THE 'WINDOWLESS' APPROACH TO SCINTILLATION COUNTING: LOW-LEVEL ^{14}C AS AN EXAMPLE

Henry Polach, Steve Robertson, Derek Butterfield* and John Gower
Radiocarbon Research Laboratory and *Department of Chemistry,
Australian National University, Canberra ACT, Australia

Erkki Soini
Research Department, Wallac Oy, Turku, Finland

ABSTRACT

Multichannel analyser (MCA) relative pulse height (RPH) spectra were used to monitor linear and logarithmic output liquid scintillation counters. Spectra for radiocarbon dating reference standards, known age samples and background were recorded. The counters tested were optimized for low-level ^{14}C counting. Their high voltage (EHT) was reduced, the phototubes were masked and the limited counting efficiency windows (65-80%) were set at balance point. The total count from the conventionally set window as well as the 1000 (0-250 KeV) channel relative pulse height (RPH) spectrum was recorded. Spectral analysis could be carried out for the total 0 - 1 MeV energy range or within the counter set lower and upper limit window discriminators. This enables (i) MCA and set window computations to be compared, (ii) counter windows to be checked or optimized (iii) merit of computations based on total spectral information to be evaluated and (iv) merit of linear or logarithmic data presentation to be evaluated. It will be shown that pulse height (PH) spectra analysis based on WINDOWLESS data acquisition is superior in all aspects to the conventional set window mode of data analysis. Full energy spectra information allows speedy recognition of: non-Poisson (non-random) events, quenching, radioactive impurities, counter instability and, especially for low-level counting, evaluation of long term spectral stability. Applied to radiocarbon dating it allows computations of minimal errors.

It is suggested that PH analysis and WINDOWLESS data acquisition is even more pertinent to high count rate liquid scintillation spectrometry and that there is no difference if based on linear (lin)

or logarithmic (log) output provided some 80 channels of analysis can be brought to bear on the isotope of interest or each individual region of interest.

INTRODUCTION

Liquid scintillation spectrometers, as manufactured today, give an opportunity to monitor integrated counts in variable width windows. These windows, in general terms, view the region of interest by adjusting their width through lower and upper limit discriminators. These energy (pulse height) discriminators are either program preset or user variable to accommodate isotope detection needs. The number of available windows vary from 1 to 5 and cover the energy range of 0 to 2 MeV. It is true that modern counters, for example Beckman, LKB-Wallac, Packard and Philips do store internally multichannel (MCA) relative pulse height spectra in 80-1000 channels, depending on make. In the most expensive versions of these, the spectra can be viewed. It remains nevertheless true that only internal, often hardware programs, can make use of the information they contain. The spectra are not readily user accessible and the programs are not user friendly. That is, they cannot be modified by the user. Further, only one sample, one spectrum at a time can be accumulated. Once calculations are performed, the spectral information is lost unless dumped to an external device.

We consider this to be a handicap, especially for low-level counting, where long counting time (days), long term stability (years) and reproducibility (10 years or more) are important factors for maximum precision. In this paper, we shall examine a WINDOWLESS approach to counting using an external multichannel analyser on line with a central computer to store and analyse all data. We shall compare windowless with fixed window based results and demonstrate the advantages and disadvantages of external MCA pulse height energy spectra acquisition for radiocarbon dating and liquid scintillation spectrometry.

EXPERIMENTAL PROCEDURES

Counters. Available for our study, carried out at ANU, were counters manufactured by Beckman, Packard and LKB-Wallac. Their characteristics are given in Table 1. It is interesting to note the different ways data presentation was achieved, a point we shall return to later. All the above counters were modified by us in collaboration with manufacturer trained, Australian distributor, engineering staff. The modifications allowed variable EHT and gain in a calibrated, re-settable mode. For the last three counters we have also include variable coincidence bias and reduction in coincidence resolution time. These changes were carried out to optimize counters electronically for highest radiocarbon dating factor of merit[1].

Table 1. Characteristics of LS counters as supplied to the RC Dating Research Laboratory, ANU.

