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ABSTRACT

As agent for the Commonwealth Scientific and Industrial Research Organisation, the Australian Atomic Energy Commission is responsible for the maintenance of the Australian standard of activity. The standard comprises activity measurement procedures involving the operation of $4\pi\beta$-γ coincidence counting equipment.

The coincidence method requires the application of correction factors which depend on detection efficiency, such as arise for complex decay schemes and internal conversion. These corrections approach unity as the detection efficiency in the β-channel approaches 100 per cent. By performing activity determinations for a range of β detection efficiencies, an 'efficiency extrapolation' analysis can be applied which eliminates the need to determine the absolute detection efficiency for each channel.

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AAEC; AUSTRALIA; BARIUM 133; BETA PARTICLES; CALIBRATION STANDARDS; COINCIDENCE CIRCUITS; COINCIDENCE METHODS; DEAD TIME; EFFICIENCY; EXTRAPOLATION; GAMMA RADIATION; RADIATION DETECTION; RADIOACTIVITY; SENSITIVITY; STATISTICS

EDITORIAL NOTE

The Australian Nuclear Science and Technology Organisation replaced the Australian Atomic Energy Commission on 27 April 1987. Reports issued after April 1987 have the prefix ANSTO with no change of the symbol (E, M, S or C) or numbering sequence. Several reports issued in April 1987 have retained the AAEC prefix, their publication having been delayed by unavoidable production problems.

CONTENTS

1. INTRODUCTION

In the field of radiation physics, the term 'activity' refers to the rate of disintegration of nuclei in radioisotopes. Defined as the number of disintegrations per second, the unit of activity is the becquerel (Bq). The quantity determined by measurement is the 'radioactivity concentration', which may be expressed as either kBq g^{-1}, or Bq mg^{-1}. The maintenance of standards refers in this instance to the establishment of a measurement procedure. There is no single set of apparatus which can determine activity for all radioactive species. Rather, methods are developed which depend on the decay processes of the selected isotope. As there are about one thousand known radionuclides, the radiation standards group must restrict its attention to those energetic β- and γ-emitting radionuclides which have simple decay schemes and are of interest to research workers.

2. THE COUNTING SYSTEM

This report describes the use of an efficiency extrapolation method for the absolute determination of radioactivity, an analysis carried out by the Australian Atomic Energy Commission* at the Lucas Heights Research Laboratories (LHRL) as part of its program for the maintenance of an Australian standard of activity.

The apparatus consists of a sodium iodide crystal and 4π gas flow proportional counter, with associated electronics. The sodium iodide detector has for gamma radiation a detection efficiency of approximately six per cent, determined effectively by the counting geometry. By contrast, the proportional counter has a near-zero detection efficiency for gamma rays, and about eighty per cent for beta particles. This latter figure is heavily dependent on the source material and the source mounting. Hence the beta detection efficiency may be readily varied by using thicker VYNS source mounting films, or by placing films over the source, thereby increasing self-absorption. Pulses arising in these detectors as a result of nuclear decay are recorded as counts. In addition to the beta and gamma counts, the number of 'coincidences' is also recorded. The coincidence count represents the number of times beta and gamma pulses occur simultaneously in the detectors, and are therefore most likely to have arisen from the same nuclear disintegration.

Analysis of the counts permits determination of absolute activity. However, deviations from ideal behaviour in the counting system lead to the need for correction factors, most of which are intrinsically difficult to determine with good precision. The concept of 'efficiency extrapolation' is discussed as a method for avoiding the absolute determination of these correction factors, thereby increasing the precision of absolute activity measurement.

With the observations appearing as counts, their analysis requires the use of statistical techniques based on the Poisson probability density function. The statistical analysis is given in **section 5.3.** Ultimately, the objective of the experiment is to determine the radioactivity concentration with as high a precision as possible. This report provides a method of analysis for placing accuracy statements on the results.

3. COINCIDENCE COUNTING PRINCIPLES

The methodology of coincidence counting has been given by Campion [1959]. For a given source, count rates are determined for the beta and gamma emissions. In addition, the occurrence of coincidences between the beta and gamma pulses is recorded by a separate electronic mixer. With gamma emission following promptly after beta decay, these coincident counts are considered to arise from disintegration of the same atom.

With N_o the activity of the source, N_β the observed beta count rate, N_γ the observed gamma count rate and N_c the count rate for coincidences we may write

$$N_\beta = \varepsilon_\beta N_o \quad , \qquad (1)$$

$$N_\gamma = \varepsilon_\gamma N_o \quad \text{, and} \qquad (2)$$

$$N_c = \varepsilon_\beta \varepsilon_\gamma N_o \quad . \qquad (3)$$

Here, ε_β is the efficiency for detection of beta particles in the 4π proportional counter, and ε_γ is the efficiency for detection of gamma rays in the sodium iodide crystal.

Solving **equations 1, 2** and **3** yields

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* Now known as the Australian Nuclear Science and Technology Organisation (ANSTO)

$$N_o = \frac{N_\beta N_\gamma}{N_c} , \tag{4}$$

$$\varepsilon_\beta = \frac{N_c}{N_\gamma} , \text{ and} \tag{5}$$

$$\varepsilon_\gamma = \frac{N_c}{N_\beta} . \tag{6}$$

Equation 4 yields the absolute activity which, in principle, is independent of detection efficiency for either beta or gamma channels. However, this independence is not achieved in practice, as the determination of activity is influenced by other effects, such as deadtime in the counter and the occurrence of 'accidental' coincidences. The correction factors for these effects are mentioned in **section 4**.

It should be noted that N_o, ε_β and ε_γ are correlated, *i.e.* a variation in one gives rise to variation in the other two. Analysis of these types of data requires a knowledge of the covariance matrix. For Poisson variates, the covariance matrix is readily derived. Further, N_c depends on N_β and N_γ, with the interesting result that [NCRP 1985]

$$N_c = \text{cov}\,(N_\beta, N_\gamma) \tag{7}$$

Equation 7 is the basis for an alternative method to coincidence counting known as correlation counting.

4. CORRECTION FACTORS FOR COINCIDENCE COUNTING

For low count rates and isotopes having energetic decay products, the errors in the expression for activity N_o in **equation 4** are quite low. Generally, the errors are of two kinds, those dependent on detection efficiency and those which are not. The latter group includes errors due to deadtime and accidental coincidences:

- Dead time losses occur when a second disintegration takes place while the counter is still paralysed from the first event.
- Accidental coincidences arise when beta and gamma counts from separate nuclei occur during the coincidence resolving time.

At low count rates, satisfactory correction factors for deadtime and accidentals have been derived by Bryant [1963]:

$$N_{oc} = \frac{N_\beta N_\gamma}{N_c} \, \frac{N_c}{N_c - 2T_R N_\beta N_\gamma} \left\{ 1 + \frac{2N_cT - 2T_R\,(N_\beta + N_c)}{2 - N_\beta T - N_\gamma T} \right\} , \tag{8}$$

$$\varepsilon_{\beta c} = \frac{N_\beta}{N_o(1 - N_\beta T)} , \text{ and} \tag{9}$$

$$\varepsilon_{\gamma c} = \frac{N_\gamma}{N_o\,(1 - N_\gamma T)} , \tag{10}$$

where deadtime is denoted by T and resolving time by T_R. The subscript c in N_{oc}, $\varepsilon_{\beta c}$ and $\varepsilon_{\gamma c}$ indicates that correction has been made.

Errors also arise which depend on detection efficiency. These include

- internal conversion, where the conversion electron is counted instead of a gamma ray;
- gamma sensitivity of the β detector, with the result that a β pulse is recorded, even though the β particle was lost by absorption in the source mounting; and
- complex decay schemes.

The correction for a complex decay scheme, say ^{60}Co, which produces two gamma rays of different energy, and where a and b are the branching ratios, takes the form

$$\frac{N_\beta N_\gamma}{N_c} = N_o \left\{ 1 - \frac{ab(\varepsilon_{\beta a} - \varepsilon_{\beta b})\,(\varepsilon_{\gamma a} - \varepsilon_{\gamma b})}{a\,\varepsilon_{\beta a}\,\varepsilon_{\gamma a} + b\varepsilon_{\beta b}\,\varepsilon_{\gamma b}} \right\} \tag{11}$$

Inspection of **equation 11** reveals that the correction is unity for 100 per cent beta detection efficiency. Consequently, an extrapolation to 100 per cent beta efficiency for N_o determined at a range of efficiencies should yield N_o free of such correction factors. This is the basis of the efficiency extrapolation method described in **section 5**.

