

## Chapter 27

# Experience in Off-Line Computer Processing of Liquid Scintillation Counting Data

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### INTRODUCTION

Liquid scintillation counting has developed rapidly since the work of Kallmann and Furst,<sup>1</sup> so that now, with spectrometers fitted with automatic sample changers, it is a technique that can give rise to a large mass of numerical results; it is therefore an obvious field for the application of electronic computers. Many workers have found it worthwhile to apply a computer to liquid scintillation counting (either 'on-line',<sup>2</sup> with a 'desk-top' computer<sup>3</sup> or with a large 'off-line' computer). Often the computer is used in a data processing role to make very simple calculations quickly and reliably on the results of counting large numbers of samples, but sometimes also to carry out more complex calculations.

In this chapter I shall concentrate on the work done at Thornton Research Centre, having chosen this approach deliberately, not with any intention of belittling the work of others active in this field, but because it may be of interest if I describe how this application of computers has developed in one laboratory. The equipment and type of computer program will be considered, and examples will be given of the use of a computer in determining the stability of instruments and in fitting curves to experimental data. Finally, I shall mention briefly some relevant published work and try to define some of the future needs of radiochemists.

The earliest applications of liquid scintillation counting at Thornton were carried out with a home made apparatus containing a single photomultiplier tube. The increasing numbers of samples involved in a wide variety of radiotracer experiments led to the installation in 1961 of a Packard TriCarb automatic liquid scintillation spectrometer (Model 314 EX), to which was added a punched paper tape output. This instrument has been in use for nearly ten years, but was supplemented in 1969 by a more modern counter, a Packard TriCarb Model 3320, which is a three-channel, 200-sample instrument with external standard and teletype output. Paper tape has been found to be an entirely satisfactory medium for the collection of data from these instruments for off-line processing, as we are dealing with low rates of collection of data (of the order of fifty characters after each counting period of, say, 10 min).

It was decided to use off-line computing of results because a Ferranti Pegasus computer had been installed at Thornton for other purposes. Since 1965, links to larger central

computers have been used, first to a Univac 1107 run by a computer service bureau in Birmingham and more recently to a Univac 1108 at Shell Centre in London. The programs for radioactivity counting experiments have been written in FORTRAN (FORmula TRANslation) computer language. The use of such a widely recognized programming language has the advantage that the programs will be readily applicable to other types of computer.

## **THE COMPUTER PROGRAM**

The general form of the program is dictated by the fact that we have at first to carry out many standard operations (of taking in counting data in a standard form) but that, after this stage, we must make different calculations depending on the nature of the experiment (for instance, whether it involves internal standard, external standard, or channels ratio methods of determining counting efficiency). The appropriate form of program for this is a general routine to be used in each application, together with a collection of optional subroutines appropriate to particular experiments. The various stages of the program will now be considered, beginning with the form of the counting data, which of course depends on the type of scintillation counter/interface/output device used. For simplicity the following sections have been confined to the system Packard TriCarb 3320/Teletype ASR 33.

Data are produced in a standard form as sample number (three digits), time (six characters, including a decimal point) and three six-digit numbers for the counts in each of three channels, which may be followed by three further six-digit numbers if the external standard is used. These data are punched automatically on paper tape in the teletype, groups of digits being separated by spaces and the printout from each sample being terminated by separating symbols ('carriage return' and 'line feeds'). Eight-hole paper tape is used in which each digit is represented by two, four or six punched holes, so that a punching error causing the addition or omission of one hole produces a major error, rather than the substitution of one digit for another (parity check). This is the so-called ASCII (American Standard Code for Information Interchange).

