

Chapter 18

Accuracy and Merit in Liquid Scintillation Counting

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INTRODUCTION – STATES OF THE ART

The practical accuracy of any analytical method is revealed when a number of laboratories cooperate in the analysis of a common (standard) sample of complex composition. Such was the case, for example, in a recent intercomparison organised by the National Bureau of Standards, for the analysis of calcium in human serum. Two sets of results are shown in Fig. 1. Those to the left of the diagram were obtained by laboratories using a standard reference material (SRM) and adhering to the carefully defined protocol of the reference method (RM) for calcium.^{1,2} When the same sample was sent 'blind' to hospital and commercial laboratories, the results to the right of Fig. 1 were obtained: three-quarters of the (12) results were in error by more than the desired medical limit of uncertainty, and one-third of these could have resulted in dangerous mis-diagnoses. Upon investigation, it was learned that the relatively large errors were connected with factors other than the particular method of analysis employed.

This example is representative of what one finds also in the field of radiochemical analysis and liquid scintillation counting. The precision and accuracy of measurements on difficult samples – such as complex biological materials or those containing trace levels of radioactivity – tend to be far more dependent upon the skill and knowledge of the scientist than upon the specific analytical method. Regrettably, erroneous results for just such samples are likely to have the greatest practical consequence.

In order to assess the current performance in the field, the literature was examined for actual limits of intra- and interlaboratory errors as deduced from replication, analyses of standards and intercomparison exercises. As shown in Fig. 2, it was necessary to partition the measurements into three broad classes according to skill of the scientist ('laboratory') (*and* the quality of his equipment), the complexity of the sample and the level of radioactivity. The relative error is seen to vary by about a decade from class to class, and the inaccuracy, as measured by the interlaboratory errors, is many-fold greater than the imprecision. The importance of Fig. 2 is that it indicates what is currently possible with liquid scintillation counting – i.e. class 1 (exemplary) – given sufficient skill and care; at the same time, it indicates what is

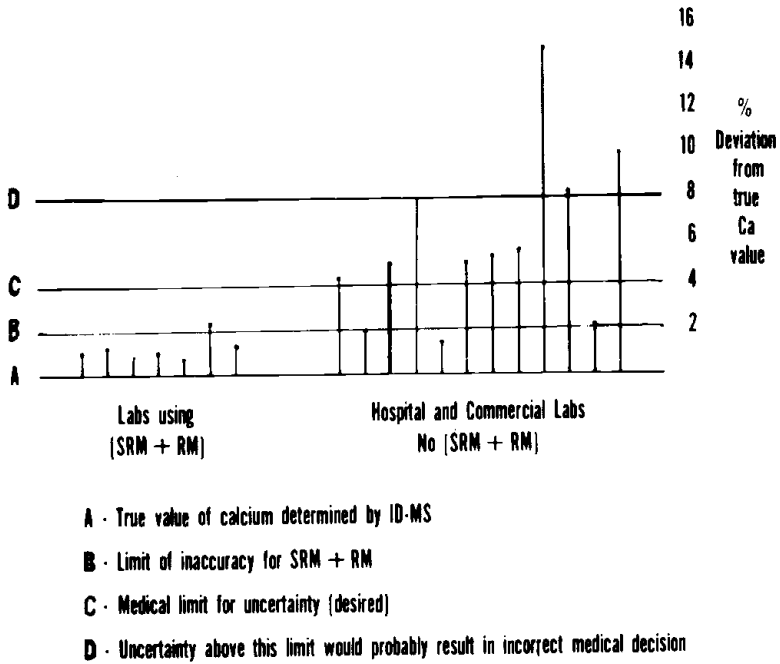


Fig. 1. Calcium in human serum.^{1,2} Results obtained in connection with the development of a referee method by the National Bureau of Standards. Seven results to the left of the diagram were obtained through the use of the referee method (RM) and a standard reference material (SRM); those to the right (same sample) involved neither.

actually achieved under less optimal circumstances. The potential benefits of paying close attention to careful sample preparation and possible sources of error — to be discussed below — are thus made manifest; and the danger of basing uncertainty intervals and performance (merit) wholly upon Poisson counting statistics becomes evident. Illustrations of the three performance classes will be given in the following paragraphs.

Vaninbroukx and Stanef³ reviewed results of high accuracy liquid scintillation counting (*class 1*), typically as performed by national standardising laboratories. These authors quoted overall calibration errors (inaccuracies) ranging from 0.1% for α -emitters to 1–3% for low-energy β -emitters. Imprecision was generally two to ten times smaller. Typical of *class 2* are the many laboratories engaged in careful measurements of ^{14}C or ^3H in natural materials at low and intermediate levels. For example, an IAEA-sponsored intercomparison (35 laboratories) of tritium-spiked water samples (10–250 TU) yielded relative standard deviations ranging from 2.6% to 25%, depending upon the concentration and method employed: gas or liquid scintillation counting, with or without electrolytic enrichment.⁴ A significant conclusion from the study was that a residual error of about 3.5% arose from the enrichment process itself. The importance of intercomparisons and standard sample analysis for radiocarbon ages was cited by Kim,⁵ who reported a relative error of 5% from the analysis of 80 interlaboratory cross-check samples. Inter- and

Laboratory; Procedure	Sample	Relative Error	
		<i>intralab</i>	<i>interlab</i>
Exemplary	Pure radionuclides ideal preparations ($S \gg B$)	$\lesssim 0.3\%$	$\lesssim 2\%$
Experienced	Restricted nuclides and matrices, careful preparation	$\lesssim 5\%$	$\lesssim 20\%$
Less experienced	Varied and mixed nuclides, complex matrices ($S \lesssim B$)	$\lesssim 50\%$	$\lesssim 10^2$

Fig. 2. States of the art. Liquid scintillation performance classes.

intralaboratory variations in measurements of natural radiocarbon were evaluated also by Polach,⁶ who demonstrated the significantly greater dispersion for comparative measurements *among* laboratories – apparently due to contamination and isotopic fractionation during sample preparation.

The results of routine inter- or intralaboratory comparisons should not be interpreted as limiting. When higher accuracy is demanded, it may be met by taking special precautions: Glass⁷ noted the achievement of better than 0.5% accuracy ('95% confidence limits') for industrial ¹⁴C and ³H measurements, and Downes⁸ succeeded in achieving 0.4% agreement among relative measurements in which ¹⁴C was employed to determine the mean diameter of wool fibres.

Radiochemical measurements at low levels in complex matrices (*class 3*) present difficulties, particularly when several radionuclides are present and when less experienced workers participate. Because of the importance of such measurements, however, a number of intercalibration exercises have been sponsored by the International Laboratory of Marine Radioactivity of the International Atomic Energy Agency. Though the results of these exercises may appear discouraging, the chief objective is being accomplished: education and improvement of the performance of the participants. One example, taken from one of the earliest exercises,⁹ concerns the measurement of ¹⁰⁶Ru (and other fission products) in seawater. The 22 positive results which were reported for sample SW-1-2 ranged from 0.3 to 62 pCi/kg! Restricting one's attention to the 16 laboratories reporting relative uncertainties smaller than 10% narrows the range to about a factor of six. (Serious conceptual or experimental systematic errors on the part of four of the participants is believed to account for most of this residual range.)

The foregoing examples demonstrate the importance of adequate internal and external quality control (QC) in order to achieve and maintain the desired level of accuracy. External QC, in the form of 'blind' measurements of standard materials and interlaboratory comparisons, provides a direct measure of practical accuracy in the field.^{10,11} Internal QC, on the other hand, is a prerequisite for the production of stable, meaningful measurements by any laboratory. Both are essential for protection against blunders

(mistakes in experiment *or* theory) which may occur at any step of the measurement process.