Manufacturer	Model	(year)	Data Presentation (achieved by)	Gain	EHT
Beckman	LS-200	(1966)	pseudo-log MPT last dynode bias	V	F
Beckman	LS-250	(1976)	log - amplifier	V	F
Beckman	LS-7500	(1980)	log - amplifier	F	F
Packard	3255	(1974)	lin - amplifier	V	V
LKB-Wallac	1215	(1980)	log - lin - amplif. - log discrim.	F	F

V = Variable; F = Fixed; MPT = multiplier phototube

All counters upon receipt (as per year in Table 1) were interfaced to an external MCA. In the early stages, up to 1975, we used the MCA for instructional and window setting purposes. Only during our work in preparation for the 1976 Banff conference, when studying the effects of vial holders on ^{14}C and ^{3}H efficiency[2], did we realize the intrinsic value of MCA data for low-level computations. Thus, whilst early interfaces were elementary, the aim of recent designs was to enable the monitoring of singles and coincident pulse height spectra prior to and after discrimination. We were not always entirely successful as discriminator limits were often distorted and smudged.

However, for our latest counters, Beckman and LKB-Wallac, modern design and stable noise free components, sophisticated interfaces[1,3,4] and a Canberra series 40 MCA resulted in sharp and clear spectra and discriminator limit definitions. This enabled a precise comparison of the set window and windowless approaches under identical conditions.

The spectra and their usage for RC dating. All spectra are shown as the logarithm of relative pulse height (energy), mostly in terms of channel numbers. The spectra cover the range of 0-250 KeV with 1000 channel resolution. A typical ^{14}C + Background spectra based on the 198.2% Modern Harwell (U.K.) radiocarbon standard[5], is given in Fig 1. The spectrum is typical of our modified counters with reduced EHT (~ 300 V less than factory set) and gain adjusted for maximum stability[1] and spectral symmetry. For purposes of this study the coincidence bias was set at the lowest permissible limit of 1 mV[1]. Typical background is inset in Fig 1, based on a counter with 5 cm lead shielding. Note, that due to EHT reduction, and especially due to rejection of optical cross-talk using amplitude disparity discrimination[1,6] the background is almost flat; it is lacking the characteristic sharp rise generally expected at lower energies[7]. Vials used for test were borosilicate glass, 5 ml shielded of design 'A' using pure benzene + butyl PBD[8].

For radiocarbon dating, counting is not conducted at the optimal window[9] as recommended by the manufacturers but at the balance point window[1,10,11], the stability of which has been demonstrated[12]. (The difference between optimal and balance point window is also demonstrated in Fig 7 later in the text). A computer generated balance point family of curves is given in Fig 2, based on C14 + B spectrum of Fig 1 by integration of a preselected number of channels (Soft Window, SW). SW is an odd number so that the sum of counts can be plotted against SW mid-point (SWM). SW is moved from left to right in increments of one (for example 0-201, 1-202 etc.). We have chosen to plot % efficiency (%EF) against SWM so that we could further calculate the Figure of Merit at various points as per Fig 6. Moving the SW across the spectrum as in Fig 2 and others, was done beyond the region of interest for ^{14}C isotope for demonstration purposes only.

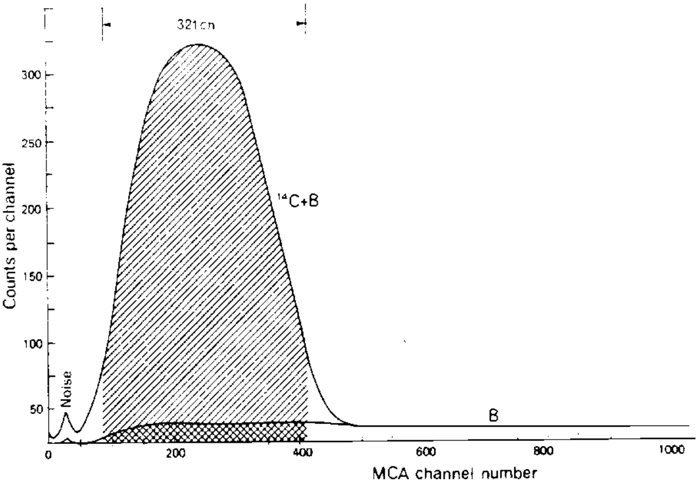


Figure 1. MCA spectra for ^{14}C + B and background (B) from 0-250 keV with 1000 channel resolution. Shaded area represents the computer selected limited window best suited for ^{14}C dating.

