

International comparison of activity measurements  
of a solution of  $^{137}\text{Cs}$  (May 1982)

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Abstract

Nineteen national or international laboratories participated in a comparison organized by the Bureau International des Poids et Mesures (BIPM), in order to test the method of efficiency tracing. Samples of similarly composed and highly pure solutions of  $^{137}\text{Cs}$  and of  $^{134}\text{Cs}$  were prepared and distributed by the Physikalisch-Technische Bundesanstalt (PTB). The activity concentration of the  $^{134}\text{Cs}$  solution (tracer) was determined by means of the International Reference System (SIR). A fraction of about  $3 \cdot 10^{-4}$  of the activity has been found to remain adsorbed on the ampoule walls.

Details concerning counting equipment, counting data and source preparation are presented. The efficiency-tracing method is outlined and formulae for calculating the activity concentration are discussed. The participants measured the  $\beta$ -particle counting efficiency  $\epsilon_{\beta}$  by various coincidence techniques, by anti-coincidence counting or by the selective-sampling method. Source-preparation procedures mostly followed current practice. First- and second-order functions for extrapolation to a  $\beta$  efficiency  $\epsilon_{\beta} = 1$  were used about equally often.

Uncertainty components and the influence of conversion electrons and photons from  $^{137}\text{Ba}^{\text{m}}$  are discussed. The final results and their uncertainties are presented in tabular form and on a graph, where they are compared with results from a previous comparison and from independent SIR results. The total range of 2.4% and the standard deviation of 0.66% of the mean are larger than usual in BIPM comparisons, but reflect the inherent difficulties of the method applied.

## 1. Introduction

With its long half life of about thirty years and its single  $\gamma$  ray of 662 keV,  $^{137}\text{Cs}$  is widely used for calibrating  $\gamma$ -ray spectrometers. It had already been noted down for an international comparison of activity measurements in 1963 [1] but, in view of certain difficulties which were to be expected, this enterprise was postponed. Since the decay scheme (Fig. 1) is now known sufficiently well,  $\gamma$ -emission rates can be derived from activities with a rather high accuracy. It therefore seemed adequate to conceive this comparison as a measurement of the more essential quantity which is activity.

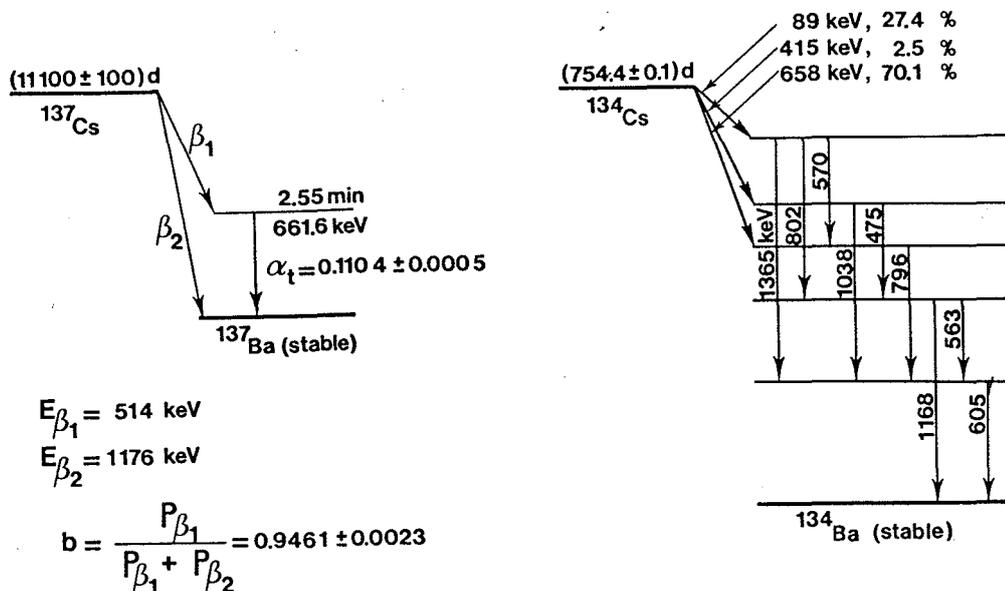


Fig. 1 - Decay schemes of  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$ . The values of the nuclear parameters were taken from ref. [2] to [5].

The experience gained by a preliminary comparison, arranged by the Bureau International des Poids et Mesures (BIPM) among a restricted number of participants [6], had provided useful hints as to organizational details and made clear that efficiency tracing is likely to present some difficulties. However, this technique is certainly most appropriate for absolute activity determinations of this nuclide and was therefore applied by nearly all the participants. Consequently, a large part of the following discussion will be devoted to this method and the results thus obtained. Although the participants were free to choose other tracer nuclides ( $^{60}\text{Co}$ ,  $^{82}\text{Br}$ , ...), they all preferred  $^{134}\text{Cs}$  which was distributed along with the solution to be standardized.

The national laboratories manifested considerable interest in these measurements, as is reflected by the large number (23) of participants. However, only nineteen of them have finally submitted their results (Table 1).

The details of the organization were explained in a circular letter dated 30 March 1982, and accompanied by an extensive reporting form based on previous experience. The reference date chosen was 1982-05-01, 0 h UT. The filled-in forms were received, with one exception, by the end of September 1982. A preliminary report [7] summarizing the information contained in the forms was issued soon afterwards.

## 2. Description of the solutions distributed; purity tests

Two different solutions were prepared and distributed by PTB. Each participant received one ampoule of  $^{137}\text{Cs}$  and one ampoule of  $^{134}\text{Cs}$ , the latter being recommended as efficiency tracer. Both solutions had similar chemical compositions and activity concentrations. The following values had been adopted: about  $600 \text{ kBq g}^{-1}$  in aqueous solutions of  $0.2 \text{ mol HCl per dm}^3$  with  $20 \text{ }\mu\text{g}$  of  $\text{CsCl}$  per gram of solution. The ampoules were of the type currently used by PTB and contained each about  $4 \text{ cm}^3$  of solution. The mass of the solution had been determined accurately and was notified to the participants. BIPM received three NBS-type ampoules of each solution for ionization-chamber measurements.

Since the results were to be expressed in terms of activity, they contained several nuclear parameters the values for which were proposed in the decay schemes represented in Fig. 1.

In order to simplify the measurements, it was decided that the BIPM determine the activity concentration of the tracer solution with the aid of the International Reference System for activity measurements of  $\gamma$ -ray-emitting nuclides (SIR) [8]. The mean value obtained from measurements of three ampoules of the tracer solution was  $(638.3 \pm 0.6) \text{ kBq g}^{-1}$ , on the reference date. This value was communicated to the participants soon after the distribution of the ampoules.

Seven laboratories made an additional effort by measuring this activity concentration using various methods, as indicated in Table 2.

Purity tests by  $\gamma$ -ray spectroscopy were carried out on samples of both solutions, before distribution to the participants. The results obtained by LMRI, NBS and PTB under different measuring conditions agreed sufficiently well and can be summarized as follows. While in the  $^{134}\text{Cs}$  solution no  $\gamma$ -ray-emitting impurity exceeding  $5 \cdot 10^{-5}$  of the main activity (on the reference date) could be identified, the relative content of  $^{134}\text{Cs}$  in the  $^{137}\text{Cs}$  solution was about  $4 \cdot 10^{-5}$  and represented the only impurity identified.

## 3. Ionization-chamber measurements and adsorption tests

About half of the participants carried out some measurements of activity concentration of the  $^{137}\text{Cs}$  solution by a calibrated ionization chamber, using either the original ampoule or their own type, after transferring and weighing the solution. Later on, when the ampoule was practically empty, it was rinsed twice with distilled water. The remaining activity gave a measure of the adsorption on the ampoule walls

which turned out to be of the order of three parts in ten thousand. NPL further reported important adsorption on the liquid-scintillation counting vessel. The results are presented in Table 3.

#### 4. Sources and detectors for proportional counting

Previous experience with  $^{134}\text{Cs}$  [9] had shown that, in general, no special treatment of the sources is needed in order to obtain high enough values of the  $\beta$ -counting efficiency  $\epsilon_{\beta}$  of proportional counters. Therefore, only a few participants have applied such treatments. They were as follows:

- Drop dispensing on electrosprayed pads of ion-exchange resin
- UVVVR reported that, for sources treated with the wetting agent Catex-anex, drying was delayed until twenty hours after preparation,
- SCK added carrier or just water to some sources,
- AECL added extra carrier to some of the sources for varying  $\epsilon_{\beta}$ .

Most participants used the solutions undiluted. Further details are given in Table 4. The most important data concerning proportional counters are summarized in Table 5.

#### 5. Detection of $\gamma$ rays

A typical  $\gamma$ -ray spectrum obtained from a mixed source of  $^{137}+^{134}\text{Cs}$  in the BIPM  $4\pi\beta$ - $\gamma$  counting set is shown in Fig. 2. Most of the participants used similar scintillation detectors. Some larger NaI(Tl) crystals were also employed. In order to avoid counting of photons from  $^{137}\text{Ba}^m$  (661.6 keV), a good energy resolution was important and more likely to be achieved with recent detector systems.

Two participants (LMRI and NRC) used Ge(Li) detectors. NRC, using anti-coincidence counting methods, also obtained two-parameter data sets [10] with a second  $\gamma$ -ray detector (NaF) for a (1250 - 1500) keV channel.