The great utility of control chart techniques for internal QC in liquid scintillation counting is well illustrated in the article by Wyld.¹² Redundancy can play a very important role in signalling systematic error, as shown by Glass⁷ and Mueller¹³ through the use of the double ratio technique. Computation of the sample counting efficiency via two independent measures – sample channels ratio (SCR), and automatic external standard counts (AES) or external standard channels ratio (ESCR) – provides protection against occasional blunders (misprints) as well as systematic error (assumed homogeneity or electron density). Redundancy (replication) is important also with respect to random error. Many a pitfall awaits those who would assume that random error is determined strictly by Poisson counting statistics.¹⁴

In the following two sections we shall review some of the primary sources of error in liquid scintillation counting and examine the question of merit, particularly as related to optimisation and the selection of alternative counting systems. In addition to specific references which will be given in the next section, there are a number of texts and review articles which treat the problem of liquid scintillation errors. Apart from the basic text by Birks,¹⁶ most of the volumes are proceedings of international conferences which have been held more or less annually since 1969.^{16–21} The volume edited by Bransome¹⁵ has been especially helpful because of its subdivision in accordance with the various aspects of the measurement process. Major sections on 'The Labeled Sample' and 'Quenching', for example, contain a wealth of information about individual sources of error. Excellent summaries and guides to the literature are found in the reviews by Parmentier and ten Haaf²² and Vaninbroukx and Stanef.³

THE MEASUREMENT PROCESS; SOURCES OF ERROR

A systematic assessment of the physics and chemistry underlying the overall liquid scintillation measurement process would be required to reveal all potential error sources. Such an assessment is complementary to analytical quality control (replication, measurement of standards, intercomparison exercises, etc.) in that causative factors would be identified and to some extent regulated. Principal steps of the measurement process include *sampling*, *sample preparation* – including chemical or physical purification and final preparation for measurement – *instrumental measurement*, and *data reduction and reporting*. Throughout, one must face the problem of *stability*: stability of the measuring device, stability of the sample and stability of the background. Obviously, several of the measurement steps are not peculiar to liquid scintillation counting, but they must nevertheless be considered if one is concerned with overall accuracy. Poisson counting statistics, similarly, is common to all methods of radioactivity measurement (pulse counting); and for all, it sets the lower limit to overall imprecision.

Rather than attempt an assessment of errors in each step of the measurement process, we shall, in the following text, focus upon the special problem in liquid scintillation counting: the fact that the sample itself is an integral part of the measurement device. The chemical and physical state of the sample, as well as its stability, can thus exert a profound effect on the instrument calibration (detection efficiency) and the background rate. The central issue of errors in the measurement step will therefore be discussed, after some brief comments on sampling and preparation.

Sampling and sample preparation

Sampling errors set a basic limit to the accuracy of any measurement process. Only when the entire sample is taken for analysis may such errors be presumed absent. For the most part, especially in biological experiments, the entire sample may not (even in principle) be taken, and sampling variability or even gross sampling errors (blunders) may far exceed all other sources of error. To cite just one example: measurements by Tyler *et al.*²³ of ^{14}C activity in animal tissue exhibited 2.5 times more random error in sampling than in counting. These authors demonstrated also the dependence of sampling error on sample size, as well as the effect of operator experience on overall accuracy.

Sample preparation generally leads to more reliable measurement if purification steps are involved. Apart from contamination and efficiency and stability questions, which will be discussed below, chemical (or physical) separation may, however, lead to losses and altered isotope ratios — particularly for ^3H and ^{14}C . If uncorrected, such factors can produce bias; and even if corrected, overall imprecision will be somewhat increased. Recovery (yield) correction via 'carrier' or stable- or radioisotope dilution is well known to analysts and to radiochemists. Stable strontium or ^{85}Sr , for example, has long been used in the radiochemical determination of ^{90}Sr . Isotope fractionation problems are, perhaps, less well known, except to those who routinely measure low levels of ^3H and ^{14}C . In the radiocarbon dating community fractionation effects are regularly corrected by mass spectrometric measurements of $\delta^{13}\text{C}$.²⁴ Without such corrections, radiocarbon measurements may exhibit a limiting (minimum) variability of at least 0.5%. Random variations in the isotopic enrichment factor for electrolytically enriched tritium have been shown to limit the overall accuracy of low-level liquid scintillation measurements of tritium to about 3%, largely due to differences among electrolysis cells.⁴ Because of this source of error, Sauzay and Schell²⁵ found improved accuracy *without* enrichment for liquid scintillation counting of water samples having tritium activities exceeding 500 TU.

Measurement

The major opportunity for error in liquid scintillation counting is in connection with the measurement step. Under ideal circumstances the error is entirely random and may be computed from Poisson counting statistics. In real circumstances, however, the counting mixture may be unstable or it may be otherwise non-identical with that used in calibration. Consequently, assumed efficiencies and backgrounds may be incorrect.

(1) **Instability.** This occurs when the counting mixture changes its composition as a result of (i) phase separation or precipitation, or (ii) losses to the vial walls (adsorption), through the walls (diffusion) or to the surrounding atmosphere (evaporation). Besides composition-related variability, one must sometimes pay attention to other sources, such as electronic instability, luminescence decay and external background variations. The stability of heterogeneous counting preparations (suspensions, colloids) has been studied by Fox,²⁶ van der Laarse²⁷ and others. Visual appearance and apparent (physical) stability have been shown to be unreliable as indicators of counting stability, particularly for tritium. As a result, Fox introduced the Instrument Merit Stability Quotient (MISQ) as a measure of the quality of aqueous colloids of varying composition, where MISQ takes into account the observed *counting* stability, detection efficiency and volume percentage of water.

Adsorption of a radioactive solute is troublesome in that it changes the effective solid angle from about 4π steradians to 2π , thus decreasing the efficiency by a

factor of two. Amounts and rates of adsorption depend upon the nature of the vial surface, the scintillation cocktail, and the chemical structure and specific activity of the labelled species.²⁸ In a concise review of the topic, Litt and Carter²⁹ demonstrated the prevalence and complexity of adsorption phenomena. They noted particularly the benefits from 'the addition of a pinch of carrier' and the use of plastic vials.

Though advantageous with respect to the adsorption problem, certain plastic vials have been responsible for long-term (\sim days) drifts in sample, external standard and background counting rates because of their permeability.^{25,30,31} Sample losses occur because of diffusion of the labelled species through the wall, and the background increases with time because the wall becomes a plastic scintillator, due to solvent plus solute diffusion. Rates of loss through polyethylene vials were reported by Horrocks³¹ as 130 mg/day for (tritiated) toluene and 5 mg/day for water. (The recommendations for controlling diffusion-induced errors, presented at the end of Horrocks' paper are well worth the reader's attention.)

Losses may occur also through evaporation. The problem is most serious when low activity levels dictate extended measurements, as in radiocarbon dating. For example, at the Australian National University, sample counts are generally accumulated for 1000 min each over a period of one week. The high rate of evaporation with commercially available vials — \sim 25 mg/day — led Polach³² to design a special small volume vial having a machined PTFE stopper. The rate of loss of solvent was thereby decreased to \sim 10 mg/week. Incidentally, when samples having different levels of activity remain together in the same sample changing chamber, one sample's losses become another sample's contamination.

(2) **Efficiency changes or quenching.** This must be anticipated whenever the sample composition or configuration differs in any way from that of the calibration standard. 'Quenching', which generally signifies decreased intrinsic detection efficiency, arises from diverse sources. Perhaps the most serious blunders (needless systematic errors) in liquid scintillation counting come about when improper quenching corrections are applied. In order to appreciate the nature of such errors, we must consider some of the causes and monitoring (correction) methods for quenching: (i) *causes* — 'chemical quenching' (impurity competition for the excitation energy of the solvent molecules), 'colour quenching' (molecular absorption of the fluorescence radiation) and 'photon quenching' (decreased photon yield due to self-absorption of the primary nuclear radiation); and (ii) *monitoring* — internal standard, sample channels ratio (SCR), automatic external standard count rate (AES) and external standard channels ratio (ESCR). Cerenkov counting has one major advantage over ordinary liquid scintillation counting, namely immunity to chemical quenching. The following paragraphs will illustrate a few of the pitfalls in quench monitoring; for further information the reader is advised to examine the comprehensive accounts of Parker and Elrick,³³ Neary and Budd³⁴ and Peng.³⁵

Errors may arise from a mismatch between quenching mechanism and monitoring method. If the degree of quenching is small and if the sample is in complete solution and stable, however, there should be little difficulty; all methods ought to yield similar results. Low count rates may limit the usefulness of the SCR method, however, because of the correspondingly large (Poisson) counting errors. The AES method has suffered problems in the past because of irreproducible source positioning and sensitivity to sample characteristics (volume, electron density) and vial characteristics.