In practice, it would be restricted to the region of interest as given by the shaded area of Fig 7. The background spectrum is analysed in the same manner in Fig 3. It is noteworthy that the slope is increasing from left to right and not decreasing as would be the case of high background at ^{3}H energies[7]. Based on the highest %EF achieved at balance point (Fig 2) and the equivalent SW for background (Fig 3) we can calculate the optimal window for the highest Figure of Merit (FM), based on $EF/\sqrt{B}$. This is an appropriate expression as the same volume vial (5 ml) was used throughout the experiment. The FM thus gives a relative measure of apparent improvement due to moving the soft window (SW) across the RPH spectra.

For low-level counting of ^{14}C and ^{3}H, two levels of detection sensitivity need to be evaluated. There is need to distinguish count rates close to the Modern reference standard and count rates close to background[13]. The conditions which optimize both are not the same. For ^{14}C close to Modern standard we deal, on average, with a signal to noise ratio (S/B) of 4:1 to 10:1. Therefore, the higher the efficiency, the higher the count rate, the smaller the error and the

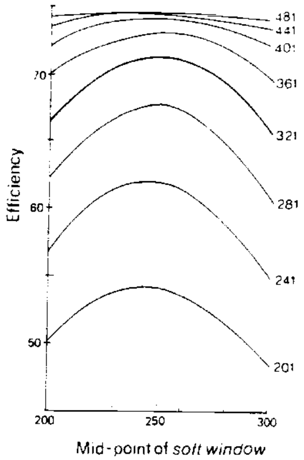


Figure 2. Balance point curves are determined by moving the SOFT WINDOW (SW) across the ^{14}C + B spectrum (Figure 1). The integrated count rate, between SW limits expressed as % efficiency, is related to the mid-point of SW for different window widths.

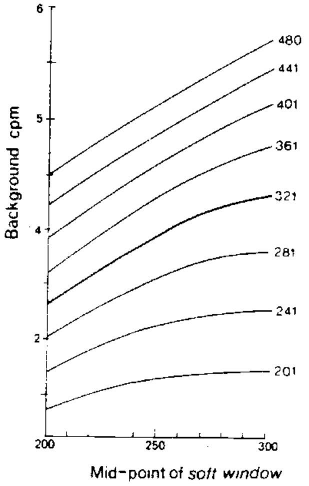
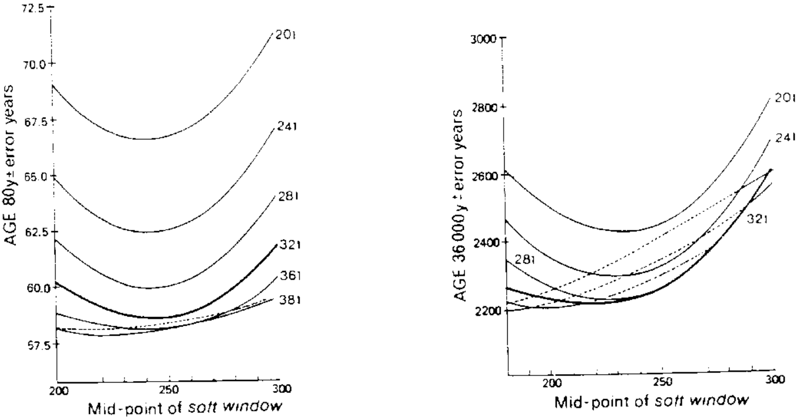


Figure 3. Moving SW across background (B) spectrum of Figure 1 gives a family of curves relating B cpm to mid-point of SW. There is no balance point.

better the resolution. From our spectral analysis point of view, the labeled isotope spectra dominate. For ^{14}C dating close to background the signal to noise ratio is inverted. The background signal (and spectrum) becomes dominant. The lower the background the better the resolution. An ideal counter would have efficiencies approaching 100% and backgrounds approaching 0 cpm. Underground installations, deep in a shaft[14] or deep under water[15] approach this ideal. Surface installations, which are prevalent, suffer in comparison. Therefore, there is need to minimize errors (optimize resolution). How the WINDOWLESS approach enables to precisely assess these errors is demonstrated in Figures 4 and 5. Counting errors (in years) are plotted against the window midpoint for a sample whose ^{14}C activity is 99% Modern (i.e. 80 y old) in Fig 4 the same counting errors are given for a sample whose ^{14}C activity is 1% Modern (i.e. 36,000 y old) in Fig 5. It can be seen that the error of determination is dependent