### **Parameter data**

Additional information is fed into the computer to identify the counting data, to specify the calculations required, to define the settings used on the counter, and to set limits of precision. This information can be punched manually on two data cards, which precede the count data, or as a heading on the paper tape on which the counts are to be recorded. It consists of the following:

1. Date of counting.
2. Counter used.  
(These two parameters have been found to be sufficient to identify the counting data, which may have been produced from instruments other than a liquid scintillation counter, e.g. a  $\gamma$ -ray scintillation counter or Geiger counter).
3. Subroutines to be used for processing the data.  
(Up to six different subroutines can be used with one set of data).
4. Number of samples counted.
5. Number of cycles.  
(Each cycle consists of one set of counts on each sample).
6. Time for sample counts.  
(This may be in seconds, in which case a typical printout from the 3320 would be

- 10.0, or in minutes, which would be printed as 10.00).
7. Preset counts.
8. Use (digit 1) or absence (digit 0) of external standard.  
(Parameters (4) to (8) provide all the information required on the settings of the counter).
9. Limits of precision for counts from samples.
10. Limits of precision for counts from the external standard.  
(These limits are required for tests, which will be described later, on the differences between statistical and experimental standard deviations of the counts).
- 11-13. Digits 1 or 0 to signify whether the external standard counts in each of the three channels exceeds  $10^6$ .

### **Form of computer programs**

FORTTRAN computer programs have been written for use with any counter that can produce data in the form described above (or in a similar form such as that produced on five-hole paper tape by counters similar to the Packard 314). These programs have been found useful with a  $\gamma$ -ray spectrometer and with a Geiger counter, as well as with a liquid scintillation counter, since the same basic calculations are required for data from any of these radioactivity detectors. The program contains a short *main program*, which reads and stores data and organizes the 'calling' of required subroutines. This is followed by *compulsory subroutines*, which are used to interpret every set of data and to carry out basic calculations. A number of *optional subroutines* (fifteen have been written to date) are used to carry out further calculations on any particular set of data and to print out results in the required form.

In FORTRAN, data and results can easily be transferred from one program or subroutine to another. By using this form of program, it has been found possible for individual users to write their own subroutines, which can be added to the existing program and can use the results of calculations carried out in other subroutines. For a particular project a subroutine can be written in such a way that results are printed out in the most suitable form.

The program and subroutines are stored on magnetic tape or on a 'drum', ready for reading into a computer. About 32 000 36-bit words of core storage are used for program and subroutines together with the data (for which storage is 16 000 six-digit numbers).

### **Main program**

The main FORTRAN program, RADIOC, is short and is concerned with the storage of data and organization of subroutines; no calculations are included in this program. The following functions are performed:

1. Computing starts when a call is made to 'execute' the main program.
2. Data are read, stored and made available for use in subroutines.
3. Compulsory subroutines are 'called'.
4. Optional subroutines are 'called', as listed on parameter cards.
5. Computing ends.

### **Compulsory subroutines**

The compulsory subroutines are used with all sets of counting data, which must be in a standard form as described above. Information from the parameter cards and from the

liquid scintillation counter are read into the computer; the counts and times are stored in a three-dimensional array, in which the first index specifies the counting channel (counting time being included as a channel), the second is the sample number, and the third is the number of the cycle. The information in this array is made available for use in any subroutine. The mean count rates in each of the three count channels are calculated for each sample and stored in a two-dimensional array, in which the first index specifies the channel and the second is the sample number.

The standard deviations of the mean count rates are calculated, expressed as a percentage. One value (SDS) of each standard deviation is found from the statistics of radioactive disintegrations. This standard deviation is found from the square root of the total counts, and is stored in a two-dimensional array. A second value (SDE) of each standard deviation is calculated when a sample has been counted more than once. This standard deviation is found from the differences from the mean of each individual count rate, and is again stored in a two-dimensional array. Finally, a test is made to determine whether or not there is a significant difference between the values of SDS and SDE. For this purpose the F-test<sup>4</sup> is used at 99% significance.

The ratio of the standard deviations is recorded as significant if  $(SDE/SDS)^2$  exceeds the critical F value, and if SDE exceeds the fixed limit as given in the data on parameter cards. The results of these tests are stored in a fourth two-dimensional matrix.