5. EFFICIENCY EXTRAPOLATION

5.1 The Technique of Efficiency Extrapolation

It was shown in **section 4** that correction factors dependent on detection efficiency approach unity as ε_β approaches 100 per cent. In this technique, measurements of radioactivity concentration are performed for a range of sources having different beta detection efficiencies.

A plot of radioactivity concentration against $f(\varepsilon_\beta)$, both corrected for deadtime and accidentals, is prepared, where

$$f(\varepsilon_\beta) = \frac{1 - \varepsilon_\beta}{\varepsilon_\beta} \quad . \tag{12}$$

A line of best fit then intercepts the radioactivity concentration axis when ε_β is unity. At that point, correction terms dependent on efficiency are also unity. Thus the radioactivity concentration is determined free of systematic effects arising from dependence on detection efficiency. A diagram illustrating the results of this extrapolation technique for ^{133}Ba is presented in **appendix B**.

5.2 Limits of Validity

The detection efficiency may be varied in a number of ways, most commonly

- by varying the self-absorption of beta particles within the source; or
- by varying the discrimination level in the beta channel pulse height analyser.

An analysis of these methods was given by Smith and Stuart [1975]. The second method is not used at LHRL, as it gives rise to non-linearities in the efficiency curve which require complex analysis.

It remains to be shown that the self-absorption method indeed yields a straight line fit to the efficiency curve. Smith and Stuart [1975] showed this to be the case when

- the output pulse amplitude from the beta detector is not proportional to the beta particle energy;
- all particles entering the detector are counted; and
- the variation in beta detection efficiency is due to absorption at or near the source.

The third condition is met immediately at LHRL, as it is the standard method of source preparation. To satisfy the first two conditions, consider the mode of operation for the beta channel electronics.

The proportional counter produces a pulse height spectrum determined by the continuous energy range of the beta particles. The pre-amplifier is specially designed to have low noise characteristics. Consequently, by increasing the gas gain of the proportional counter, it is possible to pull all (or very nearly all) of the beta pulses above the basic amplifier noise. Hence, all the electrons entering the gas volume are passed on as pulses to the main amplifier.

The main amplifier is designed to have a very rapid recovery from overloading. Thus, pulses are clipped and slightly wider in the overloaded condition. By increasing the gain of the main amplifier so that all pulses from the pre-amplifier are clipped, the output pulse height becomes independent of beta particle energy. Consequently, both the first and last conditions are met, and the straight line fit is a valid physical model.

A secondary requirement is correction for deadtime and accidental coincidence. In preparing sources with differing self-absorption characteristics, it is not practical to produce them with identical count rates. In practice, the count rates from source to source range over a factor of 1.5. Accordingly, the corrections for deadtime and accidentals also vary. For count rates below six thousand per second, the limitations of Bryant's [1963] equation may be ignored. Above this, a more advanced theory is required, such as is given by Wyllie [1987] and Cox and Isham [1977].

5.3 Statistical Analysis

For this experiment, a range of sources is counted, each source having different self-absorption properties for the beta particles. In principle, a single count for each source is all that is required for the line-fitting procedure. The accuracy is then determined by the total number of counts recorded per source.

However, there is an advantage to be gained by counting each source over an ensemble of shorter times. As, strictly speaking, they are replicates, a chi-squared (χ^2) test may be performed on each member of the ensemble. This will reveal if the counting equipment is performing incorrectly, which may be the case, for example, if the counting gas has not adequately flushed the proportional counter or the sodium iodide system is not correctly set on its plateau. For the purpose of this χ^2 test, the count time within each ensemble should be the same.

Then the reduced χ^2, χ^2/υ where υ is the number of observation n less one, is given by

$$\frac{\chi^2_{\upsilon}}{\upsilon} = \frac{1}{n-1} \sum_i \frac{(Ci - \bar{C})^2}{\bar{C}} \tag{13}$$

where, C_i are the counts, uncorrected for background, from either beta, gamma or coincidence channels, and $\bar{C}$ is the mean count. Given a satisfactory χ^2 test for each source, the variables for the extrapolation line may be derived, complete with their covariances. The line of best fit will then be regressed on each observation, and not on the means for each source. Adopting the notations C_β, C_γ and C_c for counts over time t for beta, gamma and coincidence channels, from **equation 4**

$$N_o = \frac{C_\beta C_\gamma}{C_c t} , \tag{14}$$

$$\varepsilon_\beta = \frac{C_c}{C_\gamma} , \text{ and} \tag{15}$$

$$\varepsilon_\gamma = \frac{C_c}{C_\beta} . \tag{16}$$

The information matrix for each observation is the symmetric matrix B with (**appendix A**).

$$b_{11} = N_o \left[\frac{1}{\varepsilon_\beta} + \frac{\varepsilon_\gamma}{1-\varepsilon_\beta} \right] , \tag{17}$$

$$b_{12} = -N_o , \tag{18}$$

$$b_{13} = 1 - \varepsilon_\gamma , \tag{19}$$

$$b_{22} = N_o \left[\frac{1}{\varepsilon_\gamma} + \frac{\varepsilon_\beta}{1-\varepsilon_\gamma} \right] , \tag{20}$$

$$b_{23} = 1 - \varepsilon_\beta , \text{ and} \tag{21}$$

$$b_{33} = \frac{\varepsilon_\beta + \varepsilon_\gamma (1-\varepsilon_\beta)}{N_o} \tag{22}$$

The covariance matrix A for each observation is

$$A = TB^{-1} , \tag{23}$$

where

$$T = \text{diag} \left\{ \frac{1}{t}, \frac{1}{t}, \frac{1}{t} \right\} \tag{24}$$

Then a_{11}, a_{12} and a_{13} are the variances of ε_β, ε_γ and N_o, and a_{12}, a_{13} and a_{23} are the covariances cov (ε_β, ε_γ), cov (ε_β, N_o) and cov (ε_γ, N_o).

Denoting the radioactivity concentration by Y and the efficiency function $f(\varepsilon_\beta)$ by X, then for each observation

$$Y = N_0 \frac{KR}{m} \quad , \tag{25}$$

$$X = L\left[\frac{1-\varepsilon_\beta}{\varepsilon_\beta}\right] \quad , \tag{26}$$

$$\text{var}(Y) = a_{33} \frac{K^2R^2}{m} \quad , \tag{27}$$

$$\text{var}(X) = a_{11} \frac{L^2}{\varepsilon_\beta^4} \quad \text{, and} \tag{28}$$

$$\text{cov}(X,Y) = -a_{13} \frac{LKR}{\varepsilon_\beta^2 m} \quad , \tag{29}$$

where, K is the correction for dead time and accidental coincidences from **equation 8**, m the source mass in milligrams, R the decay correction taken to the specified reference date, var ( ) denotes variance and cov ( ) covariance.

The correction factor L takes into account the effects of deadtime and accidental coincidences on the estimation of ε_β, as given in **equation 9**, where

$$L = \frac{(1-\varepsilon_{\beta c}) / \varepsilon_{\beta c}}{(1-\varepsilon_\beta) / \varepsilon_\beta} \quad . \tag{30}$$

In **equations 27**, **28** and **29**, both K and L have been treated as constants. This is not strictly the case, as both are functions of the Poisson distributed observations. However, provided that the correction factors are small ($N_\beta < 6$ kBq), the propagation of errors due to these effects may be ignored.

