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SIMULTANEOUSLY MEASURING ^{14}C AND RADON IN BENZENE DATING SAMPLES

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ABSTRACT. After benzene synthesis, radiocarbon dating samples are usually stored for 3–4 weeks before counting to allow an eventual radon contamination to decay to a negligible level. This paper presents a technique that can minimize, and often eliminate, this delay by using a simple single-phototube liquid scintillation counting system, specifically designed for ^{14}C dating. Radon contamination is assessed by pulses of ^{214}Po (a ^{222}Rn decay product, half-life 0.16 µs), identified through pulse-time analysis. For each ^{214}Po pulse, 0.49 beta particle pulses of ^{214}Pb and ^{214}Bi fall in the ^{14}C counting window, and the ^{214}Po pulses are used to correct the ^{14}C count rate. A ^{14}C sample (count rate 11.6 cpm) was measured continuously for 16 days. It was then doped with radon, which increased the first 24-hr count rate in the ^{14}C channel by 3.8 cpm, and the sample was measured for 27 more days. Radon did not measurably affect the ^{14}C-corrected count rate. Counting a sample for 2 min reveals whether it needs storing. If the radon concentration is low, the sample can be measured immediately without degrading accuracy.

INTRODUCTION

When freshly prepared radiocarbon dating samples are measured, ^{222}Rn can be a serious source of error if using a liquid scintillation (LS) detector or a gas proportional counter (e.g. Hood et al. 1989; Horvatinčić et al. 1995). Although the probability of radon contamination in newly synthesized benzene is generally low, the samples are usually stored for 3–4 weeks before counting, thus allowing radon that may be present to decay to a negligible level. This is a disadvantage when quick results are needed and also when a large number of samples must be stored. This paper presents a simple method that can quickly check whether a sample is contaminated, and in the case of moderate radon concentration, the method allows its immediate measurement. In principle, this method can also be used with conventional 2-tube systems via a software addition.

COUNTING SYSTEM

I use a simple single-phototube LS system, ICELS, specifically designed and made in my laboratory for ^{14}C dating (Theodórsson 2005). It is operated in balanced counting mode. The detector unit has a dome-shaped 3-mL quartz vial, which sits on the top of a vertical 30-mm-diameter phototube. A PC controls the counting and processes the signal. The sample is 3 mL of benzene to which 45 mg of butyl-PBD is added, and the background is 1.5 cpm at 71% ^{14}C counting efficiency.

The amplified pulses are sent in parallel to 4 fixed pulse height discriminators, D1 to D4, which are triggered when an input pulse overrides their threshold voltage, determined by a resistor chain. Only two of the discriminators are of interest here, D1 and D3, which define the lower and upper limit of the ^{14}C balanced counting window, corresponding to beta particle energies of 24.3 and 136 keV (Theodórsson et al. 2003). The computer reads the state of the discriminators every 3rd µs, and when one or more have been activated, one count is added to the number in computer summing registers, N1 to N4, corresponding to the triggered discriminators. This sequence, which takes 11 µs, is the basic ^{14}C counting program operation.

$^{214}Bi/^{214}Po$ PULSE PAIR DETECTION

Table 1 shows the radon series (^{222}Rn) and its 4 short-lived decay products, their half-lives, and the energy of the emitted alpha and beta particles. The 5 radionuclides are in secular equilibrium in the dating samples, i.e. their average disintegration rate is the same. The detection efficiency of the alpha particles is practically 100% and above 95% for the beta particles of ^{214}Pb and ^{214}Bi.

Table 1 The short-lived ^{222}Rn series.