### **Optional subroutines**

The subroutines available for use with the main program RADIOC include the following:

1. Printout of edited data.
2. Printout of mean count rates and standard deviations.
3. (Used for  $\gamma$ -counting).
- 4.&5. Net count rates per unit weight.
6. Water transfer rates (permeability experiments).<sup>5</sup>
7. Count rates corrected for radioactive decay.
- 8.&9. Channels ratio (Baillie's method).<sup>6</sup>
10. Channels ratio (parabolic curve fitting).<sup>7</sup>
- 11.&12. Net count rates  $\times$  factor/count rate from standard sample.
13. Disintegration rates (internal standard method).
14. Oil consumption rates (tritium tracer method).<sup>8</sup>
15. Disintegration rates (external standard ratio (ESR) method).

Up to six of these subroutines can be used with any one set of data. It has been found that subroutines (1) and (2) are used for almost all sets of data, for checking counting results, and these are usually followed by one or two of the other subroutines. In most cases calculations are made on net count rates of samples above a background count rate, so that the order of samples and background in the counter is fixed for each subroutine, (except (1) and (2)). The maximum numbers of samples and cycles of counts are defined (200 samples for the Packard TriCarb 3320). An increase in the numbers of samples would require only minor changes in the program, but would involve more storage space in the computer. For all these subroutines (except (1), (2) and (11)) additional data (e.g. weights) are required; such data can be punched manually on parameter cards, or on paper tape, which accompany the count data to the computer.

In most subroutines the mathematics involved are straightforward, and most of the instructions are concerned with the manipulation of data and with the production of tables of results. Examples of the output from subroutines, used for processing liquid scintillation counting data, are shown in Figs. 1 and 2 from which the calculations involved are obvious. As an added example, further details are given below of one subroutine.

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GROSS COUNT RATES AND PER CENT STANDARD DEVIATIONS

RACK NO.	RED CHANNEL			GREEN CHANNEL		
	CPM	SOS	SDE	CPM	SOS	SOE
1	26.5	3.55	4.20	23.1	3.80	5.24
2	654.8	.71	.43	570.8	.76	.65
3	12460.9	.16	.30	10854.9	.18	.28
4	620.1	.73	.19	542.0	.78	.17
5	12139.0	.17	.15	10577.2	.18	.10
6	714.2	.68	.91	624.6	.73	1.12
7	12323.1	.16	.14	10742.3	.18	.12
8	626.6	.73	.82	543.8	.78	.55
9	11568.8	.17	.45	10087.4	.18	.45
10	529.1	.79	.79	460.4	.85	.61
11	11599.5	.17	.44	10119.7	.18	.39
12	663.8	.71	.35	581.0	.76	.52
13	11617.1	.17	.13	10131.4	.18	.15
14	681.3	.70	.53	594.3	.75	.64
15	11925.8	.17	.16	10390.0	.18	.18
16	503.9	.81	.64	439.4	.87	.38
17	10276.8	.18	.19	8940.4	.19	.11
18	64.1	2.28	3.59	55.2	2.46	4.25
19	349.2	.98	.60	309.4	1.04	.78
20	17713.2	.14	.29	15772.5	.15	.32
21	371.7	.95	.98	329.0	1.01	.68
22	17445.0	.14	.23	15505.4	.15	.30
23	383.1	.93	.50	338.7	.99	.45
24	17361.0	.14	.48	15432.2	.15	.53
25	379.3	.94	1.59	335.7	1.00	1.55
26	17327.0	.14	.43	15409.8	.15	.45
27	365.0	.96	.54	324.3	1.01	.42
28	17315.9	.14	.49	15397.1	.15	.51
29	348.7	.98	.58	307.9	1.04	.87
30	17295.2	.14	.05	15376.7	.15	.07
31	351.3	.97	1.31	308.5	1.04	1.27
32	17393.4	.14	.33	15460.4	.15	.30
33	345.0	.98	.98	306.4	1.04	1.41
34	17454.9	.14	.20	15537.7	.15	.18
35	11771.4	.17	.27	7102.7	.22	*46.83
36	41.6	2.83	4.41	36.7	3.02	3.47

\* ... THE COUNTS SHOULD BE CHECKED FOR ERRORS

RACK NO.	CHANNEL	CYCLE		
		1	2	3
35	GREEN	103977	104599	4505

Fig. 1. Output from subroutine (2).

