Preprint, *Nuclear Geophysics,* 1988, v. 2, no. 2.

Performance of New Technology Liquid Scintillation Counters for ^{14}C Dating

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The results are presented of an investigation comparing the performance of commercially available liquid scintillation spectrometers claiming 'low-level' radioisotope detection abilities. Determination of ^{14}C at naturally occurring concentrations was carried out in both old and new technology liquid scintillation counters using a ^{14}C labelled benzene sample with butyl-PBD as scintillant. The signal to noise ratio and the ^{14}C detection efficiency were evaluated.

The results show a wide range of merit for radiocarbon dating of so called 'low-level' instruments.

Introduction

Radiocarbon dating is a geochronological research tool used commonly in archaeology, geosciences, climatic, floral and faunal history, oceanography, hydrology and environmental sciences such as pollution studies, volcanology and earthquake prediction. There are some 117 registered Radiocarbon Dating Laboratories (1) and at least half as many unregistered, all meeting the large demand for ^{14}C assays. Two techniques of ^{14}C radiometric determinations are practiced today: gas proportional counting as reviewed by Mook (2) and liquid scintillation counting as reviewed by Polach (3). In recent times, thanks to the generally good performance of commercially available liquid scintillation spectrometers, laboratories using these spectrometers dominate. It has thus become pertinent to evaluate the performance of the newly available liquid scintillation spectrometers claiming 'low-level' detection ability and to compare them with the old technology counters.

Experimental

Apparatus and ^{14}C standard.

Two commercially available liquid scintillation spectrometers claim 'low-level' performance, defined as high net isotope signal represented by efficiency (E) and very low background (B), resulting in a large Figure of Merit (FM = E^2/B). These are the Packard 2000 CA/LL (USA) and the Pharmacia-Wallac Quantulus (Finland) liquid scintillation spectrometers.

To monitor their performance two standards were prepared and distributed by the Australian National University, ANU-Radiocarbon Dating Research Laboratory: a labelled ^{14}C benzene sample and a ^{14}C free benzene-background sample, both with butyl-PBD scintillant at 15 g per litre (g/L). The ^{14}C concentration of the labelled standard corresponded to 25.7 $\pm$ 0.2 dpm/g benzene. This activity is equivalent to 205.6% *Modern*, where *Modern* is represented by 95% of the ^{14}C activity of the National Bureau of Standards (Washington DC) oxalic acid radiocarbon dating international standard [(4)] (0.95Ox = No cpm in the tables and figures that follow).

Each participant used the counters available to him (old or new), optimised to manufacturer specification for low-level detection of ^{14}C and reported the observed net ^{14}C and background counts for 3, 7 and 15 mL of the supplied standard solutions. Because Packard recommends a 6 g/L butyl-PBD concentration the Packard users tested the standard as supplied as well as diluted with analytical reagent grade benzene (^{14}C free) to achieve the recommended scintillant concentration.

Results

Manufacturers of nuclear counters prefer to express the performance of their equipment in terms of the Figure of Merit (FM) given by E^2/B, whilst users involved in radiocarbon dating prefer a *factor of Merit (fM)* given by $No/\sqrt{B}$ [(5)]. Both terms are given in the table of results (Table 1) but the interpretation of data is based on the *factor of Merit*.

The participants supplied data relating to counting vials (size, volume and grams of sample) and the observed results as net cpm and background. The interpretative data (%E, No cpm, *fM* and FM) was calculated at the Australian National University, as none of the participants were advised of the precise radiocarbon concentration of the standards supplied. The data as tabulated therefore enables a relative intercomparison of counting systems.

Two types of counting vials were used: the standard low potassium glass, ~ 20 mL, liquid scintillation counting vial as recommended by Packard, and the Teflon vials, Australian Nuclear Science and Technology Organisation (ANSTO) [(5)] and Australian National University [(6)] as recommended by Pharmacia-Wallac.

Four types of counting environments are represented: 1) surface location with some attenuation of cosmic radiations by ceilings of buildings (Calf and Harkness), 2) surface location with attenuation of cosmic radiation by 1.5 m of concrete on the sides and above the equipment (Polach), 3) surface location in cosmic ray shielded and γ-ray free environments (low-level lab, LoLa, Kaihola [(7)]), and 4) the laboratory in the Warragamba dam wall, with very high cosmic shielding [(8)]. Allowances will be made for these environmental factors when interpreting the results.