Channels ratio quench corrections for homogeneous solutions derive from the fact that decreased efficiency commonly accompanies changes in the amplitude or shape

of the scintillation pulse height distribution. Unless the radionuclide, quenching agent and monitoring method are all fixed, however, faulty conclusions may result from the use of a single quenching curve. The major problem arises when one fails to distinguish between the effects of chemical and colour quenching, particularly if the external monitor is employed. The difference between colour versus chemical quenching curves (ESCR) can be quite significant both for ^{14}C and ^3H , as shown in the work of Noujaim *et al.*³⁶ Although these authors found little difference in SCR curves (colour versus chemical), it has been conclusively demonstrated³⁴ that for ^{14}C and higher energy β -emitters, significant systematic error will result if one does not pay attention to the type of quenching in highly quenched solutions. Unfortunately, in practice one may not know whether quenching is primarily chemical or due largely to coloured material, particularly when biological samples are involved. A promising approach to the problem is to require concordance among two or more different quenching monitors. Lange,³⁷ for example, computed ^{14}C efficiencies of 35.2% and 24.5% using the ESCR and 'chemical' or 'colour' quench correction curves, respectively, where chlorophyll was the quenching agent. Computations based upon AES curves yielded 30.7% and 25.2%, respectively. Concluding, from the consistency between results, that the quenching was largely due to colour was indeed the correct decision: the actual efficiency was 24.9%.

Errors due to heterogeneous counting samples (suspensions, precipitates, etc.) may also be signalled through the use of redundant quench monitoring. This is one of the major features of the 'double ratio' method as proposed by Bush³⁸ and as illustrated by Glass.⁷ When sample counts are sufficiently large, a plot of SCR versus ESCR serves to monitor homogeneity. Two important points must be borne in mind with respect to heterogeneous samples, however: (i) they may not *appear* to be heterogeneous;²⁷ and (ii) erroneous efficiencies can result from the use of *either* (SCR or ESCR) quenching correction curve.¹³

Addition of an internal standard will generally yield a correct measure of counting efficiency, provided the standard has the same environment as the unknown sample, i.e. the specific activity and chemical composition of the added standard must be such that it produces no significant changes in the nature (volume, phases, etc.) of the sample. If more than one phase is present, it is essential that the added material distribute itself in the same manner as the sample radioactivity. The problem becomes acute for tritium whose radiation is so readily absorbed.³⁹ In an investigation of three-component (toluene, water, triton) emulsion counting of tritium, for example, van der Laarse²⁷ observed counting efficiencies which differed by as much as a factor of three for tritiated toluene versus tritiated water. Similarly, misleading results due to unequal phase distribution of ^{14}C -labelled components in blood have been reported by Laurecot and Hempstead.⁴⁰

Most of the foregoing sources of systematic error may be avoided through combustion if one is concerned with the measurement of ^3H or ^{14}C .^{23,41} Additional benefits include the reduction or elimination of chemiluminescence, and increased counting efficiency. Perhaps the most notable example of these benefits is the radiochemical conversion of the sample — ^3H or ^{14}C — to a scintillation solvent, benzene. Quantitative benzene synthesis has dramatically altered the role of liquid scintillation counting in radiocarbon dating.^{42,43}

(3) **The blank.** So long as sample activity levels are comparable to one another and large compared to the background, errors from contamination and background variations are of little importance. Low-activity samples, however, demand that one

pay attention to numerous additional sources of blank effects and background variations which otherwise would be inconsequential. Vial materials must be selected to yield the lowest possible background.^{44,45} Low-potassium glass or quartz or plastic (polyethylene, nylon) vials seem the most attractive in this respect. To achieve the best background accuracy, Polach³² found it necessary to determine discrete background rates for individual, numbered vials. Sample combustion apparatus can lead to inter-sample contamination due to 'memory' effects;⁴¹ and diffusion and/or evaporative losses have been responsible for sample contamination in radiocarbon dating.⁴⁶ 'Reverse' contamination (decreased sample activity) may also occur in the pretreatment of radiocarbon samples with organic solvents through exchange reactions.⁴⁷ When the lowest levels of activity are of concern, unusual sources of contamination must be considered: water samples which were exposed to the atmosphere during processing, within about 15 km of deuterium-moderated reactors, exhibited tritium contamination of 3 to 6 pCi/litre.⁴⁸

Photoluminescence and chemiluminescence are of special importance in low-level liquid scintillation counting. The former may be minimised by dark-adapting samples for an adequate period of time, and the latter by sample preparation techniques which decrease the likelihood of chemiluminescent reactions. A thorough discussion of the causes and controls for these phenomena is given in Section V of Ref. 16. One caution: chemiluminescence may persist for a period of weeks.⁴⁹ Although luminescence phenomena consist of single photon events, interference occurs even in coincidence counting systems, through pulse pile up and accidental coincidences.

Other potential sources of background error include background variations with time,²⁵ assumed background (baseline) shape under low amplitude α -peaks,⁵⁰ quenching effects³⁹ and interfering nuclear reactions.⁶² The last two deserve some amplification. With respect to quenching, Scales³⁹ has shown that the background for ^3H and ^{14}C measurements also decreases with increasing quenching. The variations with counting efficiency, however, are non-linear, and depend upon the nature of the quenching — i.e. chemical versus colour. An ancillary background effect observed by Scales, in his direct measurements of dissolved tissues, was the variable contribution due to *natural* radioactivity (^{40}K in the tissues). Errors due to an interfering nuclear reaction were encountered by Horrocks⁶² in his measurements of the production of fission-tritium from nuclear power reactors. In order to accurately determine the fission yield of ^3H from ^{235}U , Horrocks found it necessary to reduce the lithium impurity in normal uranium to less than 5 ppm, because of the interfering reaction, $\text{Li}(n, \alpha)^3\text{H}$.

(4) Example — ^{37}Ar . Departing slightly from the literal topic of this symposium, I should like to illustrate quenching and blank problems which we have encountered in the measurement of ^{37}Ar by *gas* proportional counting. In certain respects there are, in fact, close analogies between internal gas and liquid scintillation counting in terms of causes and monitoring of efficiency changes. In both cases, the sample is an integral part of the detector, so its properties can affect the efficiency and the background. Efficiency monitoring may in each case be based upon an internal standard, an external standard or the shape of the sample spectrum.

In our laboratory, it was necessary to measure extremely small concentrations (~ 0.003 dpm/litre Ar) of ^{37}Ar — a 35-day radionuclide which decays by electron capture — in connection with a study of atmospheric reactions of cosmic rays and tropospheric mixing.⁵¹ Long measurement times (\sim weeks) necessitated (quench) corrections for the shifting pulse-height spectra. Shifts in the 2.82 keV full energy

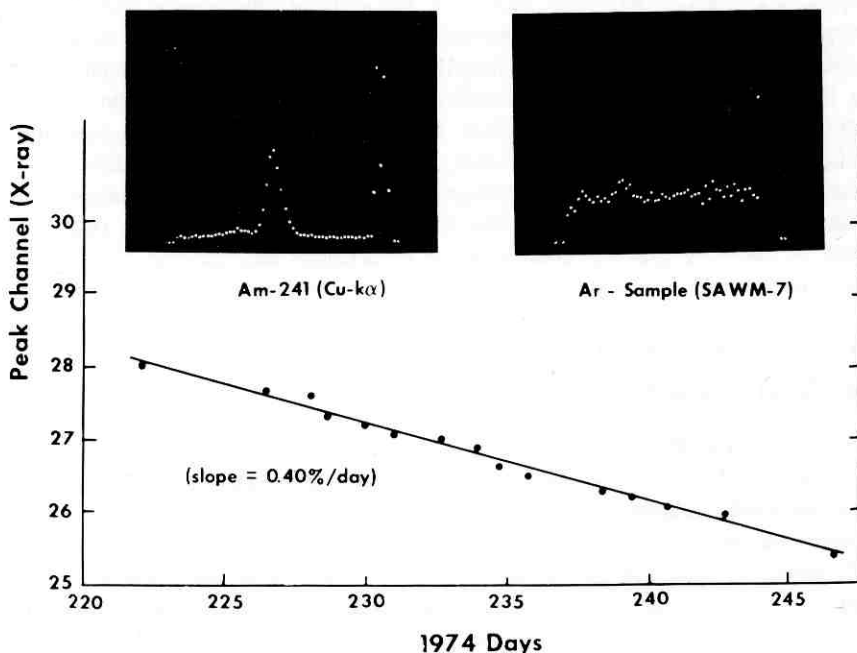


Fig. 3. Quench monitoring of ^{37}Ar . Curve shows drift in gain over 25 days. Counting gas gain and resolution were monitored via an external standard source (^{241}Am , left insert). Argon sample was far too weak to employ the sample for internal gain monitoring (SCR); right insert shows ^{37}Ar peak accumulated in 25 days of counting.