**Subroutine (14): Oil consumption rates.** This subroutine is used to determine oil consumption rates from samples of tritiated water which have been condensed from the exhaust of an engine run with tritiated oil. The technique was originated by Guinn and Coit<sup>8</sup> in 1959 and since then has been developed considerably.

The disintegration rates of tritium in the samples have been measured in a liquid

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## DISINTEGRATION RATES (RED CHANNEL) AND STANDARD DEVIATIONS

RACK NO.	SAMPLE NO.	SAMPLE		STANDARD CPM	EFFICIENCY PER CENT	SAMPLE	
		NET CPM	SDM			DPM	SDM
2	WG1	628	5	11806	15.1	4173	35
4	WG2	594	5	11519	14.7	4040	33
6	WG3	688	7	11609	14.8	4644	46
8	WG4	600	5	10942	14.0	4300	43
10	WG5	503	4	11070	14.1	3560	35
12	WG6	637	5	10953	14.0	4561	36
14	WG7	655	5	11244	14.3	4565	35
16	WG8	477	4	9773	12.5	3829	35
19	OG1	285	4	17364	26.5	1077	16
21	OG2	308	4	17073	26.0	1182	17
23	OG3	319	4	16978	25.9	1233	18
25	OG4	315	6	16948	25.8	1220	26
27	OG5	301	4	16951	25.8	1164	17
29	OG6	285	4	16947	25.8	1102	16
31	OG7	287	5	17042	26.0	1106	20
33	OG8	281	4	17110	26.1	1077	16

SDM ... ABSOLUTE STD. DEV. (MAX. OF STATISTICAL AND EXPERIMENTAL)

Fig. 2. Output from subroutine (13).

scintillation counter by the internal standard method – for accuracy this was preferred to external standard methods (*cf.* Rogers and Moran)<sup>9</sup> owing to the presence of both colour and chemical quenching, and the channels ratio method was not used owing to the low levels of radioactivity. Results showing the count rates and efficiencies are shown in Figs. 1 and 2.

The oil consumption rates have been calculated in subroutine (14) from the relation:

$$R_o = \frac{S_w}{S_o} \times R_f (K_f + A \times H_a)$$

where  $R_o$  = rate of oil consumption (g/h)  
 $R_f$  = rate of fuel consumption (g/h)  
 $S_w$  = specific activity of water (d.p.m./g)  
 $S_o$  = specific activity of oil (d.p.m./g)  
 $K_f$  = water/fuel weight ratio, on combustion of the fuel to water  
 $A$  = air/fuel weight ratio  
 $H_a$  = water/air weight ratio, depending on the relative humidity. This is calculated from wet and dry bulb temperatures from a table of relative humidities, which is included in subroutine (14).

The results of these calculations, for the eight pairs of oil and water samples (36 vials in all, including standards and backgrounds) are shown in Fig. 3. Computer processing time for this operation (giving outputs as shown in Figs. 1, 2 and 3) is about 1.5 s – compared with many man-hours of ‘hand’ calculations or several minutes with a ‘desk-top’ computer.

Two examples will now be given of further uses of an off-line computer for processing liquid scintillation counting data.

