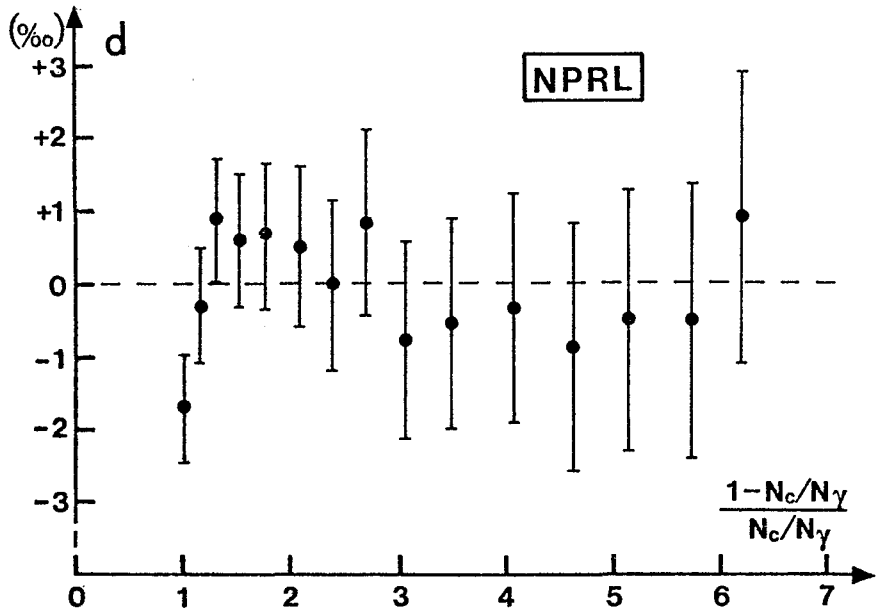

d.
+8·10⁻³
+6
+4
+2
0
-2
-4
-6
-8·10⁻³
LMRI
1-εβ/εβ
0
1
2
3

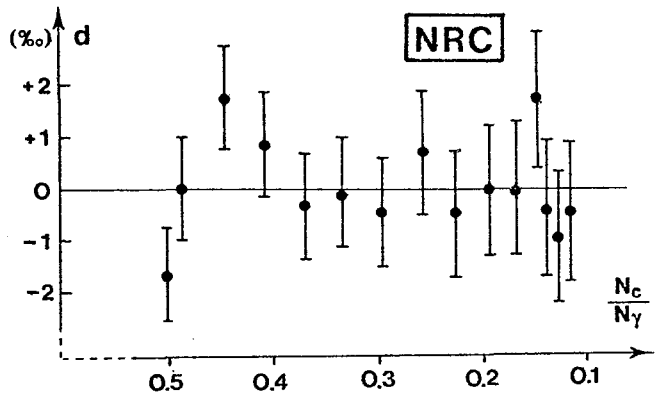

(‰)
d
+6
+4
0
-4
NBS
X
0
1
2
3
4
5
6
7
8

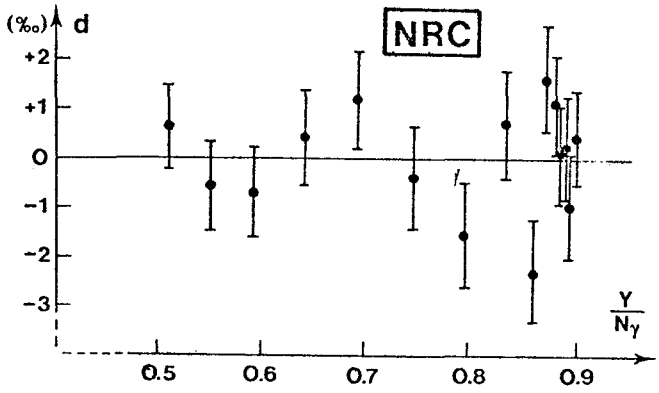

(‰)
d
+45
0
-45
4π(PC)-γ
Amp. 16