With each point on the efficiency extrapolation curve specified, it remains to perform the curve fit. The efficiency extrapolation model for the counting method in use at LHRL is given by

$$Y = a + bx \tag{31}$$

If only Y were subject to error, the weighted least squares regression estimates for a and b would be

$$\hat{a} = \frac{1}{\Delta} (\Sigma w_i X_i^2 \Sigma w_i Y_i - \Sigma w_i X_i \Sigma w_i X_i Y_i) \tag{32}$$

$$\hat{b} = \frac{1}{\Delta} (\Sigma w_i \Sigma w_i X_i Y_i - \Sigma w_i X_i \Sigma w_i Y_i) \tag{33}$$

$$\Delta = \Sigma w_i \Sigma w_i X_i^2 - (\Sigma w_i X_i)^2 \tag{34}$$

Before considering the weighting factors w_i, recall that X is also subject to variation and from **equation 29**:

$$\text{cov}(X,Y) \neq 0 \; ; \tag{35}$$

hence the regression **equations 32** and **33** are not directly applicable. The requirement to obtain a 'best fit' is achieved by minimising the function

$$\frac{\Sigma (Y_i - a - bX_i)^2}{\text{var}(Y_i - a - bX_i)} \tag{36}$$

This suggests that an iterative procedure for adjusting $\hat{a}$ and $\hat{b}$ to minimise **equation 36** would allow for the correlation between X and Y. The steps in the iteration process are as follows

- Choose an initial value for $\tilde{b}$, say $\tilde{b} = 0$.
- Calculate the weights w_i from the expression

$$w_i(\tilde{b}) = 1 / (\text{var}(Y_i) - 2\tilde{b}\,\text{cov}(X_i, Y_i) + \tilde{b}^2\,\text{var}(X_i)) \quad . \tag{37}$$

- Regress Y_i on X_i with weights $w_i(\tilde{b})$ to get $\hat{a}$ and $\hat{b}$ from **equations 32** and **33**
- If $\hat{b} = \tilde{b}$, then **equation 36** is minimised. Otherwise set $\tilde{b} = \hat{b}$ and return to **weighting expression 37** for another pass.

The value of â is then the best estimate of N_o/m, *i.e.* the radioactivity concentration. The variance of N_o/m is given by

$$\text{var}(\hat{a}) = \frac{\Sigma w_i X_i^2}{\Sigma w_i \Sigma w_i (X_i - \bar{X})^2} \qquad (38)$$

Finally, the reduced χ^2 for goodness of fit test is given by

$$\frac{\chi_\upsilon^2}{\upsilon} = \frac{\Sigma (Y_i - \hat{a} - \hat{b} X_i)^2}{\Sigma [\text{var}(Y_i) - 2\hat{b}\,\text{cov}(X_i, Y_i) + \hat{b}^2 \text{var}(X_i)]} \qquad (39)$$

and υ is the number of observations – 2.

Note that $\text{var}(\chi^2) = 2\upsilon$ (40)

so that the standard deviation (st) of $\frac{\chi_\upsilon^2}{\upsilon}$ is given by

$$\text{st}\left\{\frac{\chi_\upsilon^2}{\upsilon}\right\} = \frac{2}{\upsilon} \qquad (41)$$

6. CONCLUSIONS

When estimating the radioactivity concentration by the coincidence counting method, certain correction factors depend on the efficiency for detection of β particles. The values of these correction factors are difficult to ascertain by direct measurement.

A technique called the efficiency extrapolation method, which eliminates the need to determine these factors, has been discussed. However, the highly correlated nature of the observations from coincidence counting places constraints on the method of analysis and estimation of uncertainty for the radioactivity concentration. A rigorous procedure was given for the statistical analysis of these data, taking account of the Poisson nature of events and the correlations introduced by their manipulation. The value of radioactivity concentration, its variance, and a χ^2 goodness-of-fit parameter were also derived. A computer program, EFFEXTRAP, has been prepared to analyse this form of data.

7. ACKNOWLEDGEMENTS

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APPENDIX A

DERIVATION OF BEST ESTIMATORS FOR N_o, ε_β AND ε_γ, AND THEIR VARIANCE CO-VARIANCE MATRIX

A1. DISCUSSION

A radioactive source has disintegrations which follow a Poisson process in time with constant rate N_o. A disintegration leads to the emission of β- and γ-rays. The β particle is observed with probability ε_β, and the gamma photon with probability ε_γ. Counting the emissions for a time t gives the observed number of β particles as N_β, the γ-ray as N_γ and the number of coincident events as N_c. By a theorem of statistics, if each point in a Poisson process is independently classified into one of k types, then the k point processes of particle types are independent Poisson processes.

It follows that $N_\beta - N_c$ is Poisson distributed with mean $\varepsilon_\beta (1-\varepsilon_\gamma) N_o t$, $N_\gamma - N_c$ with mean $\varepsilon_\gamma(1-\varepsilon_\beta) N_o t$, and the coincidences with mean $\varepsilon_\beta \varepsilon_\gamma N_o t$. The number of unobserved disintegrations has a Poisson distribution with mean $(1-\varepsilon_\beta)(1-\varepsilon_\gamma)N_o t$. All four random variables are independent. In this discussion, the assumption is made that dead time effects in the counting apparatus do not significantly alter these statistics.

With the probability density functions known, estimators $\hat{N}_o$, $\hat{\varepsilon}_\beta$ and $\hat{\varepsilon}_\gamma$ of N_o, ε_β and ε_γ may be found by the method of maximum likelihood.

A2. MAXIMUM LIKELIHOOD ESTIMATION OF N_o, ε_β AND ε_γ

The likelihood of a particular data set (N_c, $N_\beta - N_c$, $N_\gamma - N_c$) is given by

$$\begin{aligned} L = {} & e^{-\varepsilon_\beta\varepsilon_\gamma N_o t} (\varepsilon_\beta\varepsilon_\gamma N_o t)^{N_c} e^{-\varepsilon_\beta(1-\varepsilon_\gamma)N_o t} \\ & \cdot \left[\varepsilon_\beta(1-\varepsilon_\gamma)N_o t \right]^{N_\beta - N_c} e^{-\varepsilon_\gamma(1-\varepsilon_\beta)N_o t} \\ & \cdot \left[\varepsilon_\gamma(1-\varepsilon_\beta)N_o t \right]^{N_\gamma - N_c} / \left[N_c!\,(N_\beta - N_c)!\,(N_\gamma - N_c)! \right] \end{aligned} \qquad (A1)$$

and

$$\begin{aligned} \ln L = {} & -\left[\varepsilon_\beta + \varepsilon_\gamma(1-\varepsilon_\beta) \right] N_o t + N_c \ln(\varepsilon_\beta \varepsilon_\gamma N_o t) \\ & + (N_\beta - N_c) \ln\left[\varepsilon_\beta(1-\varepsilon_\gamma) N_o t \right] \\ & + (N_\gamma - N_c) \ln\left[\varepsilon_\gamma(1-\varepsilon_\beta) N_o t \right] \\ & - \ln N_c! - \ln(N_\beta - N_c)! - \ln(N_\gamma - N_c)! \quad , \end{aligned} \qquad (A2)$$

where N_o, ε_β and ε_γ are to be estimated.

A given outcome is most probable when the likelihood function is at a maximum, *e.g.*

$$\frac{\partial \ln L}{\partial N_o} = 0$$

Now

$$\frac{\partial \ln L}{\partial N_o} = \frac{N_\beta + N_\gamma - N_c}{N_o} - \left[\varepsilon_\beta + \varepsilon_\gamma(1-\varepsilon_\beta) \right] t \quad , \qquad (A3)$$

$$\frac{\partial \ln L}{\partial \varepsilon_\beta} = \frac{N_\beta}{\varepsilon_\beta} - \frac{(N_\gamma - N_c)}{1 - \varepsilon_\beta} - (1 - \varepsilon_\gamma) N_o t \quad \text{, and} \tag{A4}$$

$$\frac{\partial \ln L}{\partial \varepsilon_\gamma} = \frac{N_\gamma}{\varepsilon_\gamma} - \frac{(N_\beta - N_c)}{(1 - \varepsilon_\gamma)} - (1-\varepsilon_\beta) N_o t \ . \tag{A5}$$

The best estimators in the maximum likelihood sense are $\hat{N}_o$, $\hat{\varepsilon}_\beta$ and $\hat{\varepsilon}_\gamma$, where

$$\hat{N}_o = \frac{N_\beta + N_\gamma - N_c}{[\, \hat{\varepsilon}_\beta + \hat{\varepsilon}_\gamma (1-\hat{\varepsilon}_\beta) \,] \, t} \tag{A6}$$

and $\hat{\varepsilon}_\gamma$, $\hat{\varepsilon}_\beta$ are the roots of the joint **equations A4** and **A5** after substituting **A6**, whence

$$\hat{\varepsilon}_\beta = \frac{N_c}{N_\gamma} \tag{A7}$$

$$\hat{\varepsilon}_\gamma = \frac{N_c}{N_\beta} \tag{A8}$$

$$\hat{N}_o = \frac{N_\beta N_\gamma}{N_c t} \tag{A9}$$

A3. VARIANCE - COVARIANCE MATRIX OF $\hat{N}_o$, $\hat{\varepsilon}_\beta$ AND $\hat{\varepsilon}_\gamma$

In all practical senses, the estimators $\hat{\varepsilon}_\beta$, $\hat{\varepsilon}_\gamma$ and $\hat{N}_o$ in **equations A7, A8** and **A9** are adequate. However, because their denominators may be zero with non-zero probability, they may be applied only if $N_c \neq 0$.