RESULTS OF OIL CONSUMPTION TEST WITH TRITIATED OIL

ENGINE 0.11  
 FUEL 10  
 TEST NO. 327  
 DATE OF TEST 28/6/71  
 AIR:FUEL RATIO 23.0  
 OIL 8.4635-T  
 FRACTION TRITIATED HVI 160B  
 WT. PER CENT TRITIATED 91.50  
 GMS. WATER OF COMBUSTION PER GM. OF FUEL 1.215

SAMPLE	SPECIFIC ACTIVITY		WATER DPM/G	FUEL FLOW KG/HR	HUMIDITY CORR. PER CENT	3H-LABELLED OIL CONSUMED GMS. PER HR.	SD(ABS)
	3H-COMPONENT OF OIL DPM/MG	SD(ABS)					
J1	567	8	4173	2.81	12.2	28.6	.5
J2	573	8	4040	2.81	11.9	27.3	.4
J3	597	8	4644	2.81	10.5	29.7	.5
J4	591	12	4300	2.81	10.4	27.7	.6
J5	564	8	3560	2.81	9.8	23.9	.4
J6	573	8	4561	2.81	10.4	30.3	.5
J7	575	10	4565	2.81	10.8	30.4	.6
J8	560	8	3829	2.81	10.8	26.1	.5

FIGURES UNDER THE HEADINGS SD ARE THE ABSOLUTE STANCRD DEVIATIONS CALCULATED FROM THE COUNT RATES. FURTHER ERRORS MAY BE INTRODUCED DURING SAMPLE PREPARATION. THE PRECISION OF THE RESULTS IS ESTIMATED TO BE 5 PER CENT OR SD WHICHEVER IS THE GREATER.

Fig. 3. Output from subroutine (14).

Table 1. Repeatability of count rates from tritium samples.

Sample	Counter	Time of counting (min)	Temperature (approx) (°C)	Mean counts/min (c.p.m.)	SD of mean c.p.m. (%)	
					Statistical	Experimental
PH3	314 EX	250 × 10	- 3	12573	0.018	0.055
NPH3	3320	900 × 10	+10	83119	0.0037	0.0056

### Precision of measurements

Experience has shown that instability of samples often limits the precision of results of liquid scintillation counting more than instrumental fluctuations or statistical considerations of radioactive disintegration; however, it is necessary to determine the stability of the instruments in use, and this can easily be done with the use of a computer.

As radioactive disintegration is a random phenomenon, it is necessary to accumulate a large number of counts in order to assess small instrument drifts, and this is possible with the use of a computer. To find the stability of Packard TriCarb counters over several days, under normal operating conditions, sealed standards of tritium were counted in both the Models 314 and 3320 at balance point. Ten minute repeat counts were recorded on paper tape from each of these instruments — more than one week of counting time was needed. Results of these experiments are shown in Table 1, and were obtained from the computer for which a simple FORTRAN program was written similar to that used in a RADIOC subroutine. This shows remarkably stable operation of both instruments. It would not be practicable to check by hand the calculation of the standard deviation of the mean of 900 six-digit numbers, so this was done (successfully) on a randomly selected dozen of the repeat counts.

It was found that the stabilities of each of the two channels of the ten year old instrument were similar, and that all three channels of the 3320 were even more stable by a factor of about ten than those of the 314. In routine work it is possible also to check the repeatability of operation of the external standard and we have found that, at the channel settings used by us, the ESR counts in one minute are usually steady for any given sample with a standard deviation of the mean of less than 1%.

It is concluded that instrumental drift can be negligible compared with other errors; however, reliability is all-important for computer processing of data. The ten year old instrument now produces so many errors on punched tape that it is being superseded by the modern instrument, which has a teletype output and has so far this year produced only one error in more than 500000 characters.