peak, though slow ($\sim 0.4\%/day$) were brought about by composition changes in the counting gas resulting from gradual degassing of internal counter components. Figure 3 shows the quench behaviour using an external standard (^{241}Am). Over the 25-day period covered by the plot, we monitored the peak position of the secondary (Cu-K α) X-ray induced in the brass wall of the gas counter. From the position of the copper X-ray peak, we deduced that of the argon peak and corrected for the quenching accordingly. The two spectra included in the figure represent a 5-min observation of the copper X-ray and a 25-day observation of the ^{37}Ar full energy peak for one of our atmospheric samples. As with SCR-monitoring and low-level liquid scintillation counting, it is obvious that the ^{37}Ar sample peak was far too weak to provide the 14 points on the quenching curve.*

It is instructive to examine the blank effects in the ^{37}Ar experiment, for nearly all possible types were encountered. As is evident from the sample spectrum in Fig. 3, baseline shape assumptions were important; background variations with time and counter were observed; and four distinct kinds of radioactive contamination were encountered. (i) *Radiochemical* contamination (^3H) came from argon isotopic enrich-

*The external source ^{241}Am was used to monitor the position of the ^{37}Ar peak. As with liquid scintillation counting of α -peaks, efficiency was maintained constant by correspondingly shifting the peak integration (energy) 'window'.

ment facilities which were proximate to ^3He production facilities; (ii) *isochemical* contaminants (^{222}Rn , ^{85}Kr) were present in the original atmospheric samples and/or the 'reagents' (charcoal, CaC_2); (iii) *isotopic* contamination (^{39}Ar) also came from the original atmospheric sample, due largely to cosmic ray interactions and influenced by the isotopic enrichment process; finally, (iv) *isotopic* contamination (^{37}Ar) came from atmospheric or underground nuclear tests as well as from neutron reactions with our CaC_2 reagent via the reaction $^{40}\text{Ca}(n, \alpha)^{37}\text{Ar}$. Discussion of steps taken to minimise errors from the foregoing background variations and contamination sources is beyond the scope of this chapter, but the magnitude of the problem may be perceived by the fact that the concentration of artificial atmospheric ^{37}Ar , following a nuclear test, was frequently 100 times the natural concentration.⁵²

ON THE QUESTION OF MERIT*

Conventional measures of performance

Merit and *sensitivity* are two of the most transparent, yet ambiguous, terms currently in use in chemical and radiochemical analysis. Qualitatively, the meanings are obvious: *sensitivity* relates to the ability to detect or quantify, and *merit* is a measure of relative sensitivity.

Both terms, in turn, are tied implicitly to the structure of the measurement process and to its component errors. Explicit definitions, however, depend upon the user. In spectrochemical analysis, for example, *sensitivity* is defined as the slope of the calibration function, whereas for nuclear measurements (liquid scintillation counting, activation analysis, etc.) the same term is frequently used to specify a minimum detectable quantity.^{45,53} *Merit*, likewise, takes on numerous meanings when defined by different workers; the significance of some of the more popular 'figures of merit' and their relationship to the underlying errors and assumptions will be examined below.

Despite the ambiguities, the central aim remains clear: we wish to provide an objective and quantitative means for assessing the capacity of a given procedure to detect or to measure (with a given relative standard deviation) the radionuclide (or element or compound) of interest. We wish also a quantitative measure of the relative capabilities of alternative procedures or that of a given procedure as a function of controllable variables, such as sample volume or discriminator level. These ends will be met in an easy-to-apply, yet objective, graphical procedure which may be used for the practical *planning* of radiochemical measurements and for the selection of optimal procedures or optimal values for experimental variables. Sources of error, as discussed in the preceding section, together with Poisson counting statistics, will provide the basis for the evaluation of merit in liquid scintillation counting.

To begin the evaluation of counting methods, it will be instructive to examine some of the expressions commonly used to denote merit. One finds, in perusing the liquid scintillation and radiochemical literature, the following terms (among others): Figure (or Factor) of Merit (F),^{13,41,54-56} Merit Number,²⁷ Merit Value and Merit Stability Quotient.²⁶ The term Figure of Merit, *per se*, is equated to a bewildering array of expressions ranging from S^2/B to $\sqrt{\epsilon V}$ (S = sample counts or rate, B = background counts

*Definitions for symbols used in this section will be found in Table 1.

or rate, ϵ = counting efficiency and V = sample volume). To these is added the Background Equivalent Activity ($BEA = B/\epsilon$ or $B/[\epsilon V]$) which is equivalent to the 'absolute' or 'concentration sensitivity' as used by Gelsema *et al.*⁵³ With the exception of the Merit Stability Quotient (MISQ), the expressions used to deduce the relative merit of alternative radiochemical measurement systems are based on the premise that random errors arise from Poisson counting statistics only, and that systematic errors are negligible. As has already been seen, such (non-Poisson) errors cannot always be ignored. Their importance in terms of colloid stability has been recognised and incorporated into MISQ by Fox,²⁶ and their significance in radiocarbon dating was treated by Currie⁵⁷ by resolving F into Poisson and non-Poisson components. The use of BEA as a measure of merit is predicated on the assumption that signals much smaller than the background may be unreliable, due to background variability arising from external sources. Though BEA is a helpful guide, it is in fact essential to assess the magnitude and variability of the blank if reliable conclusions are to be drawn.

The variety of expressions for merit, based upon counting statistics alone, obtains from three sources: (i) whether total activity or specific activity (per unit mass or volume) is of interest; (ii) whether the relative (inverse) variance or standard deviation is considered (e.g. S^2/B versus S/\sqrt{B}); and (iii) whether $S \ll B$ or $S \gg B$ is assumed. The first two assumptions should cause little difficulty in any particular case, provided the conventions in use are made clear. The last assumption, however, may lead to some peculiar difficulties, especially from the arbitrary application of the most commonly used Figure of Merit, ϵ^2/B . ϵ^2/B (or $[\epsilon V]^2/B$), of course, derives from one of the asymptotes of the inverse (relative) Poisson variance:

$$F = \frac{1}{\phi^2} = \frac{S^2}{S + 2B} \begin{cases} \propto \epsilon^2/B & (S \ll B) \\ \propto \epsilon & (S \gg B) \end{cases} \quad (1)$$

Apparent paradoxes which arise from ϵ^2/B (or ϵ/\sqrt{B}) are seen, for example, in reviews by Calf⁵⁸ and Cameron.⁵⁹ In the former case, direct counting of tritium as water (in dioxane) was shown to have a higher figure of merit (0.6 versus 0.4) than the benzene synthesis method, yet the benzene method was capable of quantitatively measuring a slightly smaller amount of tritium (300 TU versus 310 TU) than the water-dioxane method (in 500 min). The same situation occurs in Table III of Cameron's review for low-background gas (GM) tritium counters: #13 (Sweden) versus #15 (UKAEA). Here, one finds also the phenomenon of 'reversal' — where one method (counter) is superior at one level of activity (#13, at 500 TU), and the other is superior at a different level of activity (#15, at 5000 TU). (Determination of the activity level at which reversal takes place is one of the important questions which will be discussed later in this section.)