It is then desirable to maximise the conditional likelihood defined by:

$$L_c = \frac{L}{(1 - e^{-\varepsilon_\beta \varepsilon_\gamma N_o t})} \tag{A10}$$

Notation: The terms $1 - e^{-\varepsilon_\beta \varepsilon_\gamma N_o t}$ and $e^{\varepsilon_\beta \varepsilon_\gamma N_o t} - 1$ occur repeatedly in the following text. They are replaced here by E and F, respectively.

Then

$$\ln L_c = \ln L - \ln E$$

$$\frac{\partial \ln L_c}{\partial \varepsilon_\beta} = \frac{\ln L}{\partial \varepsilon_\beta} - \frac{\varepsilon_\gamma N_o t \, e^{-\varepsilon_\beta \varepsilon_\gamma N_o t}}{E} \ , \tag{A11}$$

$$\frac{\partial \ln L_c}{\partial \varepsilon_\gamma} = \frac{\partial \ln L}{\partial \varepsilon_\gamma} - \frac{\varepsilon_\beta N_o t \, e^{-\varepsilon_\beta \varepsilon_\gamma N_o t}}{E} \ \text{, and} \tag{A12}$$

$$\frac{\partial \ln L_c}{\partial N_o} = \frac{\partial \ln L}{\partial N_o} - \frac{\varepsilon_\beta \varepsilon_\gamma t \, e^{-\varepsilon_\beta \varepsilon_\gamma N_o t}}{E} \tag{A13}$$

Maximisation of $\ln L_c$ then leads to estimators $\tilde{\varepsilon}_\beta$, $\tilde{\varepsilon}_\gamma$, $\tilde{N}_o$ which are very close to $\hat{\varepsilon}_\beta$, $\hat{\varepsilon}_\gamma$ and $\hat{N}_o$, but involve numerical methods to solve **equations A11, A12** and **A13**.

The advantage of casting the theory in terms of L_c is to apply standard formulae for asymptotic variances free from the analytic problem concerning $\hat{\varepsilon}_\beta$, $\hat{\varepsilon}_\gamma$ and $\hat{N}_o$, namely, that they have infinite variance for all t.

Variances of the estimators $\tilde{\varepsilon}_\beta$, $\tilde{\varepsilon}_\gamma$ and $\tilde{N}_o$ will decrease to zero as t approaches infinity. The rate of convergence to zero is indicated by the formula

$$\text{var}\,(\tilde{\varepsilon}_\gamma) = \frac{a}{t} + \frac{b}{t^2} + \frac{c}{t^3} + \cdots \tag{A14}$$

where var indicates variance. Similar expressions apply for $\tilde{\varepsilon}_\beta$ and $\tilde{N}_o$.

We will find 'a' to enable the asymptotic formula to be applied, for values of t where t^{-2} is negligible but t^{-1} is not. To find the expectations of the estimators, we first form the following derivatives:

$$\frac{\partial^2 \ln L}{\partial \varepsilon_\beta^2} = -\frac{N_\beta}{\varepsilon_\beta^2} - \frac{N_\gamma - N_c}{(1-\varepsilon_\beta)^2} , \quad (A15)$$

$$\frac{\partial^2 \ln L}{\partial \varepsilon_\gamma^2} = -\frac{N_\gamma}{\varepsilon_\gamma^2} - \frac{N_\beta - N_c}{(1-\varepsilon_\gamma)^2} , \quad (A16)$$

$$\frac{\partial^2 \ln L}{\partial N_o^2} = -\frac{N_\beta + N_\gamma - N_c}{N_o^2} , \quad (A17)$$

$$\frac{\partial^2 \ln L}{\partial \varepsilon_\gamma \partial \varepsilon_\beta} = N_o t , \quad (A18)$$

$$\frac{\partial^2 \ln L}{\partial N_o \partial \varepsilon_\beta} = -(1-\varepsilon_\gamma) t \text{ , and} \quad (A19)$$

$$\frac{\partial^2 \ln L}{\partial N_o \partial \varepsilon_\gamma} = -(1-\varepsilon_\beta) t . \quad (A20)$$

In the shorthand notation, the additional term for, say **equation A11**, is

$$\frac{\varepsilon_\gamma N_o t}{e^{\varepsilon_\beta \varepsilon_\gamma N_o t} - 1} = \frac{\varepsilon_\gamma N_o t}{F}$$

Then also

$$\frac{\partial^2 \ln L_c}{\partial \varepsilon_\beta^2} = \frac{\partial^2 \ln L}{\partial \varepsilon_\beta^2} + \frac{(\varepsilon_\gamma N_o t)^2 e^{\varepsilon_\beta \varepsilon_\gamma N_o t}}{F^2} , \quad (A21)$$

$$\frac{\partial^2 \ln L_c}{\partial \varepsilon_\gamma^2} = \frac{\partial^2 \ln L}{\partial \varepsilon_\gamma^2} + \frac{(\varepsilon_\beta N_o t)^2 e^{\varepsilon_\beta \varepsilon_\gamma N_o t}}{F^2} , \quad (A22)$$

$$\frac{\partial^2 \ln L_c}{\partial N_o^2} = \frac{\partial^2 \ln L}{\partial N_o^2} + \frac{(\varepsilon_\beta \varepsilon_\gamma t)^2 e^{\varepsilon_\beta \varepsilon_\gamma N_o t}}{F^2} , \quad (A23)$$

$$\frac{\partial^2 \ln L_c}{\partial \varepsilon_\beta \partial \varepsilon_\gamma} = N_o t - \frac{N_o t}{F} + \frac{\varepsilon_\gamma \varepsilon_\beta (N_o t)^2 e^{\varepsilon_\beta \varepsilon_\gamma N_o t}}{F^2} , \quad (A24)$$

$$\frac{\partial^2 \ln L_c}{\partial N_o \partial \varepsilon_\beta} = -(1-\varepsilon_\gamma) t - \frac{\varepsilon_\gamma t}{F} + \frac{N_o \varepsilon_\beta (\varepsilon_\gamma t)^2 e^{\varepsilon_\beta \varepsilon_\gamma N_o t}}{F^2} , \quad (A25)$$

$$\frac{\partial^2 \ln L_c}{\partial N_o \partial \varepsilon_\gamma} = -(1-\varepsilon_\beta) t - \frac{\varepsilon_\beta t}{F} + \frac{N_o \varepsilon_\gamma (\varepsilon_\beta t)^2 e^{\varepsilon_\beta \varepsilon_\gamma N_o t}}{F^2} . \quad (A26)$$

Taking negative expectations yields

$$-E\left[\frac{\partial^2 \ln L_c}{\partial \varepsilon_\beta^2}\right] = \frac{N_o t}{\varepsilon_\beta} + \frac{\varepsilon_\gamma N_o t}{1-\varepsilon_\beta} - \frac{(\varepsilon_\gamma N_o t)^2 e^{\varepsilon_\beta \varepsilon_\gamma N_o t}}{F^2} , \quad (A27)$$

$$-E\left[\frac{\partial^2 \ln L_c}{\partial \varepsilon_\gamma^2}\right] = \frac{N_o t}{\varepsilon_\gamma} + \frac{\varepsilon_\beta N_o t}{(1-\varepsilon_\gamma)} - \frac{(\varepsilon_\beta \varepsilon_\gamma t)^2 e^{\varepsilon_\beta \varepsilon_\gamma N_o t}}{F^2} , \quad (A28)$$

$$-E\left[\frac{\partial^2 \ln L_c}{\partial N_o^2}\right] = \frac{[\varepsilon_\beta + \varepsilon_\gamma(1-\varepsilon_\beta)] N_o t}{N_o^2} - \frac{(\varepsilon_\beta \varepsilon_\gamma t)^2 e^{\varepsilon_\beta \varepsilon_\gamma N_o t}}{F^2} . \quad (A29)$$

and similarly for the covariance terms.

