CHAPTER 42

Statistical Considerations of Very Low Background Count Rates in Liquid Scintillation Spectrometry with Applications to Radiocarbon Dating

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ABSTRACT

The new generation of low-level liquid scintillation spectrophotometers have significantly reduced background count rates. These very low rates require non-Gaussian statistical treatment of data. An LKB Quantulus, employed for radiocarbon dating of samples of between 80 to 300 mg, received no events in 41% of the 780 20-min intervals on background benzene. The distribution of the background events is Poisson. For very small count rates, which are non-background (measurable ^{14}C content) benzene, Poisson statistical treatment of data is preferable to Gaussian treatment.

INTRODUCTION

This chapter is not meant to supercede previous discussions of counting statistics. The authors agree on strict adherence to the recommendations put forth by Stuiver and Polach[1] for use in the general case of radiocarbon date calculation. We would like to submit that very low background count rates may allow for the non-Gaussian treatment of data.

In the last decade, many advances in liquid scintillation (LS) technology have taken place. The incorporation of anticoincidence shielding reduced background count rates.[2-8] Recently, pulse shape discrimination techniques within active shielding[9] and novel energy discrimination—after pulse analysis techniques without active shielding[10,11] have decreased background count rate further.

The level of background noise in LS counters without these techniques can be approximated with Gaussian statistics. This assumes that the background events are stochastic, without a periodic noise from electronics or other source. As background events are reduced over two magnitudes of order, these events approach the predicted Poisson distribution for small numbers of stochastic events.

The statistical interpretation of ^{14}C data collected using decay counting has been discussed,[12-17] and for ^{14}C date calculations, Stuiver and Polach[1] published the guidelines which should be followed by all radiocarbon labs. Yet, when the number of measured events approaches zero, careful consideration of the distribution of the events is required.

The very low background count rates, and subsequent low sample count rates measured with LS counters, can be compared to the statistical analysis of microcontamination in ultraclean environments.[18]

VERY LOW BACKGROUND COUNT RATES

Measurements of very low background count rates (less than 0.1 cpm) have been measured both with gas proportional counting systems[17,19-21] and LS counting.[23,23]

Figure 1 shows background data collected using 0.3 mL vials in an LKB-Wallac 1220 Quantulus LS counter. These data compromise 13 different background samples counted for 60 to 20 min intervals, totaling 1200 min each. A chi-square test of this data confirms that they fit a Poisson distribution. The arithmetic mean of these data is 0.04539 cpm. The distribution of events from 11 separate 0.95 Oxalic acid ^{14}C standard samples (Figure 2), has 4 values which do not fit a Poisson distribution.

TREATMENT OF DATA COLLECTED

A Poisson distribution is quite asymmetrical when the number of counts is near zero, but it quickly approaches a Gaussian distribution by approximation as the count number increases. Tsoulfanidis states that the Gaussian distribution is almost identical with the Poisson distribution at a mean value of 25 counts/time.[24] Therefore, if an unknown were counted, such that an average 25 counts/time were measured, Gaussian statistics would be a close approximation.

Data from Otlet et al., on small gas proportional counting systems, reported background count rates ranging between 21 and 400 counts per day.[20] Increasing the time period is one method of treating the results as a Gaussian distribution. Important information on the stability and performance of an LS counter can be lost if repeat time series data is not collected.

There are many factors which can affect the fit of time series data to an expected Poisson distribution. Periodic external noise, i.e., electronic spikes, ^{222}Rn and ^{220}Rn contamination, and periodic internal noise will alter the fit of measured data. The signal and background will be a Poisson distribution if the LS counter is only recording random events.

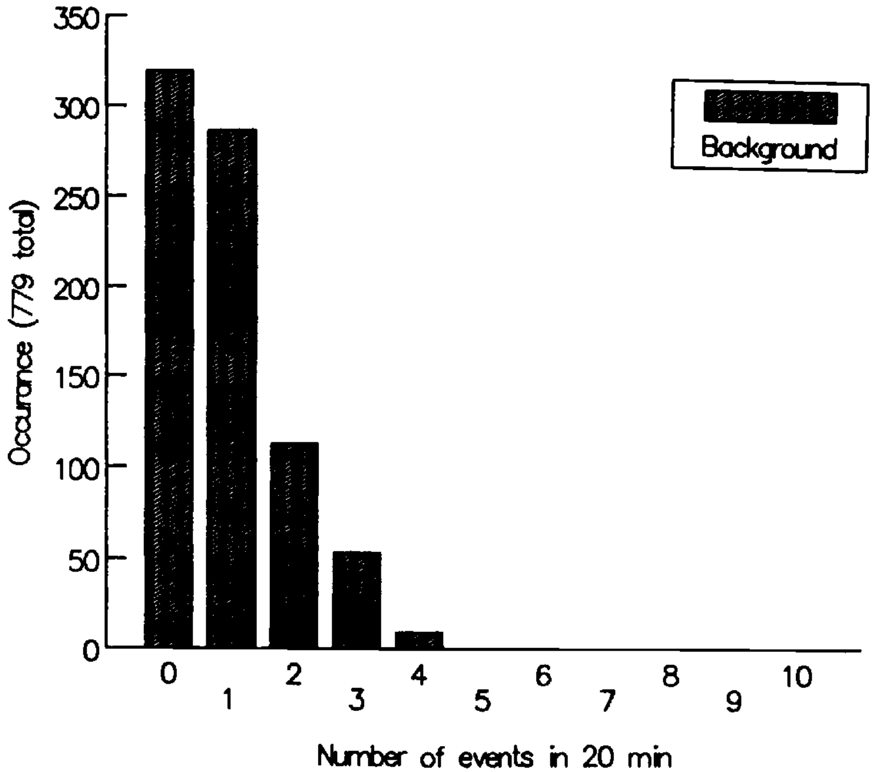

Figure 1. Distribution of background events, 780—20 min intervals.