Nuclide		Half-life	Particle, energy (MeV)
^{222}Rn	Rn	3.82 d	α 5.49
^{218}Po	RaA	3.05 min	α 6.00
^{214}Pb	RaB	26.8 min	β 0.65
^{214}Bi	RaC	19.7 min	β 1.75 (77%), 3.17 (23%)
^{214}Po	RaC'	0.16 ms	α 7.69
^{210}Pb	RaD	22 yr	β 0.018

Pulse time analysis (or pulse pair counting), based on the short half-life (0.16 ms) of ^{214}Po, is the key to the high radon sensitivity. Of the ^{214}Po alpha pulses, 98% come within 1.0 ms of the beta pulse of its parent nuclide, ^{214}Bi (Gudjonsson and Theodórsson 2000). When a pulse has been detected and registered, as described above, the computer looks for a short time interval, τ seconds, for a second pulse. If the next pulse arrives within this short period, it is recorded in a Po register. The count rate of these delayed pulses, denoted by $n(Po)$, is mainly due to ^{214}Po, but a small fraction is due to random close pulse pairs (n_{rpp}) expressed as:

$$n_{rpp} = (n_1)^2 \tau \tag{1}$$

where n_1 is the count rate (pulses per second) in N_1. A value of 1.0 ms for τ was selected. At the low count rate of dating samples, n_{rpp} is so small that it can be neglected. This ensures high radon sensitivity.

RESULTS

A ^{14}C benzene sample was first measured continuously for 16 d in counting periods of 6 hr. The mean count rate was 11.61 cpm. The benzene was then doped with ^{222}Rn by slowly bubbling 2 mL of air taken from a vial with a 5-mL aqueous solution of ^{226}Ra. This increased the first 24-hr count rate in the ^{14}C channel by 3.8 cpm. The sample was then counted for 27 d after doping.

Figure 1 shows the 24-hr average values of the count rate in the ^{14}C window, $n(tot)$, as a function of $n(Po)$ during the latter period. As expected, $n(tot)$ increases linearly with $n(Po)$ and can be described by the equation:

$$n(tot) = n(^{14}\mathrm{C}+B) + k\, n(Po) \tag{2}$$

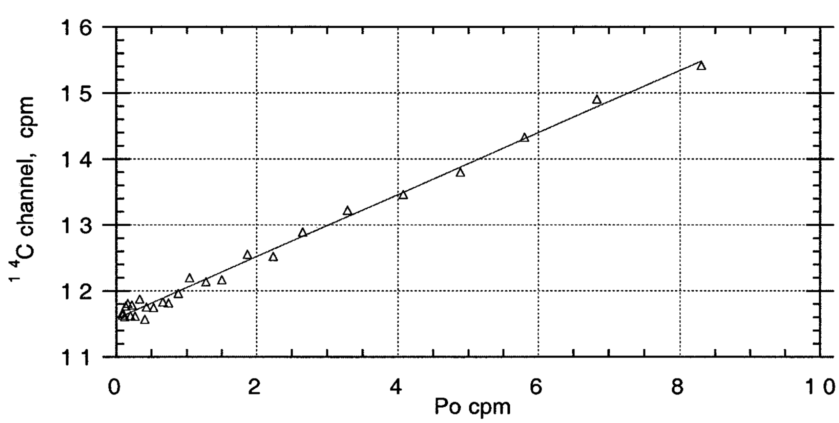

Figure 1 The 24-hr average values of the count rate in the ^{14}C window, $n(tot)$, as a function of $n(Po)$ after radon doping.

where $n(^{14}C+B)$ is the count rate of the sample in the absence of radon due to ^{14}C and background (1.5 cpm). The slope of the line (k) is 0.49, i.e. each pulse in the Po channel corresponds to an increase of 0.49 pulses in the ^{14}C channel due to the beta particles of ^{214}Pb and ^{214}Bi.

Figure 2 shows as a function of time the 24-hr average values of: a) $n(Po)$; b) the slowly declining $n(tot)$; and c) $n(^{14}C+B)$, i.e. $n(tot)$ corrected for the contribution of radon according to Equation 2.