NPL
1-εβ/εβ
2
3
4
5
6

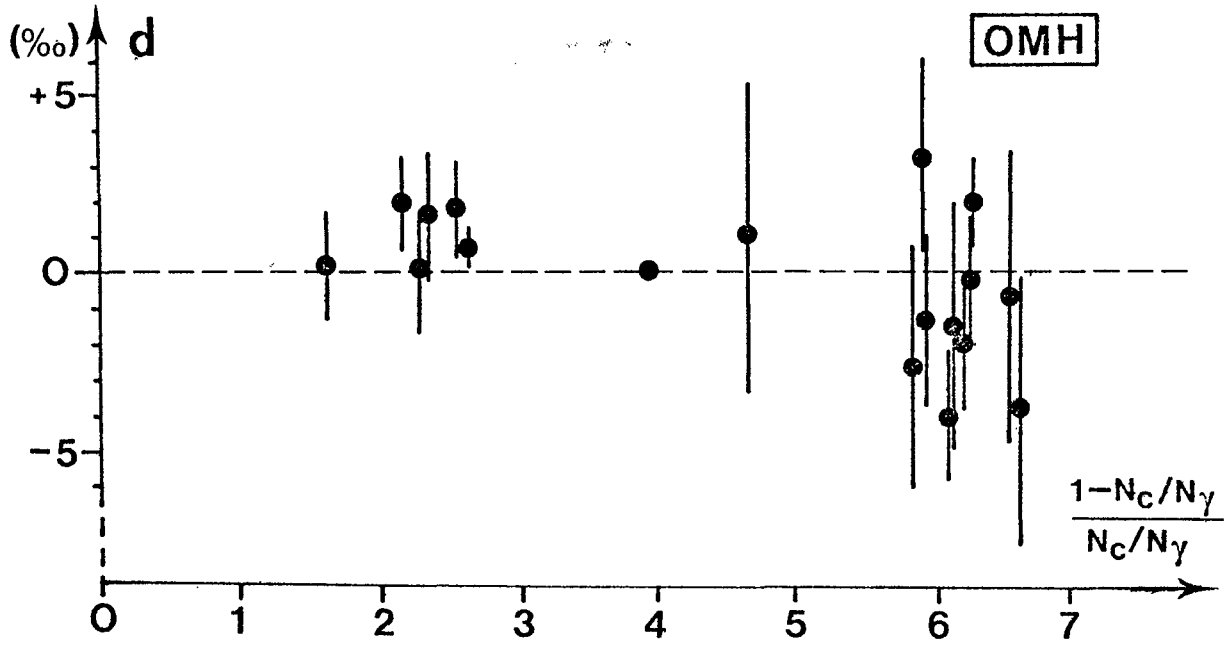

(‰)
d
+18
0
-18
4π(PC)-γ
Amp. 54
NPL
1-εβ/εβ
2
3
4
5
6

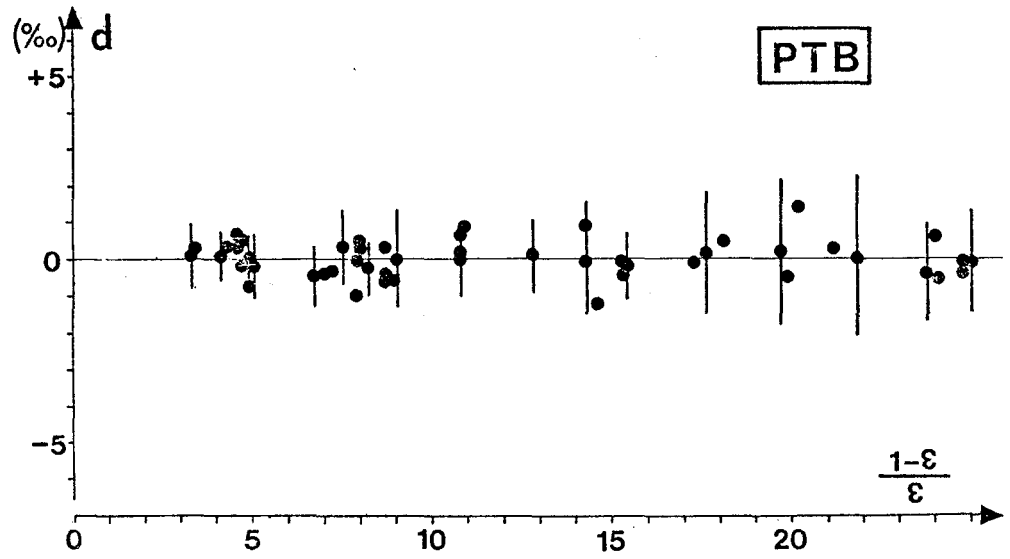

(‰)
d
+0.35
0
-0.35
4π(LS)-γ
NPL
1-εβ/εβ
2
3
4

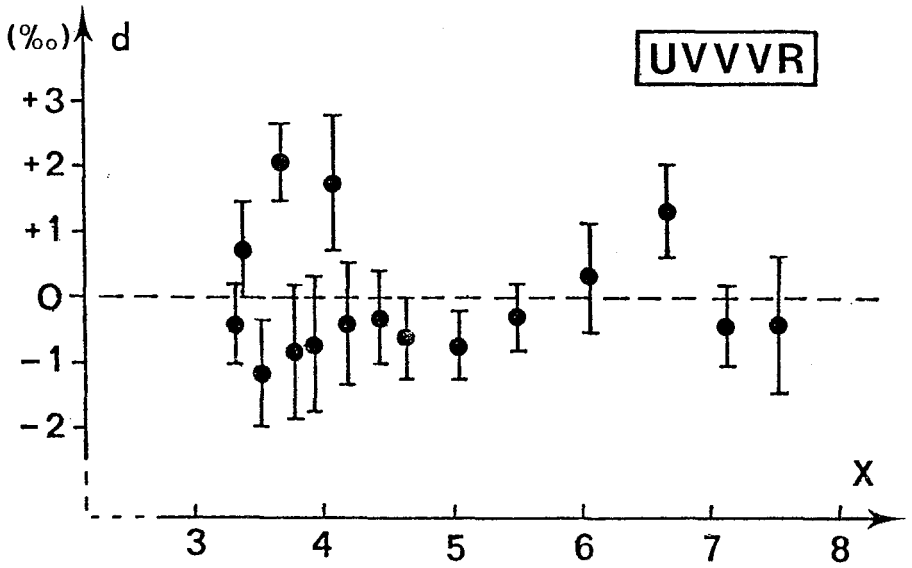

(‰) d