Figures 4 and 5. Knowing SW cpm for ^{14}C standard and background one can calculate error curves for a sample which is 80y old (Figure 4) and 36,000y old (Figure 5) for different window widths.

both on the window width and its placement. Generally the error is minimized at balance point. The integrated count rate based on the largest number of channels (i.e. full ^{14}C spectral width, 441 channels) never gives the best results. An evaluation of the windowless approach based on full MCA spectra is summarized in Fig 7. It is seen that the detection efficiency of the integrated count rate within a preselected number of channels (SW = 321) based on the gross count rate of the ^{14}C isotope spectrum rises to balance point which coincides with the lowest error of determination both for Modern (σM)

and Old (σO). It is further seen that none of these points correspond to optimal window based on Factor of Merit, Ef/$\sqrt{B}$. The shaded area indicates the region of stability where change in counting characteristics due to quenching or electronics cannot be distinguished (i.e. the computer derived results lie within ± 0.25σ). The slope of the background curve (B) is exaggerated by a factor of 4.

DISCUSSION AND CONCLUSIONS

We shall now compare fixed window to the windowless approach. Our fixed window settings are chosen at balance point experimentally determined from gross isotope count rate, the window width experimentally determined by multiple E/$\sqrt{B}$ evaluations[1], taking a minimum of 3 months to complete. The pulse height output was

Figure 6. Figure of Merit (Ef/$\sqrt{B}$) is determined for each SW based on ^{14}C signal to noise relationship with respect to SW mid-point.

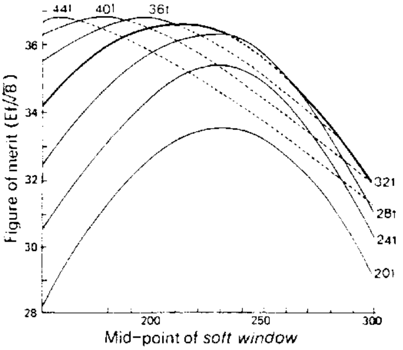
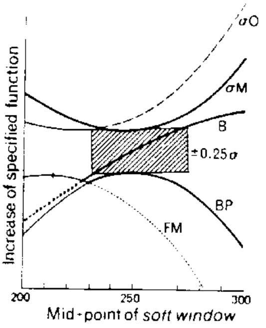


Figure 7. Complexity of relationship between minimum error of ^{14}C count rate determination for Modern (σ_M) and Old (σO), counting stability as given by balance point (BP) settings, Figure of Merit (Ef/$\sqrt{B}$) and changes in background (B), exaggerated 4x, demonstrated by the SW = 321 channels curves of previous figures. Region of maximum RC dating stability and merit is shaded. It does not coincide with max FM.



simultaneously recorded in our external MCA. Tests were carried out using a ^{14}C Modern (M) standard as reference. Sample activities, corresponding to 99%M (80y), 50%M (5730y) and 1%M (36,000y), were checked on the most recent three counters. The age calculations for fixed and windowless agreed within Poisson error limits. However, the precision of determination was not the same. The windowless Poisson error was reduced by a minimum of 10%, for the best manually set up counter, and by 30% in the worst, which we have however considered carefully optimized for low-level counting. We conclude that precise tuning is possible only by computer based analyses using the windowless approach on full MCA spectra. Should error reduction not be the researcher's aim, then this translates in reduction of time spent counting each sample. Using on-line computing facilities, the counting time could be determined by the desired error. However, other advantages of MCA analyses and windowless counting have emerged. Advantages. We could distinguish, just by looking at spectra, non-random noise pulses such as radio frequency interference, static electricity, component breakdown, noisy discriminators, or cyclic events. Fig 1 illustrates such noise, which was established to be count rate dependent, (cf. its presence on the background spectrum) hence now in all likelihood can be identified as after pulse coincident count rates[1]. The windowless approach also enables rejection of individual noise pulses, i.e. spectrum smoothing, rather than rejection of a whole counting interval, as would be the case in fixed window integrated count rate repeat sample counting.