The background data collected follow a Poisson distribution. There is not any evidence to suggest a nonrandom component of these data. The ^{14}C data have four values (of 660) which do not follow a Poisson distribution. Small amounts of Radon gas were detected in 2 of the 11 samples. We hypothesize that the non-Poisson values are the result of Radon contamination or an artifact of slight sample evaporation during the collection of data.

RADIOCARBON DATING AND POISSON STATISTICS

Questions arise when determining the age of a sample from a small number of events. How certain are the investigators that the events measured are from the original sample material and not from some source of contamination? An assessment of this can be obtained by analyzing background material which undergoes identical treatment along with samples.

A chi-square test of the time series background count rates can alert the investigator to Radon contamination or periodic noise. We also suspect that an analysis of the length of time between background events may be useful in

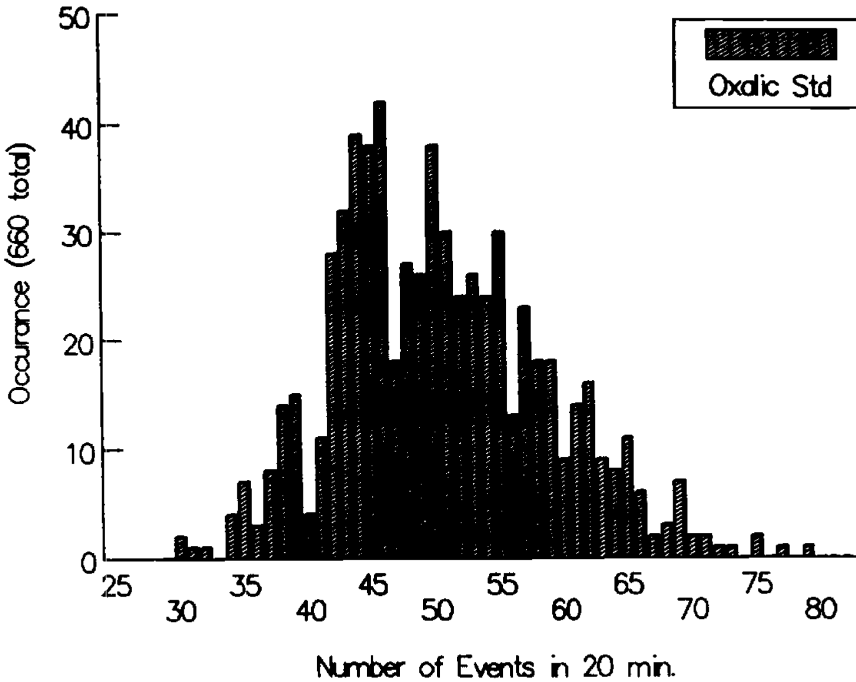

Figure 2. Distribution of 0.95 oxalic acid ^{14}C standard, 660—20 min intervals.

determining a component of periodic noise, and we will investigate this possibility further.

The introduction of systematic errors by lab personnel can be determined by measuring repeat samples of known ^{14}C activity, such as lab standards or known values quality assurance—quality control samples.

The authors would like to suggest the use of Poisson distribution tables like that of Crow and Gardner for determining acceptance criterion when calculating radiocarbon ages.[21] For the background count rate in this study, an average of 55 events is expected during the 1200 min of counting. The two criteria of Stuiver and Polach for 55 events would calculate a "greater than" age using 17 events above background.[1] The Poisson tables of Crow and Gardener would calculate the "greater than" age, 95% confidence, using 16 events.[21] Although one event may be considered trivial, it represents 2% of the total events measured. Assuming that the laboratory researchers have very carefully studied all sources of error within the lab and those associated with the LS equipment, there is a probability that one event can be considered discrete. Rejecting that one event is a loss of information. As the background count rates approach zero, this discrepancy between Gaussian and Poisson distributions becomes a greater portion of the total signal.

It may be best to measure a sample for a period of time sufficient to collect enough events so that this discrepancy is negligible. But, this may not be cost efficient, or LS technology may advance to the point that the discrete nature of single events must be taken into account.

CONCLUSION

The background events measured follow a Poisson distribution. Four of the 660 events for the ^{14}C standard did not fall within the chi-square goodness of fit criteria. These events are hypothesized to be radon contamination or artifacts of evaporation during counting. For very low background count rates, the goodness of fit to a Poisson distribution should be calculated, and an attempt to identify any nonrandom component of the background should be made. Each lab should have a degree of confidence for samples prepared in the lab based on reproduceablility of backgrounds and known activity samples. And finally, by using Poisson statistics, slightly more information may be obtained than when using the recommendations of Stuiver and Polach (1977).[1]

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