NPRL
+3
+2
+1
0
-1
-2
-3
1−Nc/Nγ / Nc/Nγ
0 1 2 3 4 5 6 7


(‰) d
NRC
+2
+1
0
-1
-2
Nc/Nγ
0.5 0.4 0.3 0.2 0.1

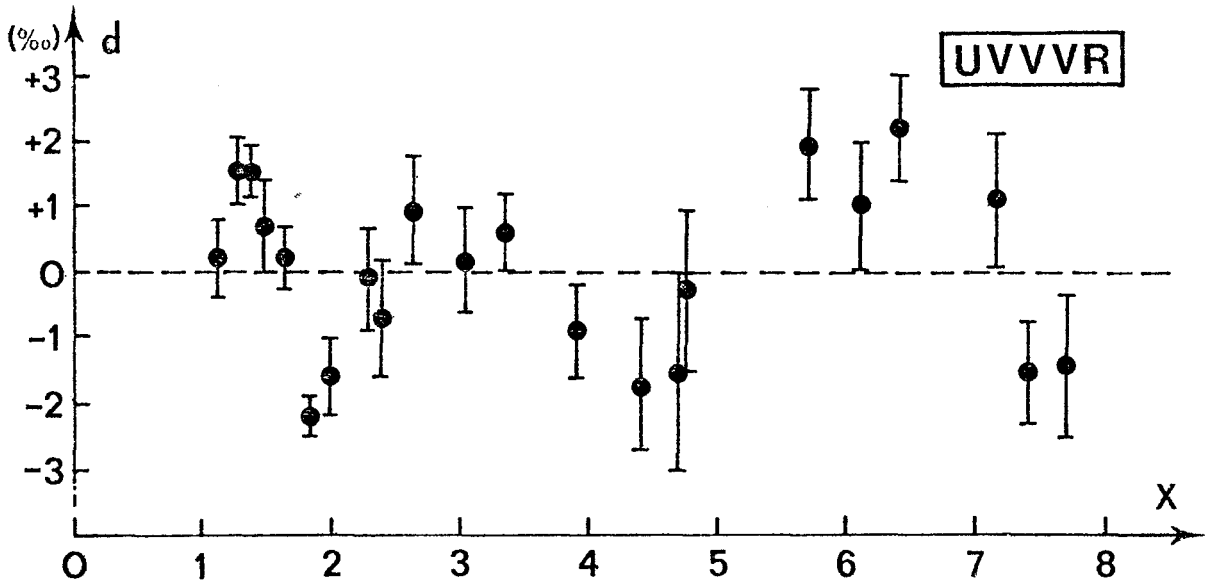

(‰) d
NRC
+2
+1
0
-1
-2
-3
Y/Nγ
0.5 0.6 0.7 0.8 0.9


(‰) d
OMH
+5
0
-5
1−Nc/Nγ / Nc/Nγ
0 1 2 3 4 5 6 7