Interpretation of results

The detection efficiency values range from a low 42.8 to a high 84.9 %E and the *factor of Merit* values range from a low 15 to a high 172 *fM*. The old technology counters, represented here by the Beckman 7500, LKB-Wallac 1215 Rackbeta-K and Packard 3330, give values for E of 70.4 to 77.6% which is as expected. The new technology counters contrast in that the Packard-2000 CA/LL efficiency values range from a low 51.7 and do not exceed 57.1% at manufacturer recommended butyl-PBD concentrations, whilst the Pharmacia-Wallac Quantulus gives systematically high values for E at 70.0 to 84.9 %. The lowest backgrounds are achieved in the Wallac low-level laboratory location by the Quantulus. This is as expected as both cosmic and environmental γ-rays have been attenuated in this special environment. The excellent background achieved by the old Packard 3330 located at the Warragamba dam (ANSTO) confirms the merit of cosmic and γ-ray shielding. The laboratory at the Australian National University does not perform the same function. With 1.5 m of ordinary concrete it attenuates the soft, therefore variable component of cosmic rays, without affecting the hard muon flux. The γ-rays from the concrete aggregate also contribute to the observed background. The design aims were to achieve a constant not a low background. Tests have shown that the background value is the same as at any other surface location at the Australian National University, albeit free of externally induced variations or interference. The values of B achieved by the old technology counters (Beckman and LKB) are therefore representative, at 3 to 4 cpm, of that type of technology which contrasts with the Quantulus at 0.14 to 1.1 cpm. The new Packard 2000 CA/LL gave results for B ranging from 0.73 to 3.5 cpm.

The relative merit of radiocarbon dating systems is given by plotting (Fig 1) the derived radiocarbon dating reference standard values (No cpm, from Table 1) against the *factor of Merit* values [9]. The countrate of No (Y axis) relates to the ultimate precision that can be achieved by a counting system / vial / sample size configuration. In practice the counting error of a sample, whose countrate is close to the modern reference standard, is primarily given by the Poisson error of the accumulated counts in a given time. The background, and thus the environment, play only a minor role [10]. The value of No is both sample size and efficiency related; therefore the precision of an age determination, for a given counting time, increases with both sample size and counting efficiency.

The *factor of Merit* is plotted on the X axis and by definition takes account of the values No and B. The derived values are thus directly related to the maximum age that can be determined [5] and are dependent on the counting efficiency of systems, their background and hence the environment in which the counters are placed. The higher the value of *fM* the higher is the ultimate age resolution and hence the greater the maximum determinable age.

The three shaded areas in Fig. 1 represent the three counting systems we wish to compare. From left to right: K and Bn are the old counters; H and C are the only marginally better new Packard; and Q is the significantly better new Pharmacia-Wallac Quantulus; all placed in virtually the same environment.

An alternative to the above presentation is to calculate (based on the cpm values of No and B, Table 1) the errors associated with the age determinations and the radiocarbon dating age limits and to plot the results as in Fig. 2.

Selected from Table 1 were, for purposes of direct comparison in Fig 2, results relating to the 3 mL sample size. The error was calculated using a 1000 minute counting error for standard (No) and background (B) and a 3000 min error for the sample, as is the practice at the Australian National University [10]. The age determination limits were calculated using the internationally accepted 2 σ detection criterion [4]. It is seen that the Packard 2000 CA/LL results give a slightly higher error (due to their lower efficiency) and slightly better old age limit (due to their lower background) than the 'Old technology' counters. Only the Pharmacia-Wallac Quantulus in a normal environment, or any liquid scintillation or gas proportional counter placed in an underground laboratory (as demonstrated by the Warragamba experiment), has achieved a higher precision and greater age resolution.

Conclusion

The results of the intercomparison of liquid scintillation ^{14}C counting systems indicate that, in terms of effectiveness for radiocarbon dating, there is a need to maintain high efficiency whilst significantly reducing the background. Background reduction can be achieved by placing any β-particle counter in a deep underground / under water / low γ- and cosmic-ray radiation environment. The results further indicate that the term *'low-level liquid scintillation spectrometer'* does not by itself guarantee improved ^{14}C resolution as compared to those liquid scintillation counters that make no such claims. This paper gives unambiguous parameters enabling the assessment of the merit of any counting system for radiocarbon dating.

References

1. *Radiocarbon,* American Journal of Science, 1956 to present.
2. Mook W. G. *Radiocarbon* **25,** 475 (1983).
3. Polach H. A. *Radiocarbon* **29,** 1 (1987).
4. Stuiver M. and Polach H. A. *Radiocarbon* **19,** 355 (1977).
5. Calf G. E. and Polach H. A. In *Liquid Scintillation Counting: Recent Advances* (Eds Stanley P. E. and Scoggins B. A.) pp. 223-234 (Academic Press, London, 1974).
6. Polach H., Gower J., Kojola H. and Heinonen A. In *Advances in Scintillation Counting* (Eds McQuarrie S. A., Ediss C. and Wiebe L.) pp. 508-522 (Univ. Alberta Press, Edmonton, 1983).
7. Kaihola L., Kojola H. and Kananen R. *Nucl. Instr. & Meth.* **B,** 509 (1986).
8. Calf G. E. and Airey P. L. In *Archaeometry: an Australasian Perspective* (Eds Ambrose W. and Duerden P.) pp. 351- 356 (Australian National University, Canberra 2601, Australia, (1982).
9. Polach H. A. *Nucl. Instr. & Meth.* **B** (1987, in press).
10. Gupta S. K. and Polach H. A. *Radiocarbon Dating Practices at the Australian National University* (Radiocarbon Dating Research, Garran 2605, Australia, 1985).