We could confirm that, due to usage of b-PBD and stability at balance point, the count rate within ^{14}C window was not affected when quench or changes in gain were simulated. The quench correction dilemma by external standardization[12] was confirmed in hours, whereas the cited experimental evidence took weeks to accumulate. We can thus further confirm the doubtful value of using high energy external standards such as ^{137}Cs or ^{137}Cs with ^{133}Ba to monitor changes in low energy isotope under balance point conditions[12,16].

The proposal of simultaneous background and sample monitoring[17] as against our quasi simultaneous short term vial multiple-vial cycling[10] becomes attractive and against consecutive single-vial one-day counting, very attractive. Based on total spectra, rather than integrated fixed window counts one may well be able to derive a precise background-count rate equivalent from the sample spectrum above the isotope end-point. (i.e. ratio of non-shaded B/shaded B Fig 1).

Presence of Radon contamination of the sample for RC dating can be recognized at once, due to the shift in ^{14}C end-point and an increase in count rate of background above end-point.

We could not distinguish merit of pseudo log, log, or lin amplification or discrimination modes of data presentation. We confirm that the initiating events, sample β^- induced or background (γ, muon, neutron, x-ray, etc.) induced, retain the same signal to noise relationship. Hence net count rates, Figures of Merit, balance points, errors, etc., are identical. The manner in which the MPT pulses are processed (log or lin) is unimportant and the user of LS counters should not be persuaded by sales literature to the contrary. We have not only checked this on all types of equipment listed in Table 1, but have also repeated the computer simulation experiments[18] with the same results: transforming lin to log spectra (or reverse) with equal ease and resolution provided there were enough channels of discrimination.

We have established that the minimum number of channels required to retain sensible information is 80, bearing on the region of interest. The emphasis is on the region of interest. Thus, should one wish to count ^3H in quenched solutions covering only the range of 0-4 KeV, this region must be covered by 80 channels. We would have reached the same conclusions for radiocarbon dating had we used only 80 instead of 450 channels to cover the 0-150 KeV range. Should we wish to monitor background simultaneously with sample count rates, then another 80 channels are required after the ^{14}C end-point up to 250 KeV, and another 80 below the ^{14}C region of interest, to cover 0-6 KeV background (or ^3H) energies. Note that a 1000 channel RPH analyser bearing on the whole 0-2 MeV range does not satisfy the above requirements hence does not satisfy the requirements of the windowless approach to data acquisition.

There is merit to use external storage of full spectra for all samples, standards and backgrounds. For low-level counting, long term stability can thus be verified and reproducibility confirmed.

Disadvantages. A number of disadvantages using an external MCA were observed. They are:

The external MCA, to be effective, needs to be microprocessor controlled and should have computing facilities in its own right. Such equipment is expensive (~ 10k USD).

To accumulate spectra for each sample individually, whilst short term cycling, required multiplexing which we failed to achieve.

Further, there is a difference in dead time between counters and the MCA. Because sampling was random, the data is valid but live-time of counters did not equal live-time of the MCA and we finished with computer outputs evaluating count rates to 1/100th of a second instead of the usual whole minutes. It did not matter, once we got accustomed to it.

The MCA could be controlled externally but the counter could not. It could not stopped or started at preset times or preset precisions by the external computer. Thus the benefit of fine windowless tuning was lost unless we were present and implemented manually the instructions. The LS counter, rather than the computer remained in control. This minimized the effectiveness of the windowless approach.

Suggestions. A picture that emerges is that an ideal LS counter (for any low-level biomedical, high-level, multi-isotope purpose) would have an inbuilt, variable energy span MCA, which through proper handshake protocol would transmit and store data in an external, user friendly micro-computer. It would allow user-accessible programming changes to supplement or modify those supplied by the manufacture of the counter. The counter itself needs only be a noise free linear amplifier of the coincident, lesser pulse, or relative pulse height discrimination type. It need only have a lower limit discriminator, set above electronic noise level. The sample changer would remain the same but sequencing, position selection, counting interval, etc. should be software selectable as should the EHT, etc. If such an ideal counter exists, it is beyond the horizon. We do not see it yet.

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