A closely related problem arises when, in the comparison of two methods, $S < B$ for one, while $S > B$ for the other. Then, clearly, neither of the limiting expressions yields a valid measure of relative merit. One solution which has frequently been taken is to calculate absolute or relative merit directly from Eqn. (1), by assuming a given level of activity, hence a given value for S . Such an approach is not entirely satisfactory, however, for it is not clear what particular activity level ought to be pre-selected for the computation. In the following sub-section we shall investigate an approach which is independent of the foregoing constraints and which can be applied singly or simultaneously to a collection of alternative procedures.

Table 1. Symbols and relationships.

t	= counting times
R_B	= background rate, B = background counts = $R_B t$
R_S	= net (sample) rate, S = sample counts = $R_S t$
ϵ	= measurement efficiency (calibration factor) ^a
A	= activity = R_S/ϵ
BEA	= background equivalent activity = R_B/ϵ
ρ	= reduced activity = $S/B = A/BEA$
ϕ	= relative standard deviation = σ_A/A
F	= merit = $1/\phi^2$

Subscripts: P, B, ϵ = variance or merit component due to Poisson, background or calibration errors, respectively.

^a ϵ represents the overall calibration factor, including both counting efficiency and chemical recovery; when specific activity is of concern, ϵ *must* incorporate also sample mass (m) or volume (V).

A consistent approach to merit

(1) **Reduced activity.** The primary aim is to develop a quantitative measure of performance which requires no prior assumption of the relative magnitude of S and B or of activity level. One such measure is the detection limit (A_D), given the counting time (t); another is the quantitative (or determination) limit (A_Q). Expressions for A_D and A_Q ⁶⁰ are as follows:

$$A_D = \frac{1}{\epsilon t} (2.71 + 4.65 \sqrt{B}) \quad (2)$$

$$A_Q = \frac{50}{\epsilon t} (1 + [1 + B/12.5]^{1/2}) \quad (3)$$

(A_D assumes 5% risks – errors of the first and second kinds – and paired observations of sample and background; A_Q assumes 10% relative standard deviation for quantitation.) Once a procedure has been specified – i.e. R_B , ϵ fixed – A_D, A_Q are functions of counting time only. One such function, $A_Q(t)$ for ³²P measured by liquid scintillation counting, is shown in Fig. 4.

Equations (2) and (3) and Fig. 4 are unsatisfactory in that separate computations or curves are required for each procedure of interest. The deficiency may be overcome, however, by the substitution of dimensionless ('reduced') coordinates, in which the activity level (ordinate) is normalised with the BEA , and the time (abscissa) with the mean interval between background counts ($1/R_B$). A universal curve results which permits us to immediately deduce for any procedure the 'reduced' quantitative activity ($\rho_Q = A_Q/BEA$) as a function of the 'reduced' time ($\tau = t/R_B = tR_B = B$). The folly of assuming a fixed expression (e.g. ϵ^2/B) to compute a single, numerical value for relative or absolute merit is implicit in the changing slope of the curve (Fig. 5). Merit must be viewed as a two-dimensional (or two-compo-

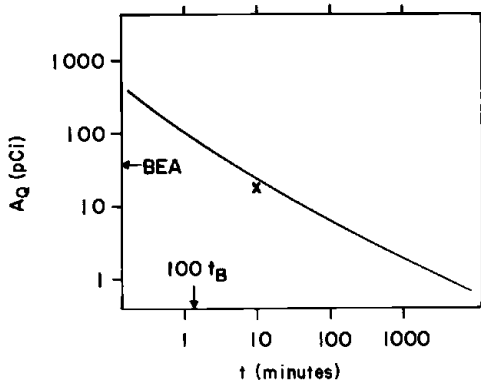


Fig. 4. Quantitation limit (A_Q , pCi) for liquid scintillation counting (L) of ^{32}P versus counting time (t). t_B equals the mean interval between background pulses ($1/R_B$); BEA equals the Background Equivalent Activity (40 pCi). Curve shows, for example, that for $t = 10$ min, $A_Q = 22.7$ pCi. Point (x) – 20 pCi – has poorer precision ($\sim 11\%$) as it lies below the curve. Assumed background and efficiency data are given in Table 2 (Method L).

ment) quantity in counting experiments; only at the asymptotes ($B \rightarrow 0$, slope $\rightarrow -1$, or $B \rightarrow \infty$, slope $\rightarrow -1/2$) does a single value suffice. An added benefit of the dimensionless representation (Fig. 5) is explicit indication of the point at which $S = B$.

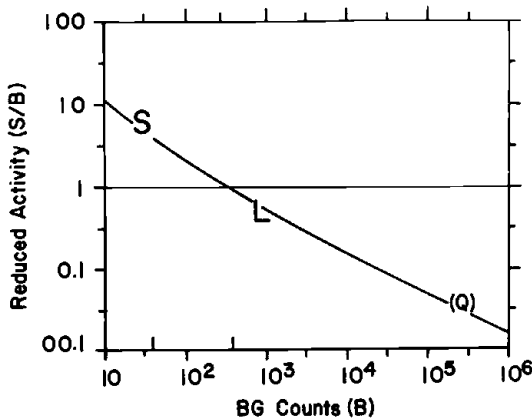


Fig. 5. Reduced activity plot, ρ_Q versus B . The curve is universal, but the points characterise the performance of liquid scintillation (L) and $2\pi\beta$ solid (S) counting methods for ^{32}P when the activity equals 20 pCi and the counting time 10 min. Note that $S > B$ for S, but $S < B$ for L. (Input data from Table 2.)

We see that $\rho_Q > 1$ ($S_Q > B$) if $B < 300$ counts, and *vice versa*. (For the detection limit, $\rho_D = 1$ at $B = 27$ counts.) These crossing points (300 counts, 27 counts) are helpful in judging the adequacy of either of the asymptotic expressions for merit, given the counting time.

Figure 5 may be used also to evaluate the relative performance of alternative

methods. Points 'S' and 'L' refer to solid (external, $2\pi\beta$) and liquid scintillation counting of ^{32}P , using counting efficiencies and background rates as given in Table 2. The points have been normalised to 20 pCi and 10 min. (Thus, the coordinates at L are $y = 20 \text{ pCi}/\text{BEA} = 20 \text{ pCi}/40 \text{ pCi} = 0.50$, and $x = (80 \text{ counts min}^{-1}) (10 \text{ min}) = 800 \text{ counts}$.) Comparing the positions of the points (L,S) with the curve, it is evident that S has the greater merit, for this particular choice of activity and counting time. S, lying slightly above the curve, has better than 'quantitative' precision ($\phi_S = 9.4\%$), whereas L, which lies below the curve, has poorer precision ($\phi_L = 11.2\%$). Levels for which each of the two methods would be quantitative ($\phi = 10\%$) derive directly from the curve ($t = 10 \text{ min}$):

$$A_Q = \rho_Q \cdot \text{BEA} = \begin{cases} (0.566) (40.0) = 22.7 \text{ pCi (for L)} \\ (4.74) (3.86) = 18.3 \text{ pCi (for S)} \end{cases}$$

The effects of changes in activity levels or counting times correspond simply to translations of the points (L,S) along the y- or x-axes, respectively. Note that for the 10 min counting time selected $\rho(S) > 1$ whilst $\rho(L) < 1$ — i.e. the net signal exceeds the background for the one procedure at the same time that the background exceeds the net signal for the other. This example should serve to illustrate (i) the utility of the dimensionless representation, as well as (ii) the potential pitfall in generally applying either FOM asymptote when comparing alternative procedures.