Some data regarding the  $\gamma$ -ray detectors are presented in Table 6. The  $\gamma$ -channel settings can be found in Table 9. From Table 11 it may be seen that most of the participants estimated the uncertainty due to the counting of (unwanted) 662 keV photons as being of minor importance.

#### 6. Liquid-scintillation counting

NAC and NPL applied  $4\pi\beta$ (LS)-coincidence counting, whereas NIM used the recently developed  $4\pi\text{LS}$  method with efficiency variation by optical filters and extrapolation [11]. Details concerning source preparation and detector systems may be found in Table 7.

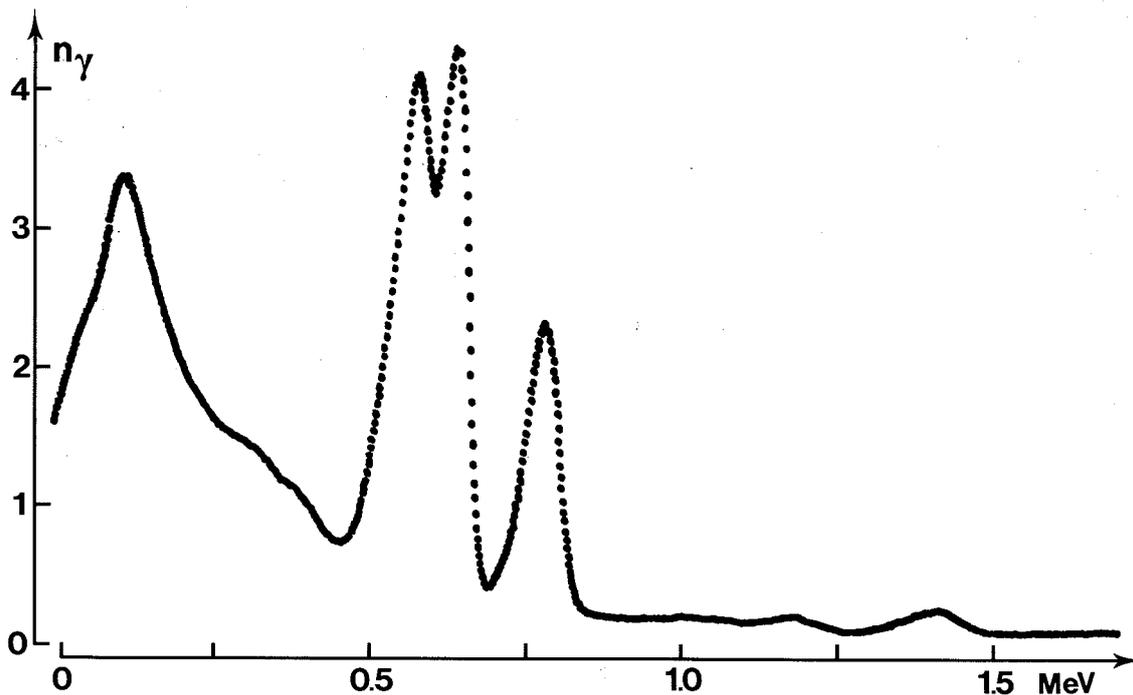


Fig. 2 - Typical  $\gamma$ -ray spectrum from a mixed source of  $^{137}\text{+}^{134}\text{Cs}$ .  
In most cases, the  $\gamma$ -channel window was set on the well-resolved peak at about 800 keV.

#### 7. $4\pi\gamma$ counting with a large NaI well crystal

In addition to  $4\pi\beta\text{-}\gamma$  coincidence measurements, SCK used a NaI well crystal in order to determine the 661.6 keV photon-emission rate. Geometrical data of the crystal had been given in [9]. Five sources were counted during 2 000 s each. A threshold was set between 70 and 160 keV, and the count rates obtained were about  $4\ 000\ \text{s}^{-1}$ . Corrections were applied for  $\beta$  particles, conversion electrons, material surrounding the crystal surface, source mount and source dimensions. The photon efficiency was calculated taking into account absorption coefficients taken from the literature [12] and accurately determined crystal dimensions.

#### 8. The efficiency-tracing method

Due to the long mean life (221 s) of the 661.6 keV state in  $^{137}\text{Ba}^{\text{m}}$ , the correlation between  $\beta$ - and  $\gamma$ -emission events has nearly completely disappeared. Therefore,  $^{137}\text{Cs}$  has to be treated, as far as activity measurements are concerned, as a pure  $\beta$ -ray emitter, for which efficiency tracing is the recommended method and may be summarized as follows. A known amount of a radionuclide emitting coincident  $\beta$  and  $\gamma$  events ( $^{134}\text{Cs}$ ) is added to the pure  $\beta$ -ray emitter. Sources of this mixture are measured by conventional  $4\pi\beta\text{-}\gamma$  counting, combined with efficiency ( $\epsilon_{\beta}$ ) variation and extrapolation. The value of the  $\beta$ -count rate  $N_{\beta}$  of the traced nuclide is obtained by subtraction of the contribution due to

the tracer alone and is plotted against  $1 - \epsilon_{\beta}$ . The extrapolated value minus the count rate of conversion electrons is finally taken as the activity of the pure  $\beta$ -ray emitter ( $^{137}\text{Cs}$ ).

This method has been developed by Campion et al. [13] and was further investigated by several authors [14 to 16]. Applications to the standardization of  $^{137}\text{Cs}$  have been reported in ref. [17] to [20]. A full description of the calculations for this method may be found in Section 9.

It is generally agreed that intimate mixing of the two components and, less stringently, comparable  $\beta$  end-point energies and spectral shapes, are the main requirements for a successful application of this technique. However, the preliminary comparison [6] had already shown that additional problems existed which had not been clearly identified previously. Also, the spread of 1.6% was about the same as that obtained from the SIR results. Therefore, a thorough analysis was needed for which a detailed reporting form asking precise questions was of high importance.

The simplicity of the method outlined would suggest that, compared with ordinary coincidence counting, nearly equally precise results can be obtained. However, this does not seem to be the case. There are in fact some pitfalls which might be responsible for the relatively large spread of the results obtained and which we shall now try to identify. They are of three different kinds:

1. Effects which distort the efficiency function  $N_{\beta}(\epsilon_{\beta})$ , namely
  - a) Differing  $\beta$ -ray spectra of tracer and traced nuclides,
  - b) Variations of  $\epsilon_{ce}$  and  $\epsilon_{\beta\gamma}$  with  $\epsilon_{\beta}$  ( $\epsilon_{ce}$  and  $\epsilon_{\beta\gamma}$  are the efficiencies of the  $\beta$ -ray detector to conversion electrons and to  $\gamma$  rays, respectively).
2. Effects which do not vanish with  $\epsilon_{\beta} \rightarrow 1$ 
  - a) Accidental coincidences,
  - b) Out-of-channel and pile-up events,
  - c) Influence of the mixing ratio,
  - d) Value of  $\epsilon_{\beta\gamma}$ .
3. Shape of the efficiency function and counting statistics.

Since no other tracer than  $^{134}\text{Cs}$  was used, the influence of 1.a) could only be judged from runs taken with different  $\gamma$ -channel settings. The following table contains the relative contributions (in percent) of the three  $\beta$ -ray branches to the coincidence-count rate.

In the few cases where results obtained with different  $\gamma$ -channel settings were reported, the differences were only small and contradictory in sign. As to the correlations between various detection efficiencies (1.b), no experimental results are available. However, it seems not unreasonable to assume that a possible distortion of an efficiency curve may be taken care of by the extrapolation procedure.

$P_{\beta}$ (%)	27.4	2.5	70.1
$\beta$ -ray end-point energy (keV)	89	415	658
$\gamma$ -channel setting (keV)			
500 to 700	39	3	57
700 to 900	26	-	74
1 260 to 1 490	100	-	-

The effects under 2) are often corrected for only approximately. Nevertheless, such estimations will help to evaluate certain uncertainty components. A few participants used different mixing ratios of the two solutions (Table 4). However, no significant influence on the results was reported, as was already observed in the foregoing comparison [6]. AECL estimated that 1/4 of the difference obtained with a wide and a narrow  $\gamma$ -ray-energy gap could be ascribed to pile-up. The important problems connected with the shape of the efficiency function will be discussed in the following section.

### 9. Efficiency functions

The  $\beta$  efficiency  $\varepsilon_{\beta}$  of the  $\beta$ -ray detector was in general determined by coincidence measurements carried out for various  $\varepsilon_{\beta}$  values and subsequent extrapolation to  $\varepsilon_{\beta} \rightarrow 1$ . The participants were asked to state in detail the expressions of the variables  $x$  and  $y$  of the efficiency function(s) used. The answers are summarized in Table 8. No attempt has been made to arrive at a more uniform notation, and we merely reproduced the formulae indicated in the reporting forms. However, since neither these formulae nor the brief explanations found in the literature ([13] to [20]) describe clearly enough the calculations and their application to the present case, it was felt that a detailed formulation\* in a simplified notation might fill this gap.