Dried source linear regression



Electrodeposited source quadratic regression

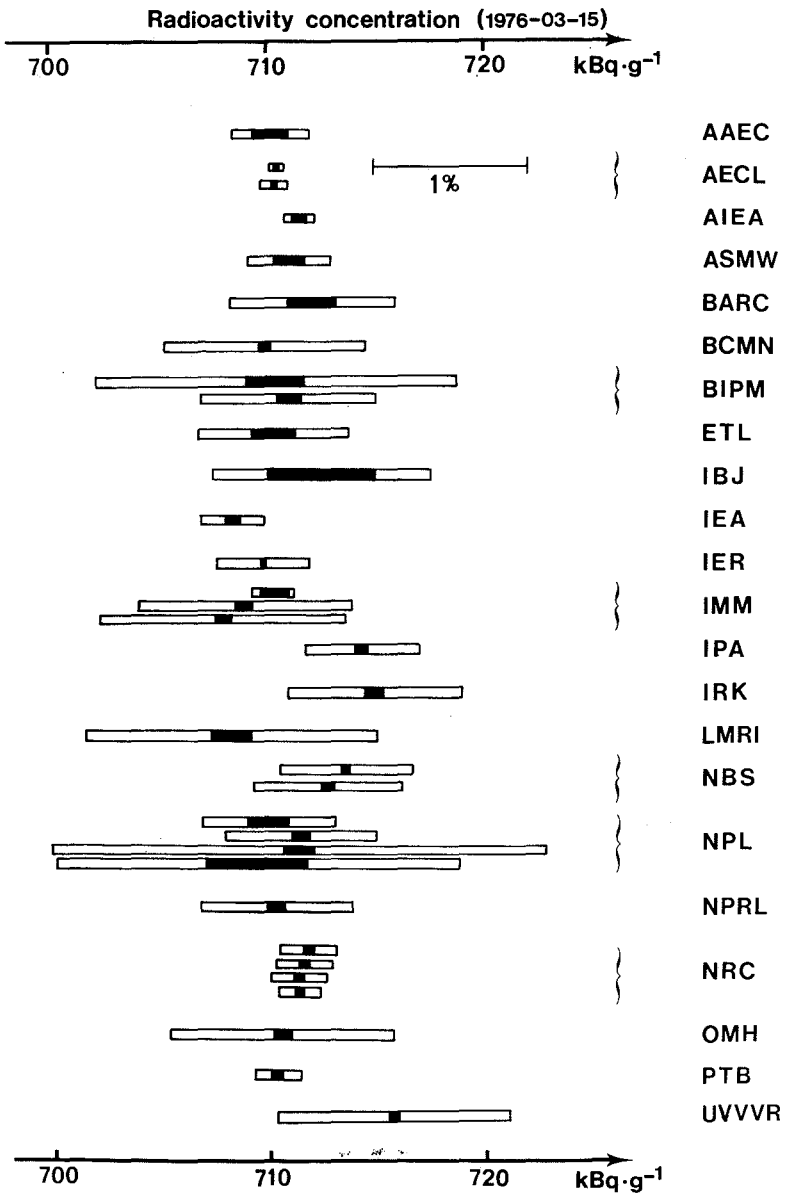


Figure 4 - Graphical representation of the results.
The black (or white) rectangles correspond to the random (or systematic) uncertainties.