Let V_t be the 3 × 3 matrix of these negative expectations. From standard theory, $t\,V_t^{-1}$ yields the matrix of quantities such as 'a' in **equation A14**.

$$\text{Then } \lim_{t\to\infty} \frac{V_t}{t} = \begin{bmatrix} N_o\left[\dfrac{1}{\varepsilon_\beta} + \dfrac{\varepsilon_\gamma}{1-\varepsilon_\beta}\right] & -N_o & (1-\varepsilon_\gamma) \\ -N_o & N_o\left[\dfrac{1}{\varepsilon_\gamma} + \dfrac{\varepsilon_\beta}{1-\varepsilon_\gamma}\right] & (1-\varepsilon_\beta) \\ (1-\varepsilon_\gamma) & (1-\varepsilon_\beta) & \dfrac{\varepsilon_\beta + \varepsilon_\gamma(1-\varepsilon_\beta)}{N_o} \end{bmatrix} \qquad \text{(A30)}$$

Matrix A30 is the information matrix for the data set (N_c, $N_\beta - N_c$, $N_\gamma - N_c$) and its inverse A is the variance - covariance matrix of the data set.

The elements of A, denoted by a_{ij}, may be interpreted as

$$a_{11} = \text{var}(\varepsilon_\beta) \quad ,$$

$$a_{22} = \text{var}(\varepsilon_\gamma) \quad ,$$

$$a_{13} = \text{var}(N_o) \quad ,$$

$$a_{12} = \text{cov}(\varepsilon_\beta, \varepsilon_\gamma) \quad ,$$

$$a_{13} = \text{cov}(\varepsilon_\beta, N_o) \quad , \text{ and}$$

$$a_{23} = \text{cov}(\varepsilon_\gamma, N_o) \quad .$$

This completes the derivation of the best estimators for N_o, ε_β and ε_γ and their variance-covariance matrix.

APPENDIX B

APPLICATION OF THE EFFICIENCY EXTRAPOLATION METHOD TO ^{133}Ba DATA

B1. DISCUSSION

The decay of ^{133}Ba is characterised by the production of photons below 81 and above 124 keV. The former have a good chance of being detected in the $4\pi\beta$ gas counter, and the latter may be detected in the sodium iodide crystal. With a suitable choice of counting windows, the two energy regimes mimic $4\pi\beta$-γ counting practices, and may be analysed in the same way.

The efficiency extrapolation method described in **section 5** was applied to ^{133}Ba data. In particular, a statistic for the goodness of fit, χ^2, was obtained for the extrapolation line. The derivation of this χ^2 statistic takes into account the correlation between radioactivity concentration and detection efficiency in the β channel. However, it is not rigorous in the sense that:

- K (**equations 8** and **25**) and L (**equations 26** and **30**) are assumed to be constants, even though they are computed from observations, and
- dead-time effects are assumed not to affect the Poisson statistics of the observations N_c, N_β and N_γ.

B2. ANALYSIS OF THE ^{133}Ba DATA

The experiment was conducted by counting nine distinct sources, each repetitively up to ten times. In addition, the detection efficiency was varied, by using different counting gases and source mount thicknesses. As a result, the observations comprise an ensemble of 15 data sets, each with a unique value of detection efficiency.

An immediate reaction to the analysis of data in this form is to determine averages for each ensemble, then apply the extrapolation line fit to these means. However, the value of the analysis outlined in **section 5.3** is that the line is fitted to each observation, not to the ensemble averages. Consequently, allowance may be made for the correlation within each observation (N_c, N_β, N_γ), and error bars derived for both the radioactivity concentration and the efficiency function.

Further, the assumption of independence for observations within a given ensemble may also be tested. A chi-squared test for repeated counts on a given source (**equation 13**) may, if low values are obtained, indicate the presence of the de-randomising effects, or if large, of additional sources of variation.

In the present case of ^{133}Ba, some of the count data showed a correlation with time, giving a near to significant chi-squared result. Ideally, these correlations would have been removed with attention to experimental techniques. Since they were not, the overall chi-squared test, computed for the goodness of fit of the extrapolation line, reflects these correlations.

B3. RESULTS OF THE ^{133}Ba ANALYSIS

The results for each data point and ensemble are tabulated in **table B1**. A plot of the efficiency function versus the radioactivity concentration is given in **figure B1**.

The value of radioactivity concentration obtained by extrapolation to 100 per cent β efficiency was 419.8 Bq mg^{-1}, with a standard deviation of 0.29 Bq mg^{-1}. The overall chi-squared result for the goodness of fit was 3.10 on 147 degrees of freedom. Slope of the extrapolation line was 15.909 and the grand mean of all observations was 424.31. The data for source reference number 1001 had a large chi-squared result for the β count. With these data excluded, the overall chi-squared results for goodness of fit fell to 2.60 on 137 degrees of freedom. The radioactivity concentration was then 420.22 ± 0.31