In what follows, the symbols  $a$ ,  $N$ ,  $D$ ,  $m$  and  $\varepsilon$  designate activity concentration at the reference date,  $\beta$ -count rate, decay-correction factor, source mass and  $\beta$ -counting efficiency, respectively. The mass numbers of the two cesium isotopes considered are referred to by subscripts 7 or 4. Thus, for a pure  $^{137}\text{Cs}$  source, the  $\beta$ -count rate per unit source mass at a reference time is given by

$$N_7 D_7 / m_7 = a_7 [\varepsilon_7 + C], \quad (1)$$

where the constant  $C$  is the contribution of conversion electrons and

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\* The Section 9 is largely based on a recent communication by D. Smith (NPL), and valuable comments by H.-M. Weiss (PTB) and G. Winkler (IRK) which all are gratefully acknowledged.

unconverted photons (see also Section 14). As the efficiency  $\varepsilon_7$  cannot be measured directly, the tracer ( $^{134}\text{Cs}$ ) is added quantitatively to produce mixture sources, and it is assumed that  $\varepsilon_7$  can be approximated as a function (usually a polynomial) of the inefficiency  $1 - \varepsilon_4$ ,

$$\varepsilon_7 = f(1 - \varepsilon_4) = f\left(1 - \frac{N_c}{N_\gamma}\right) = 1 + c_1\left[1 - \frac{N_c}{N_\gamma}\right] + c_2\left[1 - \frac{N_c}{N_\gamma}\right]^2 + \dots$$

The quantity  $\varepsilon_4 = N_c/N_\gamma$  can be readily measured with a mixture source, provided the  $\gamma$  photons from the  $^{137}\text{Cs}$  are not counted in the  $\gamma$  channel. The contribution of the tracer activity to the observed  $\beta$ -count rate is then given by

$$N_4 = \frac{m_4 a_4}{D_4} g(1 - \varepsilon_4) ,$$

$$\text{with } g(1 - \varepsilon_4) = 1 + d_1\left[1 - \frac{N_c}{N_\gamma}\right] + d_2\left[1 - \frac{N_c}{N_\gamma}\right]^2 + \dots$$

Here  $d_1, d_2, \dots$  must be obtained from separate measurements with pure  $^{134}\text{Cs}$  sources, in order to find the tracer-activity concentration  $a_4$ . Since  $N_7 = N_{\text{tot}} - N_4$ , where  $N_{\text{tot}}$  is the total  $\beta$ -count rate from a mixture source, eq. (1) can be expressed as

$$\frac{D_7}{m_7} \left[ N_{\text{tot}} - \frac{m_4 a_4}{D_4} g\left(1 - \frac{N_c}{N_\gamma}\right) \right] = a_7 \left[ f\left(1 - \frac{N_c}{N_\gamma}\right) + C \right] , \quad (2)$$

whence

$$\frac{D_7}{m_7} \left\{ N_{\text{tot}} \frac{N_\gamma}{N_c} - \frac{m_4 a_4}{D_4} \frac{g\left(1 - \frac{N_c}{N_\gamma}\right)}{N_c/N_\gamma} \right\} = a_7 \left\{ \frac{f\left(1 - \frac{N_c}{N_\gamma}\right)}{N_c/N_\gamma} + C \frac{N_\gamma}{N_c} \right\} .$$

By introducing slightly different polynomials  $g_1$  and  $f_1$ , this may be written as

$$\frac{D_7}{m_7} \left\{ N_{\text{tot}} \frac{N_\gamma}{N_c} - \frac{m_4 a_4}{D_4} g_1\left(\frac{1 - \frac{N_c}{N_\gamma}}{N_c/N_\gamma}\right) \right\} = a_7 \left\{ f_1\left(\frac{1 - \frac{N_c}{N_\gamma}}{N_c/N_\gamma}\right) + C \left[ 1 + \frac{1 - \frac{N_c}{N_\gamma}}{N_c/N_\gamma} \right] \right\} . \quad (3)$$

The functions  $f_1$  and  $g_1$  are almost identical with  $f \cdot N_\gamma/N_c$  and  $g \cdot N_\gamma/N_c$ , respectively, if second- and higher-order terms are dropped.

In fact, if one puts

$$f_1 \left( \frac{1 - \frac{N_c}{N_\gamma}}{N_c/N_\gamma} \right) = 1 + c_{11} \left[ \frac{1 - \frac{N_c}{N_\gamma}}{N_c/N_\gamma} \right] = \frac{N_\gamma}{N_c} \left\{ 1 + c_1 \left[ 1 - \frac{N_c}{N_\gamma} \right] \right\},$$

it follows that  $c_{11} = 1 + c_1$ , and similarly  $d_{11} = 1 + d_1$ . In practice, however,  $f_1$  and  $g_1$  are also experimentally fitted functions for which higher-order polynomials may be used. Thus a polynomial extrapolation of the left-hand side of eq. (2) or (3), in terms of

$$1 - \frac{N_c}{N_\gamma} \text{ or } \frac{1 - \frac{N_c}{N_\gamma}}{N_c/N_\gamma}, \text{ to the limit } N_c/N_\gamma \rightarrow 1, \text{ will give } a_7[1 + C].$$

This general method therefore extrapolates the  $^{137}\text{Cs}$   $\beta$ -count rate, per unit mass at reference time, the contribution from the  $^{134}\text{Cs}$  having been subtracted using the known, separately measured quantities  $a_4, d_1, d_2, \dots$ .

The approximation is sometimes made that  $D_7$  and  $D_4$  are constant for a series of measurements in which case the polynomials in eq. (2) and (3) can be combined as

$$N_{\text{tot}} = N_{\text{tot}}^0 \left\{ 1 + r_1 \left[ 1 - N_c/N_\gamma \right] + r_2 \left[ 1 - N_c/N_\gamma \right]^2 + \dots \right\} \quad (4)$$

and

$$\frac{N_{\text{tot}} N_\gamma}{N_c} = N_{\text{tot}}^0 \left\{ 1 + r_3 \left[ \frac{1 - N_c/N_\gamma}{N_c/N_\gamma} \right] + r_4 \left[ \frac{1 - N_c/N_\gamma}{N_c/N_\gamma} \right]^2 + \dots \right\}, \quad (5)$$

where  $N_{\text{tot}}^0 = \frac{m_7 a_7}{D_7} (1 + C) + \frac{m_4 a_4}{D_4}$ , and  $r_1, \dots, r_4$  are constants.

Thus, a polynomial extrapolation of  $N_{\text{tot}}$  in terms of  $1 - N_c/N_\gamma$  (or of  $\frac{N_{\text{tot}} N_\gamma}{N_c}$  in terms of  $\frac{1 - N_c/N_\gamma}{N_c/N_\gamma}$ ) to the limit  $N_c/N_\gamma \rightarrow 1$  will give  $N_{\text{tot}}^0$ ,

whence

$$a_7(1 + C) = \frac{D_7}{m_7} \left[ N_{\text{tot}}^0 - \frac{m_4 a_4}{D_4} \right]. \quad (6)$$

This extrapolation is not fully correct because the ratio  $D_7/D_4$  decreases with time and, therefore, differs for the various values of  $\epsilon_4$ . However, it can be shown that for a mixture source of  $^{137}\text{Cs} + ^{134}\text{Cs}$  and measurements made over 12 days, with uniformly decreasing efficiencies from 95 % to 65 %, this approximation will introduce an error in  $a_7$  of the order of 0.05 % only.

Another approximation is sometimes made by putting  $\frac{m_4 a_4}{D_4} \epsilon_4$  instead of  $\frac{m_4 a_4}{D_4} g(1 - \epsilon_4)$  for the tracer count rate. In order to illustrate this, let us rewrite eq. (2) as

$$N_{\text{tot}} = \frac{m_7 a_7}{D_7} [f(1 - \epsilon_4) + C] + \frac{m_4 a_4}{D_4} [\epsilon_4 - \epsilon_4 + g(1 - \epsilon_4)]$$

and rearrange it to the form

$$\left[ N_{\text{tot}} - \frac{m_4 a_4}{D_4} \epsilon_4 \right] \frac{D_7}{m_7} = \frac{m_4 a_4}{m_7} \frac{D_7}{D_4} [g(1 - \epsilon_4) - 1 + (1 - \epsilon_4)] + a_7 [f(1 - \epsilon_4) + C].$$

The left-hand side of this equation will extrapolate to  $a_7[1 + C]$  only if  $D_7/D_4$  is constant.

If more than one source is included in a common adjustment, individual corrections for decay and dilutions of the two components have to be applied. Therefore, some participants preferred to carry out separate extrapolations for each source and to calculate a weighted mean of the results.

## 10. Coincidence correction formulae

Before applying any extrapolation equations, the participants corrected the observed count rates  $N_{\text{tot}}$ ,  $N_\gamma$ ,  $N_c$  for background, dead times and, where necessary, resolving times using various published coincidence formulae [21-25]. The formulae are not listed in detail since the uncertainties in the corrections involved are relatively small.

## 11. Beta-gamma counting without coincidences

An alternative solution is offered by the method called "selective sampling" [26] which yields directly the value of  $\epsilon_\beta$ . This new approach is particularly useful when it can rely on the availability of a rapid speed converter [27]. The BIPM results of this comparison are based exclusively on this method which had already been applied in the 1978/79 comparison [6], but with less precision, since the new rapid speed converter was not yet available. It may be remembered that the selective-sampling method determines the rate of those  $\gamma$  events which have no partner in the  $\beta$  channel, with respect to that of all the registered  $\gamma$  events. Therefore, no coincidence mixer is needed and accidental coincidences can be disregarded completely.

Another technique which benefits from a similar advantage is "live-timed anti-coincidence counting with extended dead-time circuitry" [28]. In the present comparison, this method was applied by NBS and NRC.