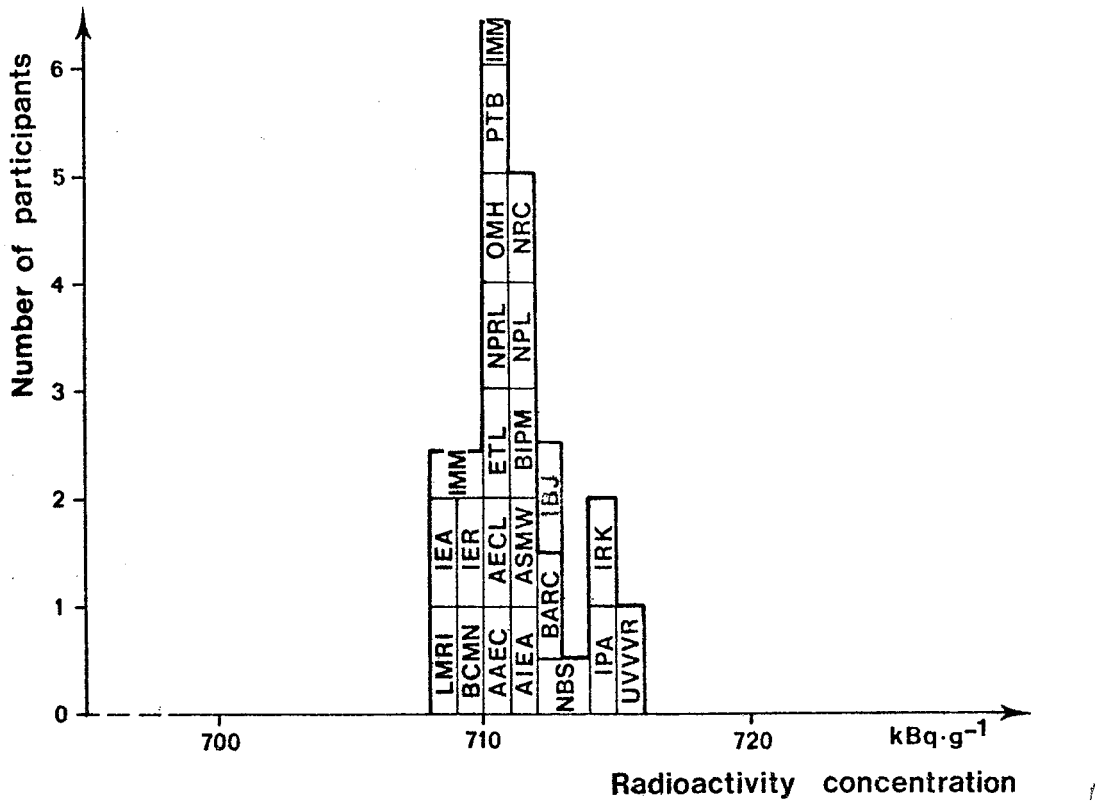


Figure 5 - Distribution of the results of the radioactivity concentration.