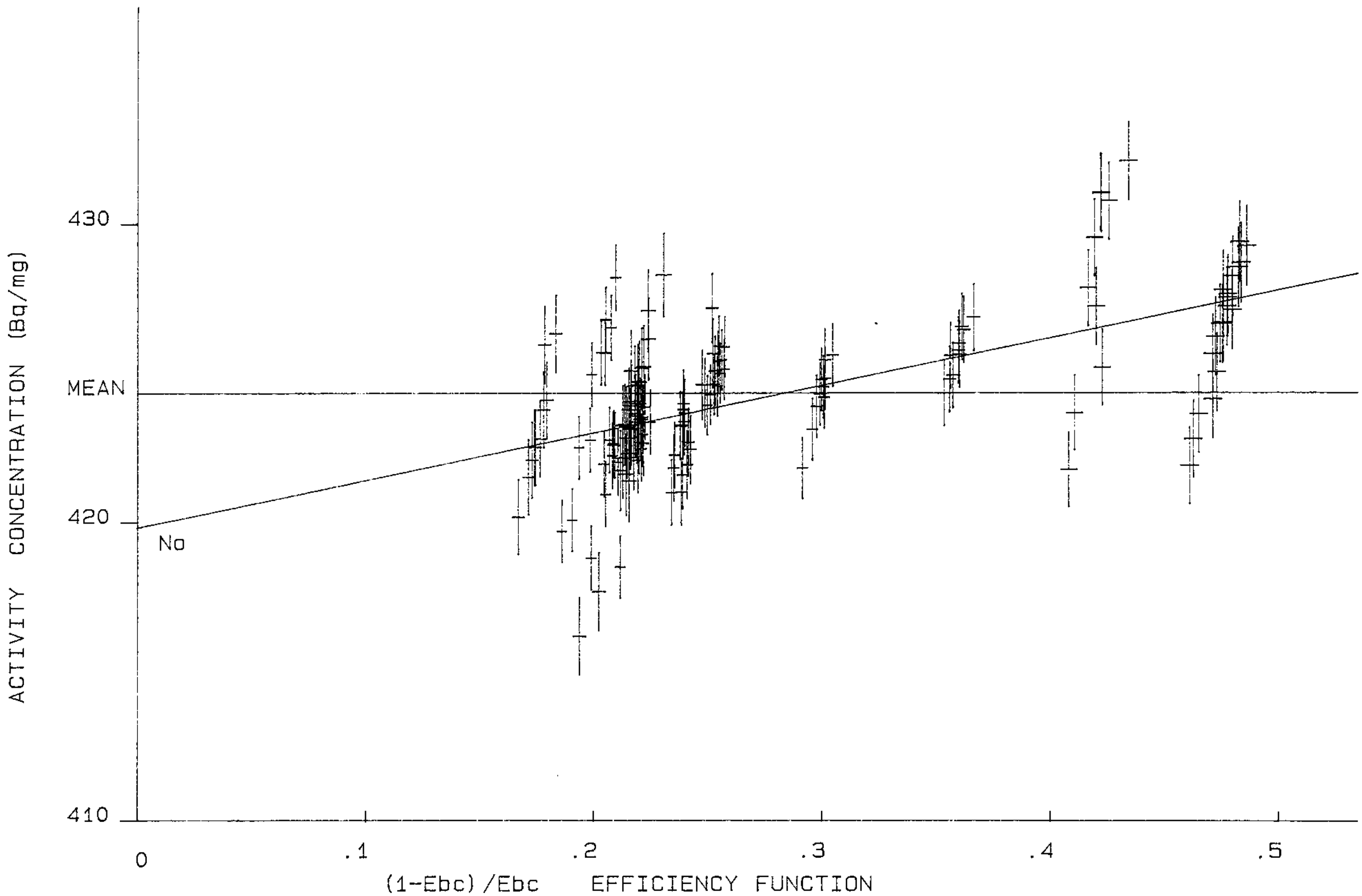

Figure B1 Efficiency function plotted against activity concentration for a range of β-particle detection efficiencies. The data comprise 149 observations of (N_c, N_β and N_γ) for ^{133}Ba. The line of best fit is extrapolated to intersect the activity concentration axis at N_o, which then represents an estimate of N_o for $\varepsilon_\beta = 1$. The mean value of all activity concentrations is also shown.

TABLE B1
COINCIDENCE COUNTING DATA FOR ^{133}Ba.

Tabulated are beta, gamma and coincidence counts, source activity, beta and gamma detection efficiencies, activity concentration and its variance, the efficiency function and its variance and the covariance of the efficiency function and activity concentration. Symbols are those defined in the main text.

Source reference number: 1339 **Mass (mg):** 12.443 **Count time:** 100 s
Background (counts s^{-1}) : beta 1.867 , gamma 25.970 , coincidence 0.040
Chi-squared : beta 0.8 , gamma 1.4 , coincidence 1.3 **Degrees of freedom**: 7

Nb	Ng	Nc	No	Ebc	Egc	Y	Var(Y)	X	Var(X)	Cov(X,Y)
402498	33198	24034	5123	0.824	0.059	421.6	1.8	0.214	0.89E-005	+0.0035
401883	33534	24274	5120	0.823	0.060	421.4	1.8	0.215	0.89E-005	+0.0035
403488	33572	24318	5138	0.823	0.060	422.8	1.8	0.214	0.88E-005	+0.0035
403293	33838	24498	5141	0.822	0.060	423.1	1.8	0.216	0.88E-005	+0.0035
402738	33254	24053	5132	0.823	0.059	422.3	1.9	0.215	0.89E-005	+0.0036
403577	33474	24146	5159	0.820	0.059	424.6	1.9	0.219	0.91E-005	+0.0036
402980	33642	24300	5147	0.821	0.060	423.6	1.9	0.218	0.90E-005	+0.0036
402174	33220	23928	5145	0.819	0.059	423.4	1.9	0.220	0.93E-005	+0.0037
402591	33450	24138	5144	0.821	0.060	423.4	1.9	0.219	0.91E-005	+0.0036

Source reference number: 1201 **Mass (mg)**: 12.582 Count time: 100 s
Background (counts s^{-1}) : beta 1.220 , gamma 25.527 , coincidence 0.043
Chi-squared : beta 0.8 , gamma 1.3 , coincidence 0.8 **Degrees of freedom**: 8

Nb	Ng	Nc	No	Ebc	Egc	Y	Var(Y)	X	Var(X)	Cov(X,Y)
409399	34661	25119	5232	0.821	0.061	424.6	1.8	0.218	0.87E-005	+0.0035
408891	34062	24758	5203	0.825	0.060	422.2	1.8	0.213	0.85E-005	+0.0034
409443	33941	25048	5130	0.838	0.061	416.2	1.7	0.194	0.74E-005	+0.0030
408738	34410	24958	5217	0.822	0.061	423.3	1.8	0.216	0.86E-005	+0.0034
408098	34132	25029	5148	0.832	0.061	417.7	1.7	0.202	0.79E-005	+0.0032
409833	34260	24908	5216	0.825	0.060	423.2	1.8	0.213	0.85E-005	+0.0034
409610	34238	24875	5217	0.824	0.060	423.3	1.8	0.213	0.85E-005	+0.0034
408925	34395	24790	5252	0.817	0.060	426.1	1.9	0.224	0.91E-005	+0.0036
408823	34069	24784	5198	0.825	0.060	421.7	1.8	0.212	0.84E-005	+0.0034
408314	34188	24717	5225	0.820	0.060	424.0	1.8	0.220	0.89E-005	+0.0035

Source reference number: 1107 **Mass (mg):** 20.999 Count time: 100 s
Background (counts s^{-1}) : beta 2.071, gamma 24.975 , coincidence 0.046
Chi-squared : beta 1.2 , gamma 2.0 , coincidence 1.0 **Degrees of freedom:** 8

Nb	Ng	Nc	No	Ebc	Egc	Y	Var(Y)	X	Var(X)	Cov(X,Y)
641447	55548	39529	8607	0.805	0.061	422.4	1.3	0.242	0.58E-005	+0.0024
643495	55903	39485	8702	0.799	0.061	427.2	1.3	0.252	0.62E-005	+0.0026
641281	56309	39789	8671	0.799	0.061	425.7	1.3	0.252	0.61E-005	+0.0025
640030	55426	39110	8660	0.798	0.061	425.1	1.3	0.253	0.63E-005	+0.0026
641103	55345	39183	8645	0.801	0.061	424.3	1.3	0.249	0.61E-005	+0.0025
641918	55250	39414	8590	0.807	0.061	421.6	1.3	0.239	0.57E-005	+0.0024
641408	55210	39405	8578	0.808	0.061	421.0	1.3	0.238	0.57E-005	+0.0024
642216	55462	39490	8612	0.806	0.061	422.7	1.3	0.241	0.58E-005	+0.0024
641486	55447	39503	8597	0.806	0.061	421.9	1.3	0.241	0.58E-005	+0.0024
642050	55381	39242	8651	0.802	0.061	424.6	1.3	0.247	0.60E-005	+0.0025

Source reference number: 926 **Mass (mg):** 21.244 Count time: 100 s
Background (counts s^{-1}) : beta 1.021 , gamma 27.507 , coincidence 0.054
Chi-squared : beta 0.3 , gamma 0.8 , coincidence 1.1 **Degrees of freedom:** 8

Nb	Ng	Nc	No	Ebc	Egc	Y	Var(Y)	X	Var(X)	Cov(X,Y)
654972	58273	41456	8772	0.808	0.063	423.3	1.2	0.238	0.54E-005	+0.0023
654630	58374	41471	8780	0.807	0.063	423.7	1.2	0.240	0.54E-005	+0.0023
653673	58327	41635	8725	0.810	0.063	421.0	1.2	0.234	0.52E-005	+0.0022
654356	58719	41892	8742	0.810	0.063	421.8	1.2	0.235	0.52E-005	+0.0022
655022	58094	41276	8782	0.807	0.062	423.8	1.2	0.239	0.54E-005	+0.0023
654382	58340	41458	8774	0.807	0.063	423.4	1.2	0.239	0.54E-005	+0.0023
654439	58251	41407	8772	0.807	0.063	423.2	1.2	0.239	0.54E-005	+0.0023
655314	58278	41413	8786	0.807	0.063	424.0	1.2	0.239	0.54E-005	+0.0023
654760	58779	41921	8751	0.810	0.063	422.2	1.2	0.235	0.52E-005	+0.0022
654923	58547	41663	8771	0.808	0.063	423.2	1.2	0.238	0.54E-005	+0.0023

Source reference number: 956 **Mass (mg):** 22.196 Count time: 100 s
Background (counts s^{-1}) : beta 1.020 , gamma 27.697 , coincidence 0.051
Chi-squared : beta 1.1 , gamma 0.5 , coincidence 0.7 **Degrees of freedom:** 8