## 12. Detailed counting data

Most of the numerical information concerning coincidence counting is presented in Table 9. It is noteworthy that the  $\beta$ -count rates seldom exceeded  $20\,000\text{ s}^{-1}$ . Due to relatively narrow  $\gamma$ -channel gates, the  $\gamma$ -count rates were low and called for rather long counting times. However, thanks to the long half lives and to the high purity of the solutions of the two Cs isotopes, this had only little effect on the overall uncertainty.

## 13. Efficiency extrapolation

The fitting of a polynomial to the experimental data has become a routine operation for which most laboratories have developed suitable computer programs. A detailed discussion of the procedure has been given in earlier reports [9, 29] and will not be repeated here.

As the intercepts and slope-to-intercept ratios depend too strongly on experimental details to be of much interest, it was felt that the only fitting data worth mentioning are the order of the polynomial, the number  $\nu$  of degrees of freedom and the "reduced"  $\chi^2$  (sum of the squared residuals divided by  $\nu$ ) of the adjustment. They may be found in Table 10.

First- and second-order polynomials were found adequate in about equal numbers of cases. The fourth order preferred by PTB is to be understood as that of a polynomial  $y = a_0 + a_1x + a_4x^4$ , with  $a_2 = a_3 = 0$ , which gave a better fit of their data near to the intercept. The justification of this choice lies in the fact that the absorption curve for a  $\beta$  radiation is practically exponential for low values of absorber thickness. Thus, after power-series development and maintaining only first-order terms, the inefficiencies for two  $\beta$  radiations become proportional to each other, and the efficiency curve is completely linear for higher  $\varepsilon_\beta$  values.

Some information about the goodness of fit can be obtained by calculating  $\chi^2 \equiv \frac{1}{\nu} \sum_{i=1}^n \frac{1}{s_i^2} [y_i - y(x_i)]^2$ , where  $(y_i, x_i)$  are the experimental data points,  $y(x_i)$  the fitted values and  $s_i$  the standard deviations of the measurements. This value  $\chi^2$  should be close to unity if the actual deviations from the fitted curve are "normally" distributed about the curve with a standard deviation compatible with  $s_i$ . The participants were asked to supply graphs of  $y(x)$  and of the residuals. Since these graphs were widely different in quality and format, we decided to reproduce only a few examples (Fig. 3).

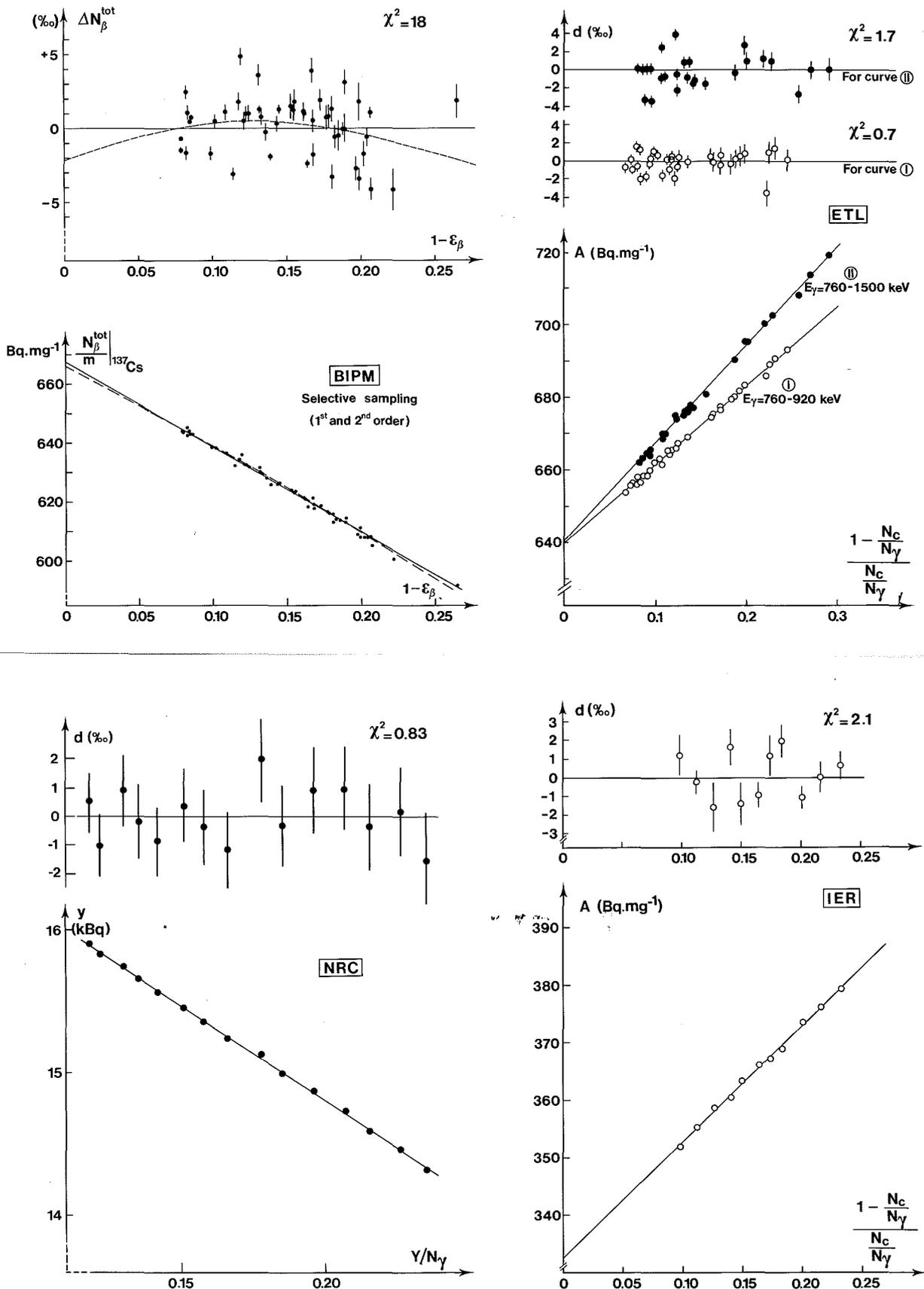


Fig. 3 - Four examples of graphs of efficiency functions. The lower part shows the experimental data points and the adjusted polynomial. The upper part represents the relative deviations from the adjusted line (residuals) and their standard deviations.



As was indicated in the reporting form, all the uncertainty components were to be considered as approximations of the corresponding standard deviations and were to be added in quadrature, with reference to recent recommendations [30]. Although the discussion of this subject is not yet definitely closed, we believe that these recommendations mark a considerable progress and help to unify the presentation of uncertainties.

## 16. Final results

The values of the activity concentration and their combined uncertainties, both taken on the reference date (1982-05-01, 00 h UT), are reported in Table 10. A comparison with the values given in ref. [7] will show that the results from BARC were included later and that three participants wished to apply corrections for the following reasons:

IFIN replaced first-order extrapolation by second order,  
 OMH " second-order " " first " ,  
 UVVVR found inconsistencies in the extrapolation procedure.

The total spread of all the "coincidence" results is 2.4 %. Some of the combined uncertainties appear to be too small compared with such a spread. Therefore, a weighted mean value of the activity concentration would be biased; an unweighted mean seems more adequate. The result obtained by PDS should be excluded from the mean because it cannot be regarded as an absolute measurement. For the four participants who reported more than one result weighted means were calculated. The unweighted mean of 18 results turned out to be

Activity concentration of the  $^{137}\text{Cs}$  solution =  $(604.1 \pm 4.0) \text{ kBq g}^{-1}$ .

The fact that fifteen results (72 %) differ from this mean by less than one standard deviation is compatible with a normal distribution. The relative standard deviation (0.66 %) is slightly larger than that of the previous comparison [6] which was found to be 0.53 %.

After completion of this report, two participants asked us to include the following modifications of their results.

BARC: 608.4 instead of 605.4  $\text{kBq g}^{-1}$  (for omission of the  $\beta$ -branching ratio of  $^{137}\text{Cs}$ )  
 NIM : 602.5 " " 597.4 " from a first-order fit to 70 data points measured by using a  $\gamma$ -channel window from 100 to 600 keV.

However, neither the standard deviation of the mean nor the data in Fig. 4 have been corrected.

Thanks to the international reference system (SIR), it is now possible to compare three groups of activity measurements of  $^{137}\text{Cs}$  with each other, namely

- 10 results from independent measurements carried out from 1976 to 1980 by eight participants in SIR,
- 10 results from the small comparison of 1979/80, and
- 19 results from the present comparison.

All these results are represented in Fig. 4 using a common scale of relative deviations from the mean value quoted above. The 1979/80 comparison is considered as belonging to the SIR results shown on the left of Fig. 4. Its mean value is slightly higher by  $+ (0.12 \pm 0.53) \%$  than that of 1982. It will be noted that the three groups have very similar spreads and (estimated) uncertainties.