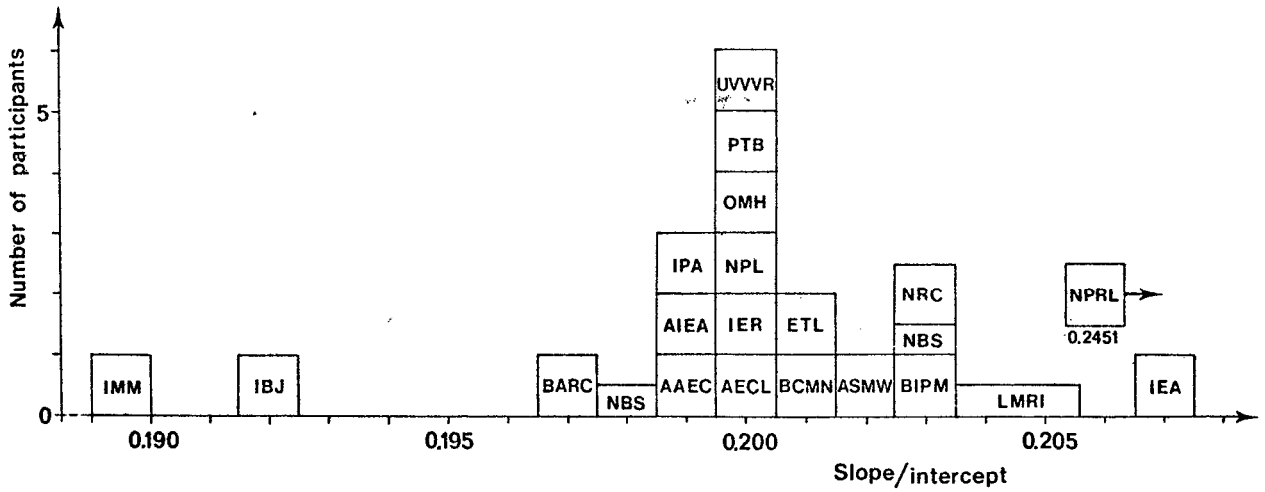


Figure 6 - Distribution of the values obtained for the slope-to-intercept ratio.

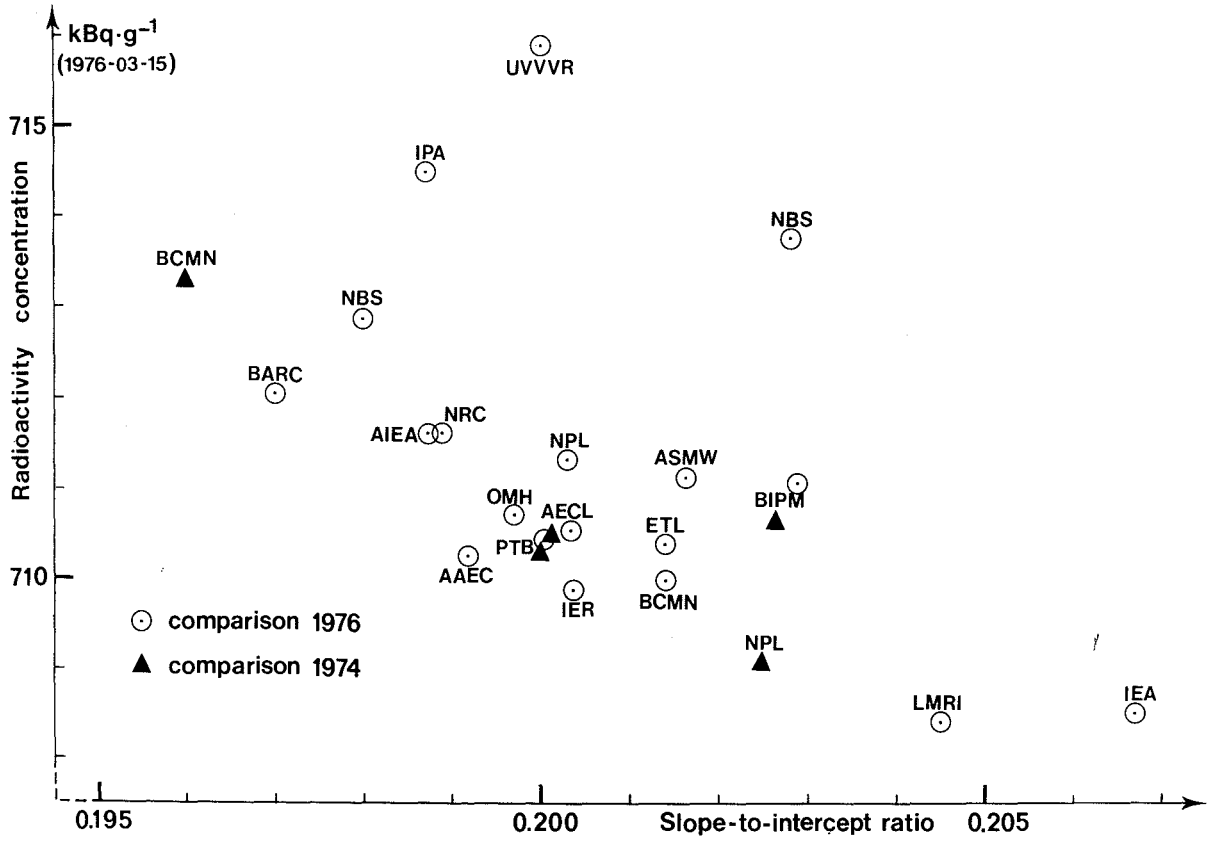


Figure 7 - Correlation of the intercept and the slope-to-intercept ratio. The normalized results of the preliminary comparison [5] are included for comparison.

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