### Curve fitting

When FORTRAN programs and a Univac 1108 computer have been used, it has been found that the fitting of curves to experimental determinations of ESR and efficiency is straightforward for the radiochemist, particularly when existing subroutines are available.<sup>10</sup> One important advantage of our off-line computer for curve fitting is that different types of curve can be fitted to experimental points, and it is possible to select the equation appropriate to the experimental conditions. This is illustrated in Fig. 4 which shows the fit of quadratic, cubic and logarithmic curves to experimentally determined values of carbon-14 efficiency and ESR. It is evident that, for the instrument settings used and the

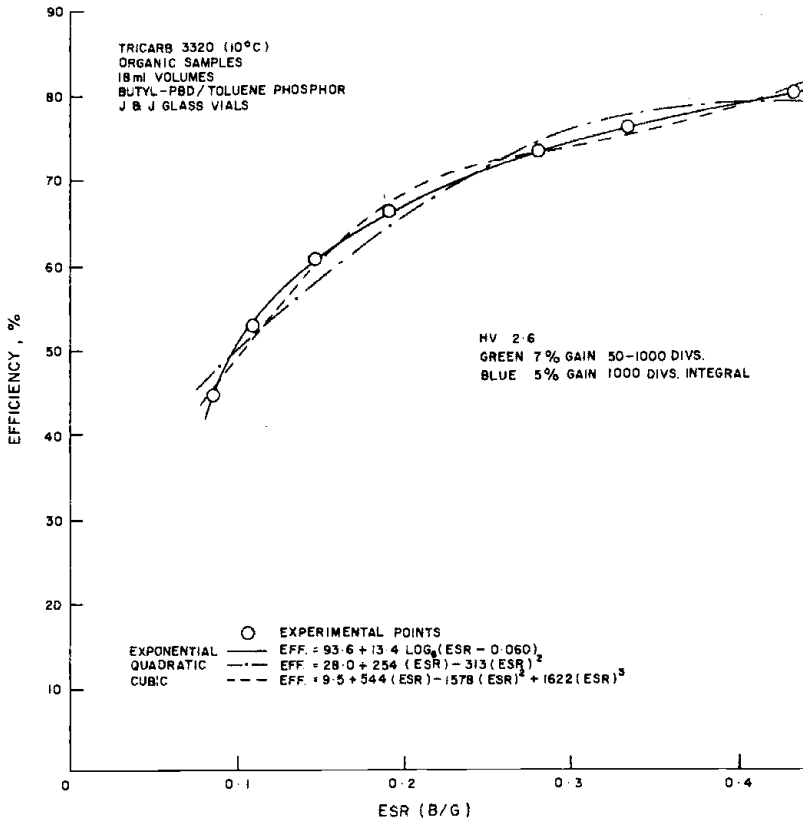


Fig. 4. Relation between carbon-14 efficiency and ESR.

range of quenching with which we are concerned, a cubic does not give as good a fit as the simple three constant expression:

$$\text{Efficiency} = A + B \log_e(\text{ESR} - C)$$

where A, B and C are constants.

## DISCUSSION

### Published work

Many papers have been published, particularly in the last few years, on the subject of data processing of liquid scintillation counting data and only a few can be mentioned here. Blanchard<sup>11</sup> described an early general program in ALGOL, which included a data checking stage and various modes of operation for the determination of background and counting efficiency. Spratt<sup>12</sup> described another general program in FORTRAN IV, which was designed for use with punched card output from the spectrometer. An unusual feature was that for application of the internal standard method two separate runs through the computer were required, for the results on the samples alone and for results on the samples plus the internal standard. This was extended to include the use of an automatic external standard.<sup>13</sup> Other recent studies have been made by Clegg,<sup>14</sup> Reich<sup>15</sup> and Glass.<sup>16</sup>

The extensive studies of Gordon and co-workers<sup>17,18</sup> covered both the channels ratio and external standard methods, and they show several interesting features. These workers recognized that when a computer is used it is unnecessary to find a setting giving a linear relation between counting efficiency and channels ratio, as has usually been preferred for graphical methods; instead they expressed the counting efficiency as a cubic in the logarithm of the channels ratio. They also examined instrumental drift with the object of avoiding the need to redetermine the whole calibration curve regularly. They found it was valid to measure just one standard each day, the instrument being then brought back on a standard calibration curve by adjustment of the gain. Their application of the external standard was to a dual-labelling experiment, and thus involved four counting efficiencies (two nuclides  $\times$  two channels). Four calibration curves were used so that the four efficiencies could all be determined from the measured external standard count rate.