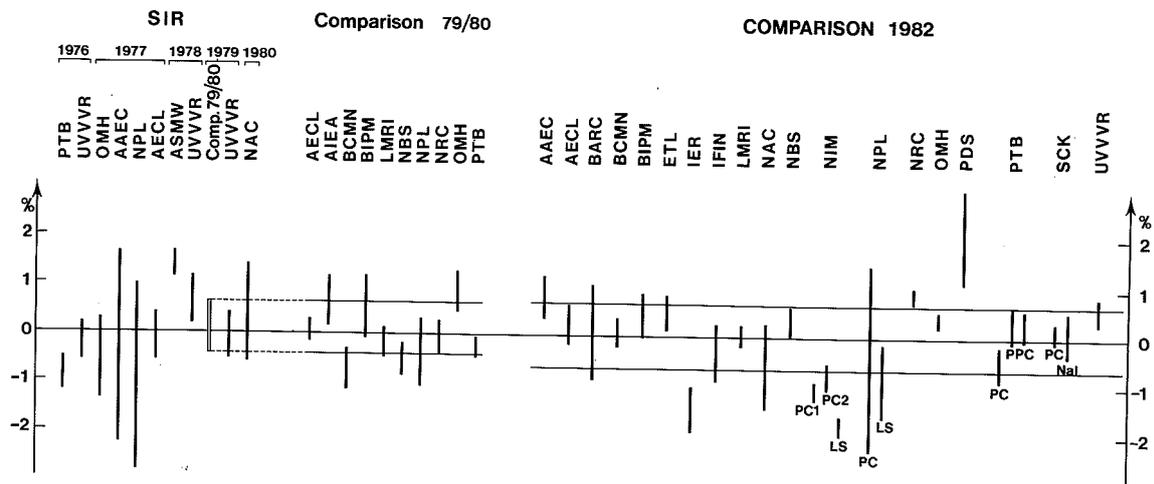


Fig. 4 - Graphical representation of the results and their combined uncertainties. The type of detector is indicated only for the four laboratories having submitted more than one result. The mean values of the two comparisons are aligned by means of the corresponding SIR results. Their respective standard deviations are represented by pairs of horizontal lines.

## 17. Conclusion

Efficiency tracing by  $^{134}\text{Cs}$  and extrapolation to 100 %  $\beta$ -counting efficiency proved to be an adequate technique for the activity measurement of  $^{137}\text{Cs}$ .

The effect of conversion electrons and photons on the  $\beta$ -ray detector can be accounted for with low enough uncertainty, provided the  $\gamma$  efficiency of the  $\beta$  detector is low and accurately known.

Uncertainties have been stated and treated according to recent BIPM recommendations which provided useful guidance. Most of the participants estimated the fitting procedure for the efficiency function to give the largest contribution to the combined uncertainty.

The fractional activity remaining adsorbed on the walls of the emptied and rinsed ampoules, of the order of  $3 \cdot 10^{-4}$ , was found to be negligible.

The total range of the results of 2.4 % and the standard deviation of 0.66 % are considerably larger than has become usual in recent comparisons concerning easier radionuclides. However, the mean value is in close agreement with the mean of a previous comparison of  $^{137}\text{Cs}$  and with the corresponding average result of independent SIR measurements. The estimated uncertainties are in general compatible with the deviations from the mean value.

Table 1 - List of the participants

		<u>Names of the persons who carried out the measurements</u>
AAEC	Australian Atomic Energy Commission, Lucas Heights, Australia	D.F. Urquhart, V. Page, H.A. Wyllie
AECL	Atomic Energy of Canada Limited, Chalk River, Canada	J.S. Merritt, L.V. Smith, R.H. Martin
BARC	Bhabha Atomic Energy Research Centre, Bombay, India	P.K. Srivastava, S. Kamboj
BCMN	Central Bureau for Nuclear Measurements, Euratom, Geel, Belgium	D. Reher, E. Celen, R. Vaninbroukx, W. Zehner
BIPM	Bureau International des Poids et Mesures, Sèvres, France	C. Veyradier, P. Bréonce, C. Colas
ETL	Electrotechnical Laboratory, Ibaraki, Japan	Y. Kawada
IER	Institut d'électrochimie et radiochimie de l'EPFL, Lausanne, Switzerland	J.-J. Gostely, P. Conte
IFIN	Institute of Nuclear Physics and Engineering, Bucharest, Romania	M. Sahagia, L. Grigorescu
IMRI	Laboratoire de Métrologie des Rayonnements Ionisants, Saclay, France	J. Bouchard
NAC	National Accelerator Centre, Faure, South Africa	J. Steyn
NBS	National Bureau of Standards, Washington, D.C., USA	B.M. Coursey, A.T. Hirshfeld
NIM	National Institute of Metrology, Beijing, China	Yu Kuí-fang, Zhōu Kè-qín, Li Zuo gán, Ma. G.H., Yang. Y.D., Wang Z.Y., Wang Z.Y., Zhang. Y.Q., Wu X.Z.
NPL	National Physical Laboratory, Teddington, UK	C.E. Grant, D.H. Makepeace, M.J. Woods, D. Smith
NRC	National Research Council, Ottawa, Canada	K. Munzenmayer, G.C. Bowes, A.P. Baerg
OMH	Országos Mérésügyi Hivatal, Budapest, Hungary	Á. Szörényi, A. Zsinka, M. Csikós
PDS	National Atomic Energy Agency, Jakarta, Indonesia	H. Arumbinang
PTB	Physikalisch-Technische Bundesanstalt, Braunschweig, FRG	K.F. Walz, E. Funck
SCK	Studiecentrum voor Kernenergie, Mol, Belgium	C. Ballaux, P. Willeborts
UVVVR	Institute for research, production and application of radioisotopes, Prague, CSSR	J. Plch, V. Zajíc

Table 2 - Activity concentration of the tracer solution  
as determined by some participants

Laboratory	Method used	Activity concentration (kBq g <sup>-1</sup> ) on the reference date
AECL	4πγ ionization chamber	639.0 ± 2.0
ETL	4πβ-γ	638.7 ± 0.9
NPL	4πβ-γ 745 < E <sub>γ</sub> < 905 keV 745 < E <sub>γ</sub>	638.9 ± 0.3 633.40 ± 0.25
NRC	4πβ(PPC)-γ AC	638.0 ± 0.3
OMH	4πβ-γ	638.6 ± 0.7
PDS	Ge(Li)	638.1 ± 4.7
SCK	4πγ NaI well crystal	639.5 ± 1.0

Table 3 - Results of ionization-chamber measurements and adsorption tests

Laboratory	Activity concentration at reference date (kBq g <sup>-1</sup> )	Activity remaining in the "empty" ampoule after 2 rinsings with distilled water (Bq)
AAEC	603 ± 12	780
AECL	607.1 ± 1.0	790
BCMNI	-	900
BIPM	604.9 ± 0.4	7 430*
IER	-	612
IFIN	-	0.08
LMRI	604.2 t	-
NAC	603	390
NBS	603.3 t	535 137Cs 532 134Cs
NIM	596.8 ± 1.8	-
NPL	593.7 t	1 100 137Cs 900 134Cs
NRC	-	500
OMH	601.8	580
PTB	605.09	500
SCK	-	357
UVVVR	609.6 t	220

t transfer to own type of ampoule

\* rinsings took place when the ampoule was fully dry

Table 4 - Source preparation for proportional counting

Laboratory	Were solutions diluted?	Activity ratio $^{137}\text{Cs}/^{134}\text{Cs}$	Range of source mass (mg)	Source backing*			Drops dispensed onto metal?	Wetting or seeding agent	Special treatment	Drying	Numbers of sources		
				Substrate Metal coating	Number of films metal layers	Total mass ( $\mu\text{g cm}^{-2}$ )					prep.	used	
AAEC	yes	0.48	23 - 63	VYNS, Au	1	1	30	yes	Ion exch. resin	Electro-sprayed	Oven	10	8
AECL	no	1.18	12 - 27	VYNS Au + Pd (20%)	1	1	12	no	Catanac SN	Extra carrier	Dry air 45 °C	16	15
BARC	no	≈ 1	17 - 52	VYNS, Au	2	2	40	no	dil. teflon suspension	-	Open air, then lamp	22	19
BCMNI	no	0.6 - 1.4	7 - 15	VYNS, Au	1	2	50	yes	Catanac, Ludox	-	Lamp	13	10
BIPM	no	1.08	26 - 96	VYNS, Au	2	2	60	no	Ludox SM $10^{-4}$	-	Open air	60	10
ETL	no	1.0	9 - 19	VYNS, Au	1	2	30	yes	Ludox SM30	-	Dry box	30	25
IER	yes	1.0	20 - 70	VYNS, Au	1	2	50	yes	Ludox SM $10^{-4}$	-	Open air	20	19
IFIN	no	0.64	5 - 11	VYNS, Au	1	2	125	yes	Ludox SM	-	Open air	18	17
LMRI	no	1.36; 0.55	15 - 27	Cellulose, Au	1	2	40	yes	Insulin	-	Open air	12	10
NBS	yes	1.5; 0.67	21 - 34	Collodion, Au	2	1	30	yes	Ludox	-	Open air	16	6
NIM	yes	0,92; 1.03	7 - 13	VYNS, Au	1	1; 2	20	no	Sil. collosol, Catanac SN	-	Open air	25	19
NPL	no	0.94	14 - 15	VYNS, Au	1	2	30	yes	Catanac $2 \cdot 10^{-4}$	-	Open air	38; 25	30; 19
NRC	no	0.95	13 - 16	VYNS, Au + Pd	1	2	40	yes	Catanac SM	-	Air 40 °C	16	16
OMI	no	0.5; 1	3 - 16	VYNS-3, Au + Pd	1	2	30	yes	Ludox + Teepol	-	Lamp	40	37
PTB	no	0.97	5 - 60	VYNS, Au + Pd	1	2	60	yes	Ludox SM $10^{-4}$ (6 sources)	Electro-sprayed resin (8 s.)	Air	14	13
SCK	no	0.85	4 - 9	VYNS, Au	2	2	50	yes	Ludox SM $10^{-4}$	CsCl; H <sub>2</sub> O	Open air	23	23
UVVVR	yes	0.55	9 - 18	VYNS, Au	3	2	45	yes	Catex-anex Ludox + Aquadak	**	-	31	31