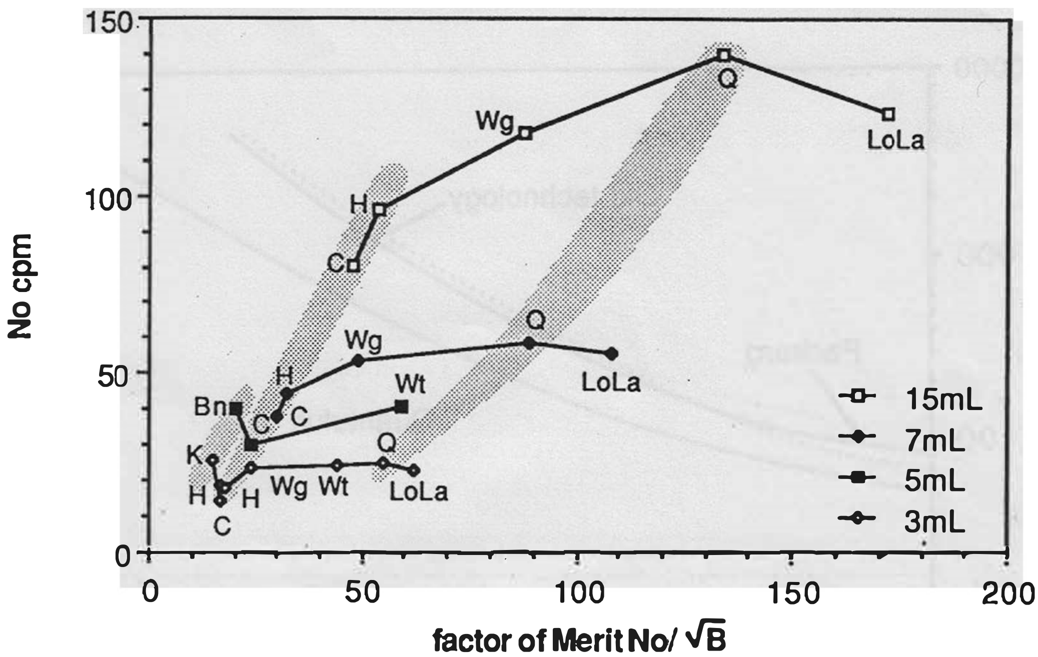

Fig. 1. Comparative merit of liquid scintillation spectrometers for ^{14}C dating. Represented are: 'Old technology', Beckman 7500 (Bn) and LKB-Wallac 1215K (K), left shade, teflon vials (t); 'New technology' Packard 2000 CA/LL (C and H), centre shade - glass vials (g); 'New technology' Pharmacia-Wallac Quantulus at the Australian National University location (Q), right shade and the Low-level Laboratory at the Wallac Oy location (LoLa), both teflon vials; and the 'Old technology' Packard 3330 at the Warragamba dam low-level laboratory (W both (t) and (g) vials). The merit of a liquid scintillation counter for radiocarbon dating is based on: (a) the countrate for the radiocarbon reference standard (No cpm), and (b) the value of the *factor of Merit*. The higher the values of (a) and (b), the better the counter.