(2) Higher dimensions; non-Poisson errors. In order to provide a general treatment to the question of measurement performance, it has been necessary to increase the dimensionality from one (scalar, asymptotic- F) to two (ρ, B). Already implicit in the discussion — through the introduction of A_D and A_Q — is a third dimension, related to the experimental precision (ϕ). A contour plot is given in Fig. 6a, for the evaluation of performance at various levels of precision. (Curves C, D and Q represent the critical (decision) level, detection limit and quantitation limit, respectively, as defined by Currie.⁶⁰

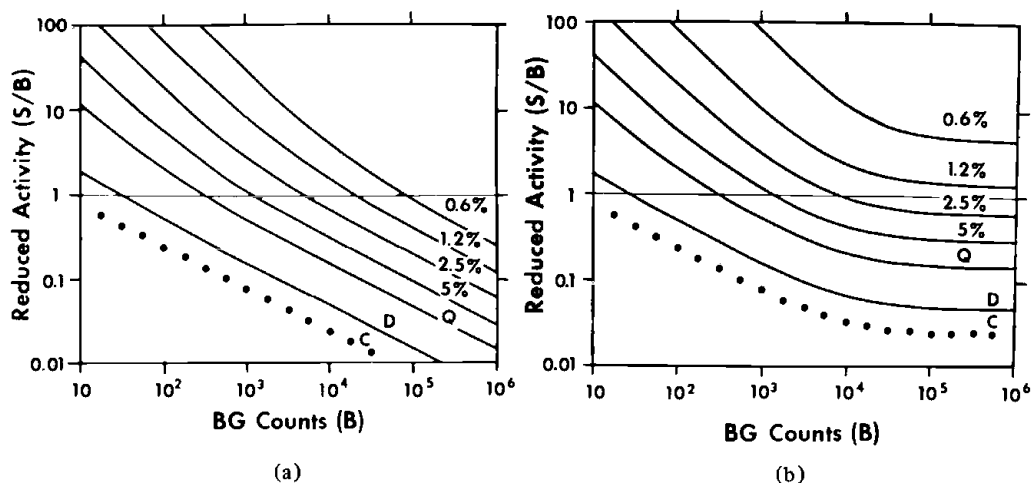


Fig. 6. Reduced activity curves. Contour plots are presented for reduced activity ($S/B = \rho$) versus background counts (B) and counting precision (ϕ). Part (a) includes Poisson errors only; part (b) incorporates additional random error (0.50% for counting efficiency, 1.0% for background variability).

Two additional parameters, ϕ_B and ϕ_e , have not yet been taken into account. These represent non-Poisson random errors in the background and calibration factor, which cannot be ignored in any real experimental situation. As four- or five-dimensional plots (even contour plots!) are quite beyond the capability of this author, a particular 'cut' of the five-dimensional figure has been selected, as shown in Fig. 6b. The values selected for ϕ_B and ϕ_e (1% and 0.5%) are, in fact, identical to those used in Fig. 1 of Currie.⁵⁷ These values were believed to be representative of about the best obtained in routine radiocarbon dating. As such, they may serve as a useful guide for a limiting merit diagram for real counting experiments, just as Fig. 6a serves as a limiting diagram for ideal counting experiments (Poisson errors only). If the *overall* performance of any specific counting experiment is to be evaluated, the corresponding values for ϕ_B and ϕ_e must be determined. These may then be incorporated into a figure similar to Fig. 6b using Eqn. (4), which follows from the definition of reduced activity (ρ) and error propagation:

$$\phi^2 = \left[\frac{1}{B} \left(\frac{\rho + 2}{\rho^2} \right) \right] + 2 \left(\frac{\phi_B}{\rho} \right)^2 + \phi_e^2$$

where the first term (in parentheses) refers to the Poisson component (contours in Fig. 6a).

The most important point to be noted in Eqn. (4) is the limiting effect of ϕ_B and ϕ_e . i.e. as counting time increases ($B = R_B t$ increases) the Poisson component approaches zero. The overall relative variance (ϕ^2) is then limited by the last two terms. The relative standard deviation of the calibration factor (ϕ_e), which depends upon random errors in efficiency (quenching and quench-corrections), isotopic enrichment, chemical recovery, etc., sets a fixed limit for the best achievable precision. The contribution from the relative standard deviation for the blank (ϕ_B), however, varies with the reduced activity level ($\rho = S/B = A/BEA$), being negligible for $S \gg B$ but becoming all-important for signals small compared to the blank ($\rho \ll 1$). For example, if one were attempting to measure quantitatively a sample whose activity was 10% of the *BEA* ($\rho = 0.1$), non-Poisson contributions would be significant unless $\phi_e < 10\%$ and $\phi_B < 0.7\%$ — the latter limit (ϕ_B) being given by the constraint that the second term in Eqn. (4) be $< (10\%)^2$. That such bounds on ϕ_B and ϕ_e are not easy to achieve was evident in the discussion of liquid scintillation errors in the preceding section.

(3) **A general definition and asymptotic relations.** In keeping with the original intent of the Figure of Merit in counting experiments we may conclude that procedures having equal precision possess equal merit. The contours in Fig. 6 are therefore equimerit contours. The index, F , when treated like a statistical weight, may be set equal to the reciprocal variance ($1/\phi^2$). F , for a given procedure, or relative F 's for alternative procedures, therefore require specification of B , ρ , ϕ_B and ϕ_e (Eqn. 4). Changing activity levels ($\rho = S/B$) or changing counting times ($B = R_B t$) will generally alter *relative* as well as absolute F 's.

Two important conclusions derive from the geometry of the points and contours in Fig. 6: (i) the relative positions of points representing different procedures are invariant with time or activity level – they are fixed by background rate and background equivalent activity (or counting efficiency) alone; (ii) the critical angle (slope) for points of equal overall merit (cf. Fig. 6b) varies continuously from -45° (slope = -1) to -0° . The first conclusion yields an extremely convenient device for evaluating relative merit and experiment adequacy, which will be utilised subsequently. The second conclusion yields limiting expressions for equal merit, as shown in Fig. 7.

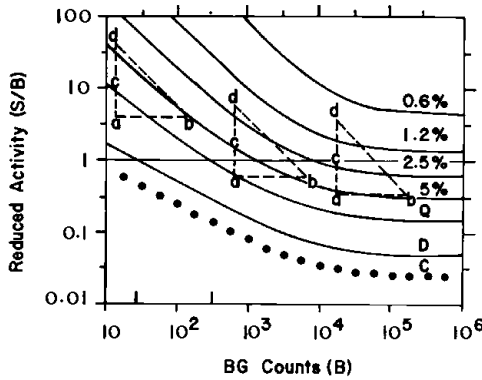


Fig. 7. Merit. Construct a-b-c-d, superposed in three different locations on Fig. 6b, shows changing relations (comparative merit) among procedures. (b,d) have equal counting efficiency, ϵ ; (b,c), equal ϵ^2/B ; and (a,b), equal background equivalent activity (or B/ϵ).

The construct, a-b-c-d, in Fig. 7 is taken to represent four alternative counting procedures, the relative positions of which are fixed by the respective R_B 's and BEA 's. Three locations, representing translations in y (activity level) and x (time), have been given for the construct. Changing relations among the relative merit (F) or precision thus become clear:

Location-1 (upper left):	$F_b \approx F_d > F_c > F_a$
Location-2 (intermediate):	$F_d > F_b = F_c > F_a$
Location-3 (lower right):	$F_d > F_c > F_b \approx F_a$

The transition from small B and large ρ is thus accompanied by a gradual change in the limiting expression for equal merit from: (i) $\epsilon = \text{const.}$ (b,d), slope = -1; to (ii) $\epsilon^2/B = \text{const.}$ (b,c), slope = -1/2; to (iii) $\epsilon/B = 1/BEA = \text{const.}$ (b,a) slope = -0. (If ϕ_B and ϕ_ϵ were always negligible, the contours would have limiting slopes of -1 and -1/2, as in Fig. 6a.) The inequalities are worth noting: for two procedures of equal efficiency (b,d), the one having the smaller background (d) is generally preferred, because $F_d \geq F_b$; for two procedures of equal ϵ^2/B (b,c), the one having the larger background is preferred, because $F_b \geq F_c$ provided non-Poisson errors are negligible.

The practical evaluation of counting experiments

(1) ³²P: merit of alternative methods of measurement. The foregoing approach to merit and the resulting contour plots can be readily adapted for very rapid planning and comparative evaluation of counting procedures. This may be accomplished by using the basic contour plot (Fig. 6), together with an array of points representing the invariant *relative* positions of the procedures in question, and x-, y- or z- (contour) translations of the entire array. (The array may be constructed simply by plotting $y = 1/BEA$ versus $x = R_B$ for each of the procedures in question.)