\* before adding further absorbers

\*\* during 20 h diffusion in water, before drying of sources with Catex-anex

Table 5 - Proportional counters used by the participants

Laboratory	Wall material	Height each half (mm)	A n o d e					Gas	Pressure (MPa)	Discri- mination level (keV)	Remarks
			Wire	Diameter (μm)	Length (mm)	Dist. from source (mm)	Voltage applied (kV)				
AAEC	See remarks	25	Pt	50	35	12	3.35	M (methane)	0.1	-	Al and gold-coated Epoxy resin
AECL	Stainless steel	21	stainl.st. or W	15	36	10	2.4	M	0.1	≈ 0.1	
BARC	Al	25	stainl. steel	12	40	12.5	2.7	LPG (see rem.)	0.1	0.2	Liquefied petroleum gas, contains Butane and Isobutane
BCMN	Perspex, Al coated	17.5	stainl. steel	50	59	10	3.0	M	0.1	0.5; 1.0	
BIPM	Brass, Au plated	20	stainl. steel	50	47	11	3.7	M	0.1	0.06	
ETL	Brass, Au coated	20	stainl. steel	50	80	10	3.4	M	0.1	0.2	
IER	Al	25	Au	100	34	12.5	3.6	M	0.1	≈ 1	
IFIN	Brass	24	W	20	40	11	3.2	M	0.1	1	
LMRI	Perspex + Au	22	stainl. steel	2	80	10	2.75	M	0.1	≈ 0.1	
NBS	Al 6061	28.3	stainl. steel	51	53	14	6.6 to 7.2	Ar + M	1.48	1 to 6	
NIM	Al	20	Constantan	25	35	10	2.45	M	0.1	?	
	Brass	36	stainl. steel	20	70	18	2.9	M	0.1	0.45	
NPL	Perspex	14	P bronze	75	75	8	2.15	Ar + M	0.1	0.23	
	Cu + Ag coated										
NRC	Al	25.4	stainl. steel	25	38	12.7	3.8	Ar + M	1.136	1 to 40	
OMH	Al	20	stainl. steel	30	40	10	2.4	M	0.1	0.7	
PTB	Al	22.5	stainl. steel	50	30	12	3.7	M	0.1	0.5	γ-chanel settings (keV) } 750 - 880 760 - 900 1 250 - 1 600
	Al	20	" "	30	40	10	4.5	Ar + M	1.1	> 0.5	
	Al	20	W, Au coated	100	34	10	7.4	Ar + M	1.1	0.5	
SCK	Brass, Au coated	20	stainl. steel	50	55	11	3.4	M	0.1	0.3	
UVVVR	Stainl. steel	18	Mo, Au coated	50	55	9	3.5	M	0.1	0.36	

Table 6 - Gamma-ray detectors, dead time, resolving times, etc.

Laboratory	Gamma-ray detector							Dead times*				Coincidence resolving time* ( $\mu$ s)	"Gandy effect"	
	Crystal	Number	Diam. (mm)	Length (mm)	Phototube	Year of purchase	Resolution at 662 keV (%)	( $\mu$ s)		Delay mismatch* ( $\mu$ s)	Effect on final result (%)			
								$\tau_{\beta}$	$\tau_{\gamma}$					
AAEC	NaI	1	75	75	EMI 9531 B	1964	8.8	9.15 (8)	18.2 (1)	1.145 (6)	0.000 (15)	0.03		
AECL	NaI	2	76	76	75 B01 (SRC)	1976/77	7.3; 7.6	1.935 (21)	1.916 (20)	0.698 3 (19)	0.040 (3)	0.002		
BARC	NaI	1	76	76	RCA 6363	1962	11	6.18 (2)	6.22 (6)	2.305 (10)	-	-		
BCMN	NaI	1	75	75	SRC 75 B01-2	1978	6.4	5.97 (6)	6.09 (6)	0.785 (16)	0.00 (5)	0.06		
BIPM	NaI	1	76	76	Harshaw 12S	1978	7.5	4.432 (10)	4.425 (10)	1.06 (1)	0.000 (15)	nil		
ETL	NaI	2	76	76	RCA 6342 A	1970	7.5 to 8	4.35 (5)	2.11 (5)	0.688 7 (28)	0.0 (1)	nil		
IER	NaI	1	76	76	RCA 4524	1977	9.6	3.201 (1)	3.196 (1)	1.075 (2)	0.000 (1)	nil		
IFIN	NaI	1	76	76	EMI-9708 E	1976	12.8	10.0 (5)	10.0 (5)	1.09 (5)	electronic compens.	nil		
LMRI	Ge(Li)	1	-	-	-	-	2 keV at 1.33 MeV	5	5	$\approx 1$	< 0.005 jitter corr.	-		
NAC	NaI	1	76	76	9531 KA	1970	9.5	1.15 (2) 1.31 (3)	1.19 (3)	0.514 (2)	-	-		
NBS	NaI	2	76	76	Harshaw	1979	7.1	4.3 extend.	10.0 (2)	-	-	-		
NIM	NaI	1	76	76	EMI-9758 K	1980	7.8	21.901 (1)	21.869 (1)	1.272 8 (1)	0.03 $\pm$ 0.01	0.05		
			75	75	EMI-9758	1981	8	5.98 (1)	1.52 (1)	1.525 (6)	< 0.15	< 0.15		

\* in parentheses: uncertainty in units of last digit

Table 6 (cont'd)

Laboratory	Gamma-ray detector							Dead times*				Coincidence		"Gandy effect"		
	Crystal	Number	Diam. (mm)	Length (mm)	Phototube	Year of purchase	Resolution at 662 keV (%)	(μs)				Coincidence resolving time* (μs)	Delay mismatch* (μs)	Effect on final result (%)		
								$\tau_{\beta}$		$\tau_{\gamma}$						
NPL	NaI	2	100	100	EMI-9758 L	1981	8.3	1.54	(1)	2.8	(5)	0.591	(5)	0.00	(1)	nil (PC)
	NaI	1	100	100	-	1960	9.8	18.7	(5)	3.27	(4)	0.705	(5)	0.000	(3)	(PC)
	(well)		25	40)						1.97	(2)	0.300	(5)	jitter	small	
NRC	NaI	1	76	76	RCA 8054	1965	8.45	5.08	(5),	common		-		Not applicable		
	Ge(Li)	1	-	-	-	-	0.45	extending $\tau$ for $\beta$ and $\gamma$						(AC counting)		
OMH	NaI	1	76	76	RCA 8054	1977	8.2	3.067	(5)	3.021	(5)	1.022	(10)	+ 0.035	(15)	0.02
PTB	NaI	1	100	100	EMI 9791 B	1977	6.6	5.06	(5)	4.73	(5)	1.01	(2)	0.00	(5)	0.05 (PC)
	NaI	1	76	76	EMI 9758	1979	6.6	5.02	(5)	5.01	(5)	1.03	(2)	0.00	(5)	0.05 (PC1)
	NaI	1	152	152	RCA 8055	1979	8.9	5.00	(2)	5.00	(2)	1.000	(3)	0.10	(2)	0.06 (PC2)
	(well)		50	100)												
SCK	NaI	1	102	102	Dumont 6363	1967	8.8	2.48	(1)	2.46	(1)	1.014	3 (3)	- 0.026	(4)	0.016
UVVVR	NaI	2	76	76	Canberra 802-4	1979	7.3	5.724	(3)	5.710	(3)	0.992	(4)	0.000	(15)	nil
										5.732	(3)	0.978	(4)			

\* in parentheses: uncertainty in units of last digit

Table 7 - Liquid-scintillation counting

Laboratory	Counting vessel	Phototubes		Spurious pulses		Composition of scintillating solution, source preparation	Sources	
		number		precautions taken	upper limit of count rate (%)		prepared	used
NAC	glass 20 cm <sup>3</sup>	RCA 8850	2 in coinc.	measured	0.07	Instagel (Packard)	11	11
NIM	glass 8 cm <sup>3</sup>	GDB 52-LD	1	threshold and $\tau_{\beta} \approx 21 \mu s$	0.05	75% toluene, 25% ethyl alcohol 5 g of PPO + 0.5 g of POPOP per l of solution	15	13
NPL	glass 13 cm <sup>3</sup>	RCA 31 000 D 1 (development tube of RCA 8850)	1	measured, $\epsilon_{\beta} \leq 0.925$	0.05	100 ml Toluene 2 g BIEUQ 0.05 g BLSMSB 5 g UNIBLEND (Koch-Light Ltd.) 0.6 g of carrier solution (CsCl in 0.1 molar HCl)  15 or 30 mg of active solutions were added to 7 cm <sup>3</sup> of this cocktail.	10	10