The value of having a standard error for every result is emphasized by Nodine.<sup>19</sup> In his programs the standard error is carried through the calculation, i.e. standard errors are calculated explicitly for intermediate results such as net counting rates. We have also considered it important that final results should have their standard errors. The calculations involved are generally longer than those used in obtaining the results alone, and are often omitted as being too tedious for manual calculation. In the case of the channels ratio method, the need for standard error calculations has led to different ways of treating the information from the two channels.

The use of the channels ratio method for dual-labelling experiments can lead us into non-linear simultaneous equations which may give multiple solutions: this approach has been studied by Krichevsky and co-workers.<sup>20</sup>

Today Spratt and others have described to us their methods of data processing of liquid scintillation counting data; we have heard about the processing of data from multi-labelled samples, and it seems evident that the computer is rapidly becoming an indispensable instrument for the radiochemist.

### **Advantages of computer processing**

The main advantage in the computer processing of liquid scintillation counting data is the saving of time. As illustrated above, results can be obtained in a few seconds of computer processing time, compared with hours for manual processing. However, the time needed for writing a correct subroutine is of the order of a few days, and the procedure is only worthwhile for experiments which are repeated many times, or where large numbers of samples are involved.

Over a period of about eight years, we have found that a further advantage of computer processing is in the elimination of errors. Many checks have been made between results from the computer and those found by calculation with a desk calculator; differences, once a correct program has been written, have always come from errors in the manual calculation.

By the use of a computer many more detailed calculations can be made on count data, calculations which would be too tedious manually, particularly in checking count data. Samples can be counted several times without the need for tedious manual averaging, so that comparisons can be made of statistical and experimental standard deviations of mean count rates, as described earlier. These have shown up several types of error, some of these being as follows.

Occasionally, errors occur in the operation of the counter; particularly, a numeral is incorrect in both the printed and the paper tape output. (One such error is shown in Fig. 1).

Systematic drifts have occurred in repeat counts of long-lived isotopes. These have been traced to adsorption of part of a sample on the walls of the counting vessel, long-term phosphorescence of dioxan-based samples due to exposure to sunlight, and increasing quenching in samples due to slow colour-producing reactions.

Finally, it has been found possible by computer processing to produce tables of results in easily readable form. Results from some subroutines (e.g. those shown in Figs. 1 and 2) are of use to the radiochemist making the measurements, and others (as shown in Fig. 3) can be given directly to the scientist in another discipline who wishes to make use of the results; in either case a well-edited table greatly assists clear communication – nowadays some scientists seem only to believe results which are produced by ‘the computer’.

### **Future developments**

There is considerable interest today in the use of small on-line computers in the laboratory. I have found that access to a large off-line computer is very convenient for radiochemical work, particularly for liquid scintillation counting. It is easier to write a FORTRAN program for a large computer with plenty of storage space than it is to write non-standard programs for small computers. It may well be more economical to use a few seconds per day on a large computer than longer times on a small computer. However, the amount of money available and the nature of the work will determine the relative merits of the off-line and the on-line computer. One of these types of computer is becoming essential; ideally, the radiochemist would like both.

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## **DISCUSSION**

**P. Stanley:** Were your measurements made at 'balance-point' or not?

**J. H. Deterding:** Yes indeed, all samples were measured at 'balance-point'.

**P. Stanley:** Balance-point operation will nullify the effects of spectrometer drift. To observe spectrometer drift it would be perhaps better to set two channels to monitor the top 5% and the whole of the energy range of the pulse height spectrum and then look for any significant change in the ratio of the two rates. A fairly hot standard (say  $5 \times 10^5$  d.p.m.) would be necessary if one wished to observe short-term drift since it would be necessary to accumulate a statistically acceptable number of counts in a short time for a valid test.