The investigation of merit in ³²P counting has been carried further in Fig. 8, where two *independent* arrays of points are given for (i) activity (pCi – methods S, C, L), and (ii) specific activity (pCi/ml – methods © and ⊙). The first set has been normalised to 20 pCi, the second to 20 pCi/ml; both sets have $t = 10$ min. Assumed input data are given in Table 2. (Note that the background and

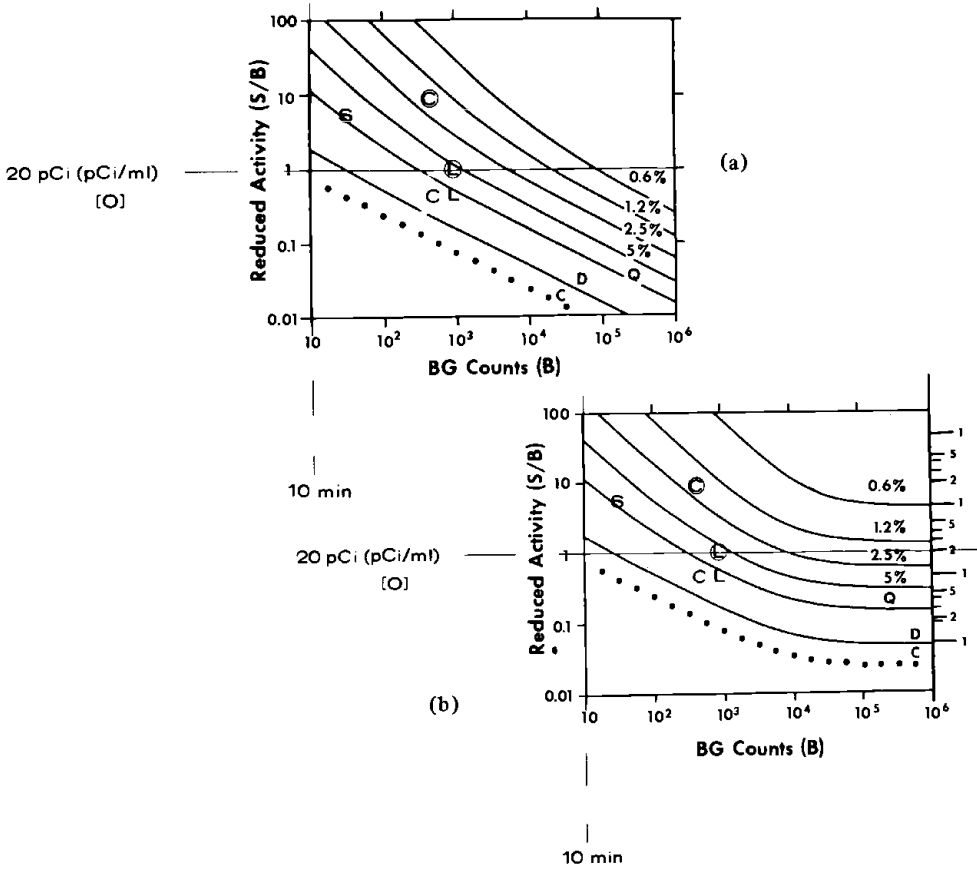


Fig. 8. Comparative performance of solid, liquid scintillation and Cerenkov counting of ³²P. Circled points refer to specific activity (pCi/ml), others to total activity (pCi). Normalisation set at 20 pCi (or pCi/ml) (y-axis) and 10 min (x-axis); input data from Table 2. Part (a) uses contours from Fig. 6a (Poisson errors only); part (b) uses contours from Fig. 6b. (Scale at left is reduced activity; scale at right, pCi. Abscissa in this and following figures indicates background counts as well as counting time (min).)

Table 2. ^{32}P Merit: input data.

	Counting method ^a				
	L	C	S	⊙	⊚
Counting efficiency (%)	90	40	35	90	40
Background rate (counts min ⁻¹)	80	40	3.0	80	40
Sample volume (ml)	-	-	-	2	20
BEA (pCi)	40.0	45.0	3.86	-	-
BEA (pCi/ml)	-	-	-	20.0	2.25

^aL, C, S = liquid scintillation, Cerenkov and solid ($2\pi\beta$) counting for total activity (pCi); ⊙, ⊚ = liquid scintillation, Cerenkov counting for specific activity (pCi/ml).

efficiencies used are assumed, plausible values, used here strictly for the sake of illustration.) For total activity measurements (S, C, L), we have already noted the somewhat better performance of S compared to L (Fig. 5 discussion). From Figure 8a it is apparent that Cerenkov counting (C) yields slightly poorer performance than either of the other methods.* On the other hand, upon examination of the points denoting specific activity, ⊙, ⊚, we find that Cerenkov counting is considerably better than liquid scintillation counting; $\phi(\odot) \sim 2\%$, $\phi(\ominus) \sim 5\%$. We see also from the figure that $\rho(\odot) \approx 1$, whereas $\rho(\ominus) \approx 10$. Therefore the normalisation activity (20 pCi/ml) equals the background equivalent activity for ⊙, so neither asymptotic expression for F would be appropriate. For ⊙, $S \gg B$, so the asymptotic form, $F \propto \epsilon V$, would be acceptable.

Figure 8b differs from Fig. 8a (i) by the addition of an activity scale at the right (set at 20 pCi or pCi/ml for $\rho = 1$), and (ii) through the use of contours incorporating the effects of non-counting errors, like Fig. 6b. Performance as deduced from Fig. 8b is scarcely different from that deduced from Fig. 8a because of the relatively small numbers of counts — i.e. Poisson errors are still limiting. The effects of large increases in counting time, however, are very different. Figure 8b shows, for example, that the limiting precision ($t \rightarrow \infty$) for both Cerenkov (C) and liquid scintillation counting (L) of 20 pCi of ^{32}P is $\sim 4\%$ due to the assumed non-counting errors.

The reversal of relative merit with activity level is illustrated in Fig. 9 which corresponds to a one decade vertical translation of the array of points to 200 pCi (still 10 min). Now, the precision of L is somewhat better than that of S, though the precision of both is improved in comparison with Fig. 8b ($\sim 2\text{--}3\%$ rather than $\sim 10\%$). Obviously, therefore, there must exist an intermediate activity level for which L and S have equal merit. The magnitude of that intermediate level may be found, by vertical translation, to be about 40 pCi, at which point both procedures have a precision of 6%.

Actually, a continuous series of points of equal merit (for L and S) exists. It is characterised by a functional relationship involving counting time (t), activity level (A) and precision (ϕ), such that all three parameters are the same for the two procedures (L,S). Equal

*Note that curve D (detection limit) corresponds to a relative standard deviation of about 30%.

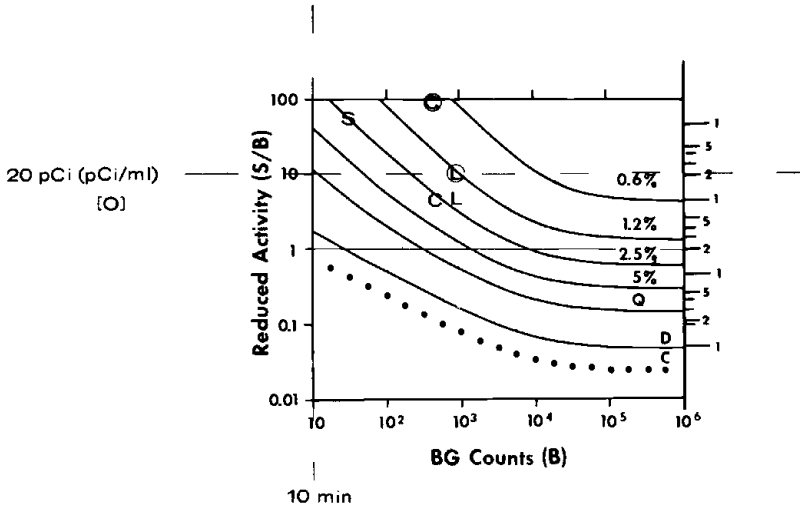


Fig. 9. Comparative performance, ³²P. Array of Fig. 8b translated one decade vertically to new normalisation of 200 pCi (or pCi/ml). Reversal of merit is exhibited by points L and S: Fig. 8b, $\phi_S < \phi_L$; Fig. 9, $\phi_S > \phi_L$ (ϕ = relative standard deviation).

merit values of t, A, ϕ may be derived from translations along the x-axis (t) or along the y-axis (A) or along a contour. The result of such an analysis is given in Fig. 10. $2\pi\beta$ (S) counting has greater merit than liquid scintillation counting (L) for all samples whose activity is less than about 35 pCi. Liquid scintillation counting is better for large activities, but the point of (merit) reversal increases gradually with counting time, having approximately doubled by the time that $t = 100$ min.