Nb	Ng	Nc	No	Ebc	Egc	Y	Var(Y)	X	Var(X)	Cov(X,Y)
687744	60637	43950	9055	0.826	0.063	418.5	1.1	0.211	0.42E-005	+0.0019
688150	60788	43592	9159	0.817	0.063	423.3	1.2	0.225	0.47E-005	+0.0020
689738	61134	44023	9144	0.820	0.063	422.7	1.1	0.220	0.45E-005	+0.0020
688679	60685	43620	9143	0.819	0.063	422.6	1.1	0.222	0.46E-005	+0.0020
689371	60760	43689	9150	0.819	0.063	422.9	1.1	0.221	0.45E-005	+0.0020
688868	60682	43649	9139	0.819	0.063	422.4	1.1	0.221	0.45E-005	+0.0020
690136	61018	44014	9133	0.822	0.063	422.2	1.1	0.217	0.44E-005	+0.0020
690149	60786	43665	9170	0.818	0.063	423.9	1.1	0.222	0.46E-005	+0.0020
689078	60567	43614	9131	0.820	0.063	422.1	1.1	0.219	0.45E-005	+0.0020
690122	60627	43578	9162	0.819	0.063	423.5	1.1	0.221	0.45E-005	+0.0020

Source reference number: 1120 **Mass (mg):** 25.021 Count time: 100 s
Background (counts s^{-1}) : beta 1.020 , gamma 27.697 , coincidence 0.051
Chi-squared : beta 1.0 , gamma 2.0 , coincidence 0.7 **Degrees of freedom:** 8

Nb	Ng	Nc	No	Ebc	Egc	Y	Var(Y)	X	Var(X)	Cov(X,Y)
651719	67434	41050	10266	0.685	0.062	421.9	1.7	0.461	0.14E-004	+0.0044
652350	67833	41258	10287	0.684	0.063	422.8	1.7	0.462	0.14E-004	+0.0044
652081	68141	40876	10428	0.674	0.062	428.7	1.8	0.483	0.15E-004	+0.0048
651889	68608	41104	10441	0.673	0.062	429.2	1.8	0.486	0.15E-004	+0.0048
653976	68448	41275	10406	0.678	0.062	427.8	1.7	0.476	0.15E-004	+0.0046
651464	68261	41062	10390	0.676	0.062	427.1	1.7	0.480	0.15E-004	+0.0047
652532	68419	41273	10379	0.678	0.063	426.6	1.7	0.475	0.15E-004	+0.0046
651290	68586	41454	10340	0.679	0.063	425.0	1.7	0.473	0.15E-004	+0.0045
652492	67962	40977	10380	0.678	0.062	426.7	1.7	0.476	0.15E-004	+0.0046
653219	68192	41213	10369	0.679	0.062	426.2	1.7	0.472	0.15E-004	+0.0046

Source reference number: 1202 **Mass (mg):** 25.354 Count time: 100 s
Background (counts s^{-1}) : beta 0.999 , gamma 27.948 , coincidence 0.049
Chi-squared : beta 0.8 , gamma 2.3 , coincidence 1.2 **Degrees of freedom:** 8

Nb	Ng	Nc	No	Ebc	Egc	Y	Var(Y)	X	Var(X)	Cov(X,Y)
660442	67917	41187	10442	0.683	0.062	423.6	1.7	0.465	0.14E-004	+0.0045
660208	68650	41169	10561	0.675	0.062	428.5	1.7	0.482	0.15E-004	+0.0047
660756	68687	41321	10536	0.677	0.062	427.5	1.7	0.477	0.15E-004	+0.0046
660849	68983	41504	10538	0.677	0.062	427.6	1.7	0.478	0.15E-004	+0.0046
660880	69357	41836	10514	0.678	0.063	426.6	1.7	0.474	0.14E-004	+0.0045
660451	69153	41624	10529	0.677	0.062	472.2	1.7	0.477	0.15E-004	+0.0046
660759	68548	41414	10491	0.680	0.062	425.6	1.7	0.471	0.14E-004	+0.0045
660835	68906	41397	10553	0.676	0.062	428.2	1.7	0.480	0.15E-004	+0.0047
661200	69000	41370	10581	0.674	0.062	429.4	1.7	0.483	0.15E-004	+0.0047
658619	69011	41718	10453	0.680	0.063	424.1	1.7	0.471	0.14E-004	+0.0045

Source reference number: 1355 **Mass (mg):** 30.877 Count time: 100 s
Background (counts s^{-1}) : beta 1.197 , gamma 27.817 , coincidence 0.039
Chi-squared : beta 1.0 , gamma 0.9 , coincidence 1.2 **Degrees of freedom:** 8

Nb	Ng	Nc	No	Ebc	Egc	Y	Var(Y)	X	Var(X)	Cov(X,Y)
847105	82868	53221	12746	0.735	0.062	425.8	1.2	0.360	0.72E-005	+0.0027
845362	82830	52953	12778	0.732	0.062	426.8	1.2	0.366	0.75E-005	+0.0028
847636	82504	52915	12770	0.735	0.062	426.5	1.2	0.361	0.73E-005	+0.0027
846986	82746	53235	12722	0.737	0.062	424.9	1.2	0.357	0.72E-005	+0.0027
848034	83167	53658	12704	0.739	0.062	424.3	1.2	0.353	0.70E-005	+0.0026
847389	82724	53115	12753	0.735	0.062	426.0	1.2	0.360	0.73E-005	+0.0027
848779	82281	52954	12742	0.737	0.062	425.6	1.2	0.356	0.72E-005	+0.0027
847566	82950	53424	12718	0.738	0.062	424.8	1.2	0.356	0.71E-005	+0.0027
846621	82339	52855	12742	0.735	0.062	425.6	1.2	0.360	0.73E-005	+0.0027
847034	82679	53007	12766	0.734	0.062	426.4	1.2	0.362	0.73E-005	+0.0027

Source reference number: 1227 **Mass (mg):** 31.180 Count time: 100 s
Background (counts s^{-1}) : beta 1.474 , gamma 27.248 , coincidence 0.040
Chi-squared : beta 0.4 , gamma 0.8 , coincidence 0.9 **Degrees of freedom:** 8

Nb	Ng	Nc	No	Ebc	Egc	Y	Var(Y)	X	Var(X)	Cov(X,Y)
886747	83535	55865	12826	0.770	0.062	424.3	1.1	0.299	0.52E-005	+0.0021
886349	84222	56250	12841	0.769	0.063	424.8	1.1	0.301	0.52E-005	+0.0021
887523	83989	56083	12859	0.769	0.062	425.4	1.1	0.301	0.52E-005	+0.0021
886848	83546	55933	12813	0.771	0.062	423.9	1.1	0.298	0.51E-005	+0.0021
886768	83857	56415	12752	0.774	0.063	421.8	1.0	0.291	0.49E-005	+0.0020
886535	83461	55954	12791	0.772	0.062	423.1	1.1	0.296	0.51E-005	+0.0021
887213	83650	55912	12840	0.769	0.062	424.8	1.1	0.300	0.52E-005	+0.0021
886286	83478	55765	12833	0.769	0.062	424.5	1.1	0.300	0.52E-005	+0.0021
885415	83565	55816	12823	0.769	0.062	424.2	1.1	0.301	0.52E-005	+0.0021
885795	83573	55664	12864	0.767	0.062	425.6	1.1	0.305	0.53E-005	+0.0022

Source reference number: 1659 **Mass (mg):** 12.443 Count time: 100 s
Background (counts s^{-1}) : beta 0.503 , gamma 24.526 , coincidence 0.019
Chi-squared : beta 1.0 , gamma 1.1 , coincidence 1.1 **Degrees of freedom:** 8

Nb	Ng	Nc	No	Ebc	Egc	Y	Var(Y)	X	Var(X)	Cov(X,Y)
403945	33997	24745	5149	0.823	0.061	423.8	1.8	0.215	0.87E-005	+0.0035
403742	34054	24769	5151	0.822	0.061	423.9	1.8	0.216	0.87E-005	+0.0035
403420	34325	24889	5166	0.819	0.061	425.2	1.8	0.221	0.89E-005	+0.0035
404277	34121	24671	5189	0.817	0.061	427.1	1.9	0.224	0.92E-005	+0.0036
402576	34187	24817	5148	0.820	0.061	423.6	1.8	0.219	0.89E-005	+0.0035
403222	33849	24501	5167	0.819	0.060	425.2	1.9	0.222	0.91E-005	+0.0036
404814	34312	24970	5165	0.822	0.061	425.1	1.8	0.216	0.86E-005	+0.0035
403213	34066	24495	5204	0.813	0.060	428.3	1.9	0.230	0.96E-005	+0.0038
403749	33711	24541	5142	0.824	0.060	423.2	1.8	0.214	0.87E-005	+0.0035
403392	33982	24645	5161	0.820	0.061	424.7	1.9	0.220	0.90E-005	+0.0036