Table 8 - Variables of the efficiency functions used by the participants

	x	y
AAEC	$1 - \frac{(N'_c - N_{acc})[1 - \tau_\beta(N_c - N_{acc})]}{N'_\gamma(1 - \tau_\beta N'_\beta)}$	$(1/m) \left( \frac{N'_\beta}{1 - \tau_\beta N'_\beta} - B_\beta \right)$
AECL	$(1 - N_c/N_\gamma)/(N_c/N_\gamma)$	$N_\beta N_\gamma / (m N_c)$
BARC	$(1 - \epsilon_\beta)/\epsilon_\beta$	$N_\beta/m$
BCMN	$1 - N_c/N_\gamma$	$N_\beta$
BIPM	$1 - \epsilon_\beta$	$N_\beta^{tot}$
ETL	$\frac{1 - N_c/N_\gamma}{N_c/N_\gamma}$	$N_\beta N_\gamma / (m N_c)$
IER	"	$(1/m) \left( \frac{N_\beta N_\gamma}{N_c} - A_{134} \right)$
IFIN	$1 - N_c/N_\gamma$	$N_\beta/m$
LMRI	$(1 - \epsilon_\beta)/\epsilon_\beta$	?
NAC	$N_\gamma/N_c$	$N_\beta N_\gamma / N_c$
NBS	$(1 - \epsilon)/\epsilon; \quad \epsilon = (1 - Y/N_\gamma)$ Y = observed anti-coinc. rate	$N_\beta / (1 - \epsilon)$
NIM	$1 - \epsilon_\beta; (1 - \epsilon_\beta)/\epsilon_\beta; I_o/I_n^*$	$(N_\beta - \epsilon_\beta A_{134})/m; N_\beta N_\gamma / N_c; N$
NPL	$1 - N_c/N_\gamma$	$1/m_{137} (N_\beta^{m_{134}} A_{134} f(\epsilon_\beta))$ , see also eq. (3) in Section 9
NRC	$Y/N_\gamma$	$N_\beta^{tot}$
OMH	$\frac{1 - N_c/N_\gamma}{N_c/N_\gamma}$	$N_\beta N_\gamma / (m N_c)$
PDS	-	-
PTB	$1 - N_c/N_\gamma$	$\frac{N_\beta (m_{134} + m_{137})}{m_i m_{137}} - \frac{m_{134}}{m_{137}} \epsilon_\beta A_{134}$
	$\frac{1 - N_c/N_\gamma}{N_c/N_\gamma}$	$\frac{N_\beta N_\gamma m_{134} + m_{137}}{N_c m_i m_{137}} - \frac{m_{134}}{m_{137}} A_{134}$
		$m_i = \text{mass of the source number } i$
SCK	$1 - N_c/N_\gamma$	$N_\beta^{tot}/m$
UVVVR	$(1 - \epsilon_\beta)/\epsilon_\beta; 1 - \epsilon_\beta$	$N_\beta N_\gamma / N_c; N_\beta/m$

\*  $I_n$  = anode current with optical filter,  $I_o$  = current without filter

Table 9 - Count rates and related data

Laboratory	Method (beta-ray detector)	$\gamma$ -channel setting (keV)	Typical count rates ( $s^{-1}$ )			Background rates ( $s^{-1}$ )			Number of data points; time per point (s)	Range of $\epsilon_{\beta}$ (%)	Method for varying $\epsilon_{\beta}$	Time of the measurements
			$N_{\beta}$	$N_{\gamma}$	$N_c$	$B_{\beta}$	$B_{\gamma}$	$B_c$				
AAEC	PC	795 - 900	7 000	75	65	1	0.8	0.002	50; 1 700	95 - 85	Changing foil thickness	June-August
		755 - 1 445	7 000	150	140	1	5	0.015	20; 1 200	94 - 83		
		1 250 - 1 500	7 000	0.6	0.45	1	0.2	0.001	10; 4 000	86 - 68		
AECL	PC	740 - 1 510	12 900	630	560	0.4	1.9	0.02	168; 1 000	91 - 78	Extra carrier (3 sources) Catanac (5 " )	May 14-20
		740 - 900	12 900	450	410	0.4	0.6	< 0.01	202; 1 000	93 - 81		" 18-20
BARC	PC	1 250 - 1 610	37 000	55	40	0.77	0.5	0.002	45; 3 600	79 - 38	By adding inactive carrier	August 9-Sept. 21
		680 - 1 700	36 750	274	208	0.77	4.75	0.12	46; 2 400	82 - 45		
BCMN	PC	> 750	14 000	500	400	0.3	2.4	0.2	9 to 11	93 - 69	Foil absorption	June 29-July 16
		> 730	to 19 000	to 800	to 600				per source			
BIPM	PC	700 - 900	30 000	800	670	1.35	2.1	0.17	51; 7 000	91 - 80	Foil absorption	June 30-July 23
ETL	PC	760 - 930	7 000	150	135	4.19	2.78	0.052	34; 2 100	94 - 80	Addition of carrier; metallized VYNS foils	June 8-18
		760 - 1 500	7 000	250	225	4.19	9.48	0.13	28; 2 100	92 - 77		" "
IER	PC	785 - 890	16 800	190	168	2.9	0.92	0.01	600; 100	94 - 78	Gold-coated VYNS	June 29-July 7
		785 - 1 600	18 000	293	240	3.9	4.4	0.03	580; 100	90 - 75		July 13-22
IFIN	PC	750 - 980	8 000	150	120	6	2.5	0.01	49; 1 000	91 - 64	Metallized VYNS, Mylar	June 3-30
LMRI	PC	760 - 820	14 000	40	36	2.3	0.6	0	22; 3 000	93.5-76	Foils	July 1
NAC	LS	> 950	32 575	197	161	9.3	3.3	0.2	11; 1 000	83 - 72	Pulse-height selection	May 13
NBS	PPC	755 - 870	6 000	155	14-30	1.0	0.7	AC 0.7	5-15; 3 000	92 - 75	Vary threshold discr. level	May 10-August 5
		> 890	6 000	170	17-30	1.0	4.7	AC 4.7	per source			

Table 9 (cont'd)

Laboratory	Method (beta-ray detector)	$\gamma$ -channel setting  (keV)	Typical count rates ( $s^{-1}$ )			Background rates ( $s^{-1}$ )			Number of data points; time per point (s)		Range of $\epsilon_{\beta}$ (%)	Method for varying $\epsilon_{\beta}$	Time of the measurements
			$N_{\beta}$	$N_{\gamma}$	$N_c$	$B_{\beta}$	$B_{\gamma}$	$B_c$					
NIM	PC1	769 - 952	11 695	328	226	1.5	6.7	0.004	70;	900	95 - 80	Foil absorption VYNS foils	July 25-Aug. 23
	PC2	740 - 867	8 000	90	80	1	0.1	0.003	190;	900	97 - 80		July 19-Aug. 28
NPL	PC	745 - 905	15 000	850	700	2	1	0.03	34;	1 000	83 - 70	Addition of VYNS and Al foils	June 30-July 8
		745	15 000	2 200	1 600	2	10	0.5	28;	1 000	78 - 63		
	LS	770 - 920	4 300	75	65	2.9	0.28	0.002	35;	1 000	92.5-77	Computer discrimination	June 30-July 2
		1 230 - 1 630	4 300	48	41	2.9	0.49	0.002	35;	to	89 - 68		
		1 850 - 2 200	4 300	2.7	2.0	2.9	0.08	0.002	35;	2 000	76 - 35		
NRC	PPC Ge(Li)	780 - 810	18 000	55	(AC)	0.5	0.058	(AC)	240;	700	91 - 73	$\beta$ -threshold variation (Note: Two $\gamma$ -ray channels for 2-param. data fit)	May 3-10
	Ge(Li)	780 - 810	18 000	55	5-15	0.5	0.058	0.058	240;	to	91 - 73		May 10-30
	NaI	1 250 - 1 500		17	3-7		0.756	0.756		2 000	82 - 60		
OMH	PC sol 1	750 - 1 500	11 500	350	290	4.8	5.2	0.01	23;	1 000	94 - 74	Different amounts of inactive carrier	June 7-10
	" 2		10 500	230	200				14;	1 500	93 - 74		
PTB	PC	750 - 880	7 000	170	160	2.6	0.69	0.004	59;	10 000	93 - 65	Absorption Discrimination "	June 4-20
	PPC1	760 - 900	6 600	103	76	0.57	0.33	0.002	320;	2 400	90 - 70		"
	PPC2	1 250 - 1 600	7 000	650	550	0.6	1.8	0.006	780;	1 600	88 - 76		May 28-June 5
SCK	PC	745 - 930	8 300	295	251	2.17	2.56	0.027	54;	5 000	94 - 82	Addition of gold-coated VYNS foils and/or carrier	Apr. 26-June 18
									17;	5 000	90 - 81		"
	NaI	(70 - 160)	-	4 050	-	-	25.4	-	6;	5 000	92 - 80	-	May 4-11
									100;	2 000	-	-	April 21-24
UVVVR	PC	715	5 840	486	387	1.20	6.78	0.45	56;	400	91 - 63	Addition of wetting agent	May 26