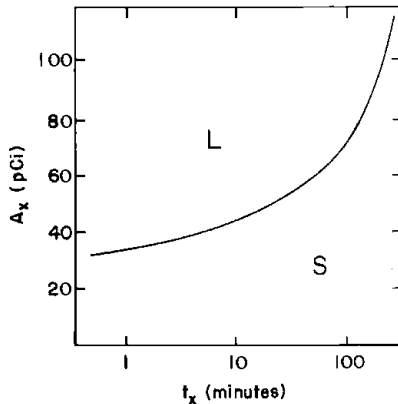


Fig. 10. Equimerit curve for ³²P (L, S). Curve for which the counting precision is identical using liquid scintillation (L) or solid, $2\pi\beta$ (S) methods. 'Crossing' activity level (A_x) versus time (t_x) may be used for selecting preferred method depending upon activity level or counting time. Liquid scintillation counting (L) is better for space above the curve; S is better for the space below.

Table 3. Tritium merit: input data.

	Counting method ^a				
	G	a	A	b	B
Counting efficiency (%)	90	34	53	34	53
Background rate (counts min ⁻¹)	0.20	0.89	1.72	1.68	2.98
Sample volume (ml)	-	10	10	50	50
Water equivalent (g)	1.61	6.87	6.87	33.4	33.4
BEA (pCi/kg) ^b	62.2	172	213	66.6	75.8

^a G = gas proportional counter (NBS counter, approximate characteristics), a, A, b, B = liquid scintillation counter,⁴⁵ with varying volumes and efficiencies (from discriminator changes) as shown in table.

^b 1 TU (Tritium Unit) = 3.24 pCi/kg H₂O.

though G, a and A possess equal merit, the gas counter requires only about 25% as much sample as the (10 ml) liquid scintillation counter; (ii) use of $(\epsilon V)^2/B$ as a measure of merit would be incorrect, for the signal exceeds the background ($\rho > 1$) for all five of the methods.

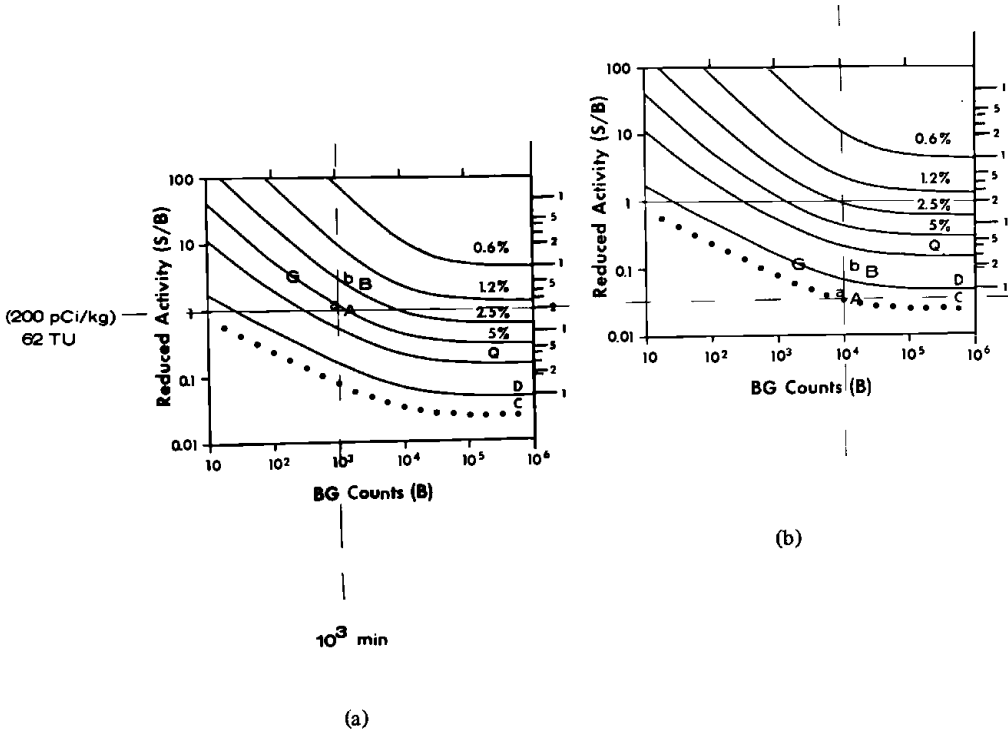


Fig. 12. Comparative merit for tritium methods. Comparison of gas counting (G) and liquid scintillation counting (a, A, b, B) of tritium. Liquid scintillation counting is illustrated for four different efficiency-sample volume choices. (See Table 3 for symbol identification.) Part (a) is normalised to 200 pCi/kg H₂O (62 TU) and 1000 min; part (b) exhibits the lowest feasible detection limit (for method G, 1 week of counting). Note the change in relative merit and optimum variable (discriminator, volume) combination.

A second comparison of interest treats the case where the gas counter is pushed to its limit. In Fig. 12b we evaluate the smallest detectable tritium concentration (counter G) when the maximum feasible counting time (10^4 min \sim 1 week) is employed. This concentration is seen to be about 7 pCi/kg (\sim 2 TU), where the signal is just slightly greater than one-tenth of the background. G is no longer equivalent to a and A, however. In fact, it is now intermediate between a, A and b, B in performance; a, A lie below the detection limit, and b, B may be measured with a precision of about 20%. A final observation: the relative merit of b, B is reversed. The larger volume liquid scintillation samples still provide the best merit, but now the small efficiency member (b) has a slight edge. $(\epsilon V)^2/B$ would be appropriate for ranking the five methods based upon Poisson errors alone, since $\rho \ll 1$. However, B (background counts) is so large, especially for the liquid scintillation measurements, that the effects of background reproducibility must also be considered.⁶¹

CONCLUSION

The utility and general meaning of Figure of Merit (F) are clear. F is obviously extremely useful for procedure selection and variable optimisation, and it must surely be defined as a measure of signal/noise or relative precision. Difficulties arise, however, once numerical values are assigned to F – even for the assessment of *relative* merit – because of equivocal or improperly used asymptotic expressions.

Taking F to be $1/\phi^2$ means that variance components translate directly into F components (eqn. 4), and that F must be viewed as a function of A , ϵ , R_B , t , ϕ_B and ϕ_E . Transformation to reduced coordinates (ρ, B), however, permits F_p (Poisson component) to be represented by a single diagram (Fig. 6a). This, in turn, permits the merit of any number of alternative procedures to be simultaneously evaluated. Figures 6a and b and Eqns. (3) and (4) yield the following caveats:

- (i) A_Q (given t) or t_Q (given A) may be used as a single index of performance. No prior assumption of activity level or S/B is required.
- (ii) Validity of alternative, asymptotic expressions for F relate to contour slopes:
 - 45° implies $F \propto \epsilon$
 - 30° implies $F \propto \epsilon^2/B$
 - 0° implies $F \propto (\epsilon/B)^2 \propto (1/BEA)^2$
- (iii) The approach to zero slope establishes maximum useful counting times and minimum useful ρ 's (S/B).
- (iv) No single asymptotic measure of merit (ϵ , ϵ^2/B , $1/BEA$) may be generally relied upon for comparing alternative procedures, for different procedures may simultaneously be in different asymptotic regions (cf. Fig. 5).
- (v) Unless two procedures have identical BEA 's a range of A 's will exist for which $S_1 > B_1$ at the same time that $S_2 < B_2$.
- (vi) If the slope of the line connecting points representing two procedures lies between -45° and 0°, there will exist a range of A 's and t 's over which merit reversal will take place (cf. Figs. 7-10).

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