Source reference number: 1039 **Mass (mg):** 12.582 Count time: 100 s
Background (counts s^{-1}) : beta 1.149 , gamma 25.631 , coincidence 0.047
Chi-squared : beta 0.9 , gamma 1.9 , coincidence 1.0 **Degrees of freedom:** 8

Nb	Ng	Nc	No	Ebc	Egc	Y	Var(Y)	X	Var(X)	Cov(X,Y)
420253	34236	25574	5204	0.849	0.061	423.8	1.6	0.178	0.64E-005	+0.0028
419852	34157	25544	5192	0.850	0.060	422.8	1.6	0.176	0.64E-005	+0.0027
420372	34048	25564	5177	0.854	0.060	421.5	1.6	0.171	0.61E-005	+0.0027
420856	34329	25533	5235	0.845	0.060	426.3	1.7	0.183	0.67E-005	+0.0029
420288	34012	25470	5189	0.852	0.060	422.5	1.6	0.174	0.63E-005	+0.0027
422136	34073	25426	5231	0.849	0.060	425.9	1.6	0.178	0.65E-005	+0.0028
420430	33854	25350	5189	0.852	0.060	422.5	1.6	0.174	0.63E-005	+0.0027
420106	33722	25130	5208	0.848	0.059	424.1	1.6	0.179	0.66E-005	+0.0028
420645	33524	25233	5161	0.857	0.060	420.2	1.6	0.167	0.60E-005	+0.0026
420371	33735	25275	5184	0.853	0.060	422.1	1.6	0.173	0.63E-005	+0.0027

Source reference number: 1149 **Mass (mg):** 20.999 Count time: 100 s
Background (counts s^{-1}) : beta 1.149 , gamma 25.631 , coincidence 0.047
Chi-squared : beta 1.3 , gamma 1.0 , coincidence 1.2 **Degrees of freedom:** 8

Nb	Ng	Nc	No	Ebc	Egc	Y	Var(Y)	X	Var(X)	Cov(X,Y)
658711	54567	39854	8595	0.830	0.060	421.9	1.2	0.204	0.45E-005	+0.0020
657052	54410	39928	8531	0.834	0.060	418.8	1.2	0.199	0.43E-005	+0.0020
655832	54815	39864	8596	0.826	0.060	422.0	1.2	0.210	0.47E-005	+0.0021
657656	55028	40080	8608	0.827	0.060	422.6	1.2	0.209	0.46E-005	+0.0021
657333	55052	40117	8600	0.828	0.060	422.2	1.2	0.208	0.46E-005	+0.0021
656355	54442	39423	8637	0.823	0.060	424.0	1.2	0.215	0.49E-005	+0.0022
656991	54986	40167	8574	0.830	0.061	420.9	1.2	0.205	0.45E-005	+0.0020
657689	54795	39908	8607	0.828	0.060	422.6	1.2	0.208	0.46E-005	+0.0021
657324	54695	39810	8607	0.827	0.060	422.6	1.2	0.209	0.46E-005	+0.0021
658736	54947	40072	8611	0.829	0.060	422.8	1.2	0.207	0.45E-005	+0.0020

Source reference number: 951 **Mass (mg):** 21.244 Count time: 100 s
Background (counts s^{-1}) : beta 1.127 , gamma 26.029 , coincidence 0.052
Chi-squared : beta 1.4 , gamma 3.0 , coincidence 1.2 **Degrees of freedom:** 8

Nb	Ng	Nc	No	Ebc	Egc	Y	Var(Y)	X	Var(X)	Cov(X,Y)
675375	57635	42281	8790	0.834	0.062	424.9	1.1	0.199	0.40E-005	+0.0019
674689	56709	41762	8741	0.838	0.061	422.5	1.1	0.193	0.39E-005	+0.0018
672758	56656	41839	8691	0.840	0.062	420.1	1.1	0.190	0.38E-005	+0.0018
674487	56309	41713	8684	0.843	0.061	419.7	1.1	0.186	0.37E-005	+0.0018
674884	57064	41632	8828	0.830	0.061	426.8	1.2	0.205	0.43E-005	+0.0020
674347	57096	41730	8806	0.831	0.061	425.7	1.2	0.203	0.42E-005	+0.0019
673304	57263	41713	8823	0.828	0.061	426.5	1.2	0.208	0.44E-005	+0.0020
674708	57445	41778	8857	0.827	0.061	428.2	1.2	0.209	0.44E-005	+0.0020
673440	56807	41453	8806	0.830	0.061	425.7	1.2	0.205	0.43E-005	+0.0020
672576	56589	41514	8746	0.835	0.061	422.7	1.1	0.198	0.41E-005	+0.0019

Source reference number: 1001 **Mass (mg):** 25.021 Count time: 100 s
Background (counts s^{-1}) : beta 1.091 , gamma 26.049 , coincidence 0.038
Chi-squared : beta 13.2 , gamma 2.0 , coincidence 0.9 **Degrees of freedom:** 8

Nb	Ng	Nc	No	Ebc	Egc	Y	Var(Y)	X	Var(X)	Cov(X,Y)
671260	66276	41379	10328	0.703	0.061	425.2	1.6	0.423	0.12E-004	+0.0041
672632	66134	41702	10246	0.710	0.061	421.8	1.5	0.408	0.12E-004	+0.0039
674264	66027	41548	10292	0.709	0.061	423.7	1.6	0.410	0.12E-004	+0.0039
675291	66077	41302	10377	0.704	0.060	427.2	1.6	0.420	0.12E-004	+0.0041
676128	67019	41505	10492	0.697	0.061	432.1	1.7	0.434	0.13E-004	+0.0042
677929	66692	41530	10461	0.702	0.061	430.7	1.7	0.425	0.12E-004	+0.0041
678912	66794	41773	10432	0.705	0.061	429.5	1.6	0.419	0.12E-004	+0.0040
679933	66102	41239	10468	0.703	0.060	431.0	1.7	0.422	0.12E-004	+0.0041
677671	66712	41805	10391	0.706	0.061	427.8	1.6	0.416	0.12E-004	+0.0040
679781	66624	41573	10467	0.703	0.060	431.0	1.6	0.422	0.12E-004	+0.0041

Source reference number: 1204 **Mass (mg):** 31.180 Count time: 100 s
Background (counts s^{-1}) : beta 1.135 , gamma 25.846 , coincidence 0.040
Chi-squared : beta 0.9 , gamma 0.5 , coincidence 0.8 **Degrees of freedom:** 8

Nb	Ng	Nc	No	Ebc	Egc	Y	Var(Y)	X	Var(X)	Cov(X,Y)
905915	82388	56818	12723	0.796	0.062	425.1	1.0	0.257	0.40E-005	+0.0018
907165	82180	56705	12733	0.796	0.062	425.4	1.0	0.256	0.40E-005	+0.0018
907161	82316	56745	12746	0.795	0.062	425.9	1.0	0.257	0.40E-005	+0.0018
906801	81941	56570	12720	0.797	0.062	425.0	1.0	0.255	0.40E-005	+0.0018
907744	82069	56769	12709	0.798	0.062	424.6	1.0	0.253	0.39E-005	+0.0018
908146	82572	57202	12698	0.799	0.062	424.3	1.0	0.251	0.39E-005	+0.0017
906710	82128	56754	12707	0.797	0.062	424.6	1.0	0.254	0.40E-005	+0.0018
908353	82526	57229	12688	0.800	0.062	423.9	1.0	0.250	0.38E-005	+0.0017
908161	82331	56882	12731	0.797	0.062	425.4	1.0	0.254	0.40E-005	+0.0018
908784	82141	56718	12747	0.797	0.062	425.9	1.0	0.255	0.40E-005	+0.0018