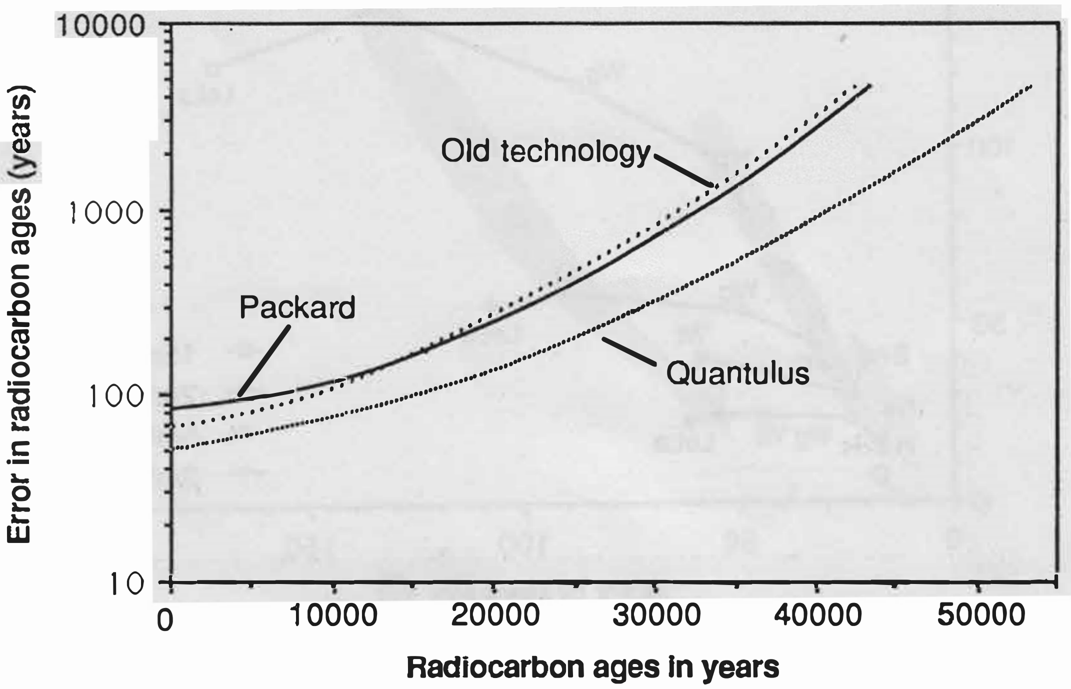

Fig. 2. Achievable resolution of liquid scintillation spectrometers in terms of the radiocarbon dating error and age resolution. Selected for comparison purposes were the results (Table 1) relating to 3 mL of the same sample. Thus, the performance of various counters can be assessed on equal terms. The 'Old technology' and the Pharmacia-Wallac Quantulus are represented by the dotted lines. The Packard 2000 CA/LL (solid line) crosses but nevertheless closely follows, in terms of resolution for radiocarbon dating, the performance of the 'Old technology' counters.

Table 1. Performance of selected liquid scintillation counters for radiocarbon dating

Name counter	Vial type	Vial mL	b-PBD g/L	benzene mL	benzene g	Net count cpm	B count cpm	Efficiency %	No (95% Ox) cpm	*factor of M* $No/\sqrt{B}$	Fig of M E^2/B
CALF	glass	20	15	15	13.115	166.0	2.85	49.2	80.74	48	850
Packard-2000		20	6	15	13.167	183.0	3.48	54.0	89.01	48	840
CA/LL											
		20	15	7	6.125	78.0	1.58	49.5	37.94	30	1550
		20	6	7	6.162	84.0	1.87	53.0	40.86	30	1500
		20	6	5	4.445	62.0	1.55	54.2	30.16	24	1900
		20	15	3	2.634	29.0	0.73	42.8	14.11	17	2510
		20	6	3	2.631	35.0	1.19	51.7	17.02	16	2250
CALF	glass	20	15	15	13.115	241.7	1.79	71.7	117.56	88	2870
Packard-3330		20	15	7	6.125	110.9	1.21	70.4	53.94	49	4100
in		20	15	3	2.634	48.5	0.96	71.6	23.59	24	5340
Warragamba											
dam	Tef/ANSTO	5	15	5	4.394	84.3	0.48	74.6	41.00	59	11590
		3	15	3	2.656	49.8	0.31	72.9	24.22	44	17140
POLACH	Tef/Cu	15	15	15	13.191	288.2	1.10	84.9	140.18	134	6560
Pharmacia-Wallac		7	15	7	6.155	121.3	0.44	76.6	59.00	89	13340
Quantulus		3	15	3	2.641	51.9	0.21	76.4	25.25	55	27820
Beckman 7500	Tef/Cu	7	15	5	4.395	82.1	3.92	72.6	39.94	20	1350
LKB-Wallac 'K'	Tef/Cu	3	15	3	2.641	52.7	3.02	77.6	25.63	15	1990
HARKNESS	glass	20	7	15	13.124	195.2	3.16	57.9	94.95	53	1060
Packard-2000		20	15	7	6.121	84.8	1.70	53.9	41.22	32	1710
CA/LL		20	15	3	2.642	37.7	1.27	55.5	18.32	16	2420
		7	6.5	7	6.171	90.5	2.02	57.1	44.00	31	1620
		7	15	3	2.641	36.8	1.01	54.3	17.91	18	2910
KAIHOLA	Tef/Cu	15	15	15	13.185		0.52	75.0	123.80	172	10820
Quantulus		7	15	7	6.150		0.27	73.0	56.20	108	19740
in 'LoLa'		3	15	3	2.640		0.14	70.0	23.10	62	35000

ANSTO = Australian Nuclear Science and Technology Organisation; B = Background; M = Merit; LoLa = Low-level Laboratory, Wallac Oy; Tef/Cu = Teflon/Copper