Table 10 - Extrapolation data and final results

Laboratory	Method used	Order of fitted polynomial	Number of degrees of freedom	Reduced $\chi^2$	$\gamma$ efficiency of $\beta$ -ray detector		Final result (kBq g <sup>-1</sup> ) on ref. date (1982-05-01)		
					$\epsilon_{\beta\gamma}$ (%)	how determined	Activity concentration	Combined uncertainty (kBq g <sup>-1</sup> )	(%)
AAEC	4 $\pi$ $\beta$ (PC)- $\gamma$	1	6	1.0	0.1	Estimated from [31] and [32]	608.8	2.6	0.43
		1	6	1.1					
		1	6	1.1					
AECL	"	1	43 to 86	1.3 to 2.0	0.1 $\pm$ 0.03	Estimated from [33]	605.4	2.4	0.40
BARC	"	2	42	50	0.05	From slope/intercept for diff. nuclides	605.4	5.9	0.97
		2	43	87					
BCMN	"	1	7 to 9 per source	40 to 200	0.11 $\pm$ 0.03	According to [31]	604.5	1.8	0.30
BIPM	" select. sampl. coinc.	2	48	18	0.3 $\pm$ 0.1	From results with <sup>54</sup> Mn	606.7	2.7	0.44
		2	48	76					
EIL	4 $\pi$ $\beta$ (PC)- $\gamma$	2	31	0.7	0.25	According to [34]	607.0	2.2	0.36
			25	1.7					
IER	"	1	9	2.1	0.10 $\pm$ 0.05	Estimated from [32]	595.1	2.8	0.47
		1	9	109					
IFIN	"	2	46	5.0	0.54 $\pm$ 0.01	<sup>137</sup> Cs between absorbers	602.1	3.5	0.59
IMRI	"	1	20	3.3	0.11	According to [35]	604.3	1.3	0.22
NAC	4 $\pi$ $\beta$ (LS)- $\gamma$	2	12	0.9	8.0	Simulation	600.5	5.2	0.86
NBS	4 $\pi$ $\beta$ (PPC)- $\gamma$	1	4 to 14	0.02	0.54 $\pm$ 0.02	Measured for <sup>7</sup> Be, <sup>51</sup> Cr, <sup>54</sup> Mn	605.9	1.9	0.31

Table 10 (cont'd)

Laboratory	Method used	Order of fitted polynomial	Number of degrees of freedom	Reduced $\chi^2$	$\gamma$ efficiency of $\beta$ -ray detector		Final result (kBq g <sup>-1</sup> ) on ref. date (1982-05-01)		
					$\epsilon_{\beta\gamma}$ (%)	how determined	Activity concentration	Combined uncertainty (kBq g <sup>-1</sup> )	(%)
NIM	4 $\pi\beta$ (PC)- $\gamma$ 1	1	8	0.61	0.29 $\pm$ 0.05	Foil absorption	597.4	1.1	0.19
	4 $\pi\beta$ (PC)- $\gamma$ 2	1	8	0.22	0.4	Absorber method	599.2	1.6	0.27
	4 $\pi$ (LS)- $\beta$	-	-	-	5.6 $\pm$ 0.1	From <sup>60</sup> Co value	593.1	1.2	0.20
NPL	4 $\pi\beta$ (PC)- $\gamma$	2	32	34	0.32	According to [31]	601.5	11.4	1.9
		2	26	14					
	4 $\pi\beta$ (LS)- $\gamma$	2	32	1.1	7.0 $\pm$ 0.5	Interpolated	598.8	4.5	0.75
		2	32	1.3		<sup>60</sup> Co, <sup>95</sup> Nb, <sup>139</sup> Ce, <sup>123</sup> I			
		2	32	0.9					
NRC	4 $\pi\beta$ (PPC)- $\gamma$ AC	1	13 per source	0.5 - 1.5	0.32 $\pm$ 0.03	Calcul. and measured according to [32] and [36]	609.3	1.0	0.16
	4 $\pi\beta$ (PPC)-2 $\gamma$ AC (Two-parameter)	1	12	0.83					
OMH	4 $\pi\beta$ (PC)- $\gamma$	1	22	0.83	0.10 $\pm$ 0.05	Estimation	606.4	1.0	0.16
		1	13	0.37					
PDS	Ge(Li) $\gamma$	-	-	-	-	-	616.6	5.9	0.96
PTB	4 $\pi\beta$ (PC)- $\gamma$	4	56	4.3	0.11 $\pm$ 0.02	Interpolation	600.9	2.2	0.36
						<sup>7</sup> Be, <sup>54</sup> Mn, ...			
	4 $\pi\beta$ (PPC)- $\gamma$ 1	4	300	0.9 to 2.6	0.25 $\pm$ 0.06	"	605.8	2.2	0.36
	4 $\pi\beta$ (PPC)- $\gamma$ 2	4	780	4.6	"	"	605.6	2.0	0.31
SCK	4 $\pi\beta$ (PC)- $\gamma$	1	52	0.44	0.25 $\pm$ 0.05	From eff. functions for <sup>54</sup> Mn, <sup>60</sup> Co	604.8	1.2	0.20
		1	15	3.7					
		1	4	13.8					
	4 $\pi$ (NaI) $\gamma$	-	-	-	-	-	604.6	2.7	0.45
UVVVR	4 $\pi\beta$ (PC)- $\gamma$	1	54	-	0.320 $\pm$ 0.013	Interpolation	607.5	1.6	0.27
						<sup>7</sup> Be, <sup>54</sup> Mn, <sup>85</sup> Sr			

Table 11 - Uncertainty components of the final result (in %)

Component due to	AAEC	AECL	BARC	BCMNI	BIPM	ETL	IER	IFIN	IMRI	NAC	NBS	NIM coinc. LS	NPL PC LS	NRC AC	OMH	PTB PC PPC 1 PPC 2	SCK coinc. NaI	UVVVR
counting statistics	0.1	0.045	0.85	0.2	0.257	0.17	0.15	0.04	0.11	0.088	0.12	0.12 0.05	0.047 0.15	0.035	0.020	0.1 " "	0.096 0.087	0.049
weighing	0.1	0.011	0.13	0.2	0.033	0.08	0.03	0.05	0.03	0.033	0.2	0.11	0.03 0.03	0.05	0.005	0.005 " "	0.020	0.054
dead times	0.2	0.003	0.25	<0.05	0.030	0.03	0.005	0.21	<0.001	0.085	0.02	0.002 0.002 4	0.04 0.09	N.A.	0.005	0.04 " 0.01	0.008 0.05	<0.001
resolving time	0.05	<0.001	0.30	<0.05	0.030	0.04	0.009	0.01	<0.001	0.005	-	0.05 0.002	0.03 0.01	N.A.	0.010	0.01 " 0.005	<0.000 2 -	0.007
delay mismatch	0.03	0.002	-	0.06	0.020	0.1	-	-	<0.001	-	-	0.05 0.024	0.03 0.002	N.A.	0.05 " 0.1	0.006 -	0.021	
pile-up	-	0.04	-	-	0.020	-	-	-	-	-	-	-	- 0.10	0.001	-	0.01 " -	0.01 -	-
fitting procedure	0.15	0.37	0.15	0.13	0.340	0.13	0.42	0.20	0.08	0.820	0.12	0.15 0.12	1.9 0.5	0.096	0.050	0.3 " "	0.097 0.037	0.163
background	0.2	<0.012	0.10	<0.05	0.005	0.08	0.05	0.04	-	0.020	0.02	0.05 0.05	0.03 0.02	0.001	0.005	0.01 " 0.02	0.012 0.003	0.029
timing	0.01	<0.01	0.001 3	-	<0.001	-	0.002	0.01	0.003	-	0.004	0.002 -	0.003 0.003	0.01	0.005	0.003 " "	0.000 3 -	<0.001

Table 11 (cont'd)

Component due to	AAEC	AECL	BARC	BCMNI	BIPM	ETL	IER	IFIN	IMRI	NAC	NBS	NIM coinc. LS	NPL PC LS	NRC	OMH	PTB PC PPC 1 PPC 2	SCK coinc. NaI	UVVVR	
$\alpha_t$ and branching	0.05	< 0.057	0.01	< 0.05	0.049	0.07	0.07	0.075	0.05	0.071	0.072	0.04 0.045	0.066 0.066	0.044	0.055	0.06	0.044	0.170	
$^{137}\text{Ba}^m$ photons in $\beta$ -ray detector	0.08	< 0.024	0.005	< 0.05	0.085	0.05		0.01	0.007	0.18	0.02	0.02	0.02 0.04	0.02 0.5	0.025	0.040	0.03 0.07	0.035	0.010
" " " $\gamma$ -ray detector	0.1	< 0.072	0.006	< 0.05	-	0.05		0.02	0.10	-	0.025	0.05	0.05 -	0.01 0.02	-	0.005	0.05	0.09	0.014
half lives	0.015	< 0.001	0.008	< 0.05	0.005	0.01	0.004	0.002	0.004	-	0.001	0.01 0.000 12	0.001 0.001	0.002	0.005	0.001	0.001 0.000 5	0.002	
tracer activity	0.18	< 0.094	0.10	< 0.05	0.094	0.19	0.094	0.15	0.09	0.094	0.06 -0.13	0.1 0.09	0.07 0.07	0.099	0.100	0.1	0.10	0.088	
impurities	0.01	< 0.005	0.004	< 0.05	0.010	0.01	-	0.01	-	0.004	0.001	- 0.006	0.005 0.005	-	0.005	0.005	0.016 0.07	0.020	
adsorption	0.03	< 0.001	-	< 0.05	-	0.05	0.03	0.06	-	-	0.02	-	0.015 0.015	0.013	0.025	0.02	< 0.01 0.01	0.005	
other effects	0.05	-	-	-	-	0.1	-	0.30	-	-	-	-	- 0.052	-	-	0.05 " "	- 0.43	0.057	
Combined uncertainty	0.43	0.40	0.97	0.30	0.44	0.36	0.47	0.47	0.22	0.86	0.31	0.27 0.20	1.9 0.75	0.16	0.14	0.36 0.36 0.31	0.20 0.45	0.27	

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