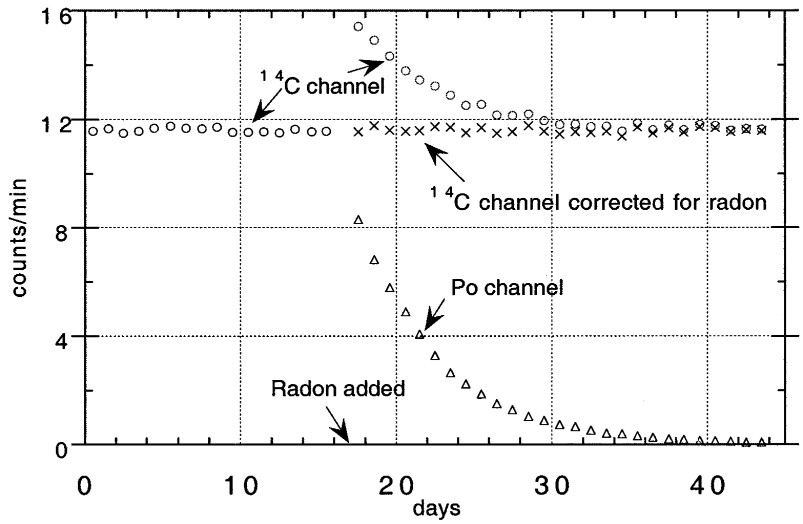

Figure 2 The 24-hr average count rates: a) of ^{214}Po; b) in the ^{14}C window, $n(tot)$; and c) $n(tot)$ corrected for the contribution of radon.

Figure 3 shows the 24-hr and 4-d average count rate in the ^{14}C channel before radon doping and the corrected count rate after doping, with theoretical error bars. The standard deviation of measured average values is in both cases only slightly larger than the calculated values. The average ^{14}C radon corrected count rate the first days after doping shows no significant deviation from the average value before doping (Figure 3). The results of these measurements, average values, and the standard deviation (σ), measured and calculated, are given in Table 2.

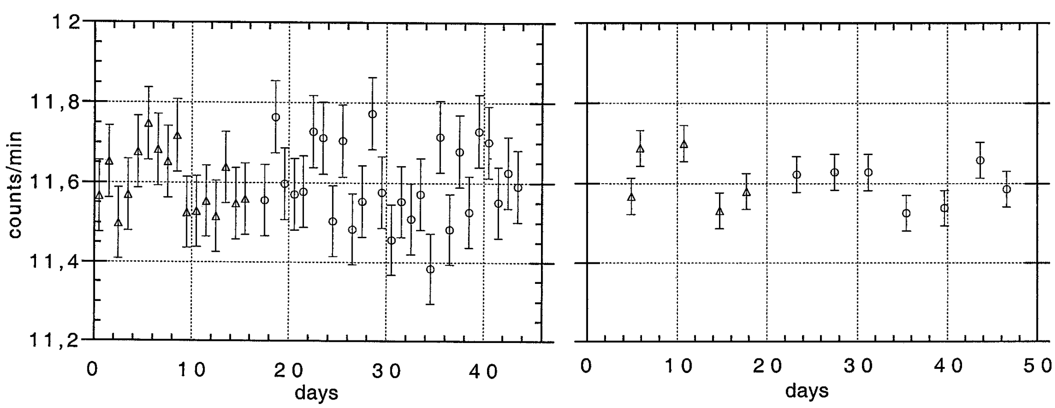

Figure 3 The 24-hr and 4-d average values of count rate in the ^{14}C window, corrected for radon. Error bars are calculated from statistical deviations. Triangles: without radon; circles: after radon doping.

Table 2 Average values and statistical deviations (σ).

	Average cpm	σ measured cpm	σ calculated cpm
Before radon doping			
24-hr average	11.61	0.078	0.090
4-d average	11.61	0.059	0.044
After radon doping			
24-hr average	11.60	0.10	0.09
4-d average	11.60	0.050	0.044

From these results, one can conclude that an initial 2-cpm contribution from radon in the ^{14}C window can safely be corrected without degrading the measurement accuracy. This radon concentration initially gives 4 cpm in the Po-channel, which can be checked with sufficient confidence by counting a sample for 2 min, as the background in this channel is negligible.

DISCUSSION

This paper presents a technique in which the contribution of radon contamination in dating samples can be corrected for by counting ^{214}Po pulses, identified through time analysis. A counting time of 2 min is sufficient to know whether a sample needs to be stored due to radon contamination. In the case of a small amount of contamination, samples can be counted immediately. The net ^{14}C count rate can then be determined with nearly the same precision as in the absence of contamination by applying an appropriate correction, determined from the count rate of ^{214}Po, for the contribution of the radon series to the pulses registered in the ^{14}C counting window.

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