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Optimization of simultaneous tritium–radiocarbon internal gas proportional counting

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ABSTRACT

Specific environmental applications can benefit from dual tritium and radiocarbon measurements in a single compound. Assuming typical environmental levels, it is often the low tritium activity relative to the higher radiocarbon activity that limits the dual measurement. In this paper, we explore the parameter space for a combined tritium and radiocarbon measurement using a natural methane sample mixed with an argon fill gas in low-background proportional counters of a specific design. We present an optimized methane percentage, detector fill pressure, and analysis energy windows to maximize measurement sensitivity while minimizing count time. The final optimized method uses a 9-atm fill of P35 (35% methane, 65% argon), and a tritium analysis window from 1.5 to 10.3 keV, which stops short of the tritium beta decay endpoint energy of 18.6 keV. This method optimizes tritium-counting efficiency while minimizing radiocarbon beta-decay interference.

1. Introduction

1.1. Tritium and Radiocarbon Backgrounds

Both tritium (^{3}H) and radiocarbon (^{14}C) are extensively used for age dating in a wide variety of circumstances. These two standard radioisotopes span the period of 10's to 1000's of years: tritium with a half-life of 12.32 years and radiocarbon with a half-life of 5730 years. Both tritium and radiocarbon undergo beta decay; tritium is a low-energy beta emitter with an endpoint of 18.6 keV [25] whereas radiocarbon has an endpoint energy of 156.5 keV [5].

Tritium concentrations are typically reported in tritium units (TU) where 1 TU indicates one tritium atom per 10^{18} hydrogen atoms. This corresponds to 0.015 disintegrations per day per standard cubic centimeter of methane [10]. Before atmospheric nuclear testing began in 1945, tritium concentration levels in precipitation were around a few TUs from natural processes, e.g., cosmic bombardment of atmospheric nitrogen. Atmospheric tritium concentrations peaked in 1963 at thousands of TU, depending on the geographic location (the majority was injected into the northern stratosphere). After the last atmospheric nuclear test in 1980, atmospheric tritium concentrations have been steadily decreasing and have now returned to nearly pre-test levels [20,26].

Radiocarbon concentrations are typically [22] reported in *percent Modern Carbon* (pMC), defined relative to an oxalic acid standard with a reference value of 95 percent of the activity in AD 1950, such that 100 pMC is 10.5 disintegrations per day per standard cc of methane [17,23]. Before 1945, biosphere radiocarbon fluctuated around 100 pMC. At the peak of atmospheric nuclear testing, the atmospheric ^{14}C concentration was roughly double (200 pMC) that of pre-nuclear test years [13]. After the end of above-ground testing, radiocarbon concentrations have fallen to nearly pre-nuclear-age levels [15].

1.2. Tritium and Radiocarbon Measurement Techniques

There are multiple measurement techniques to count tritium including liquid scintillation counting (LSC), gas proportional counting (PC), or mass spectrometry; each method offers its own advantages [24]. This paper focuses on gas proportional counting to measure tritium, which offers the main advantages of high counting efficiency and low backgrounds. This method lends itself best to samples that are small in size (mL) or contain low concentrations of tritium. Given the high efficiency of this counting method, nearly every decaying atom of tritium can be counted in the active gas volume, so even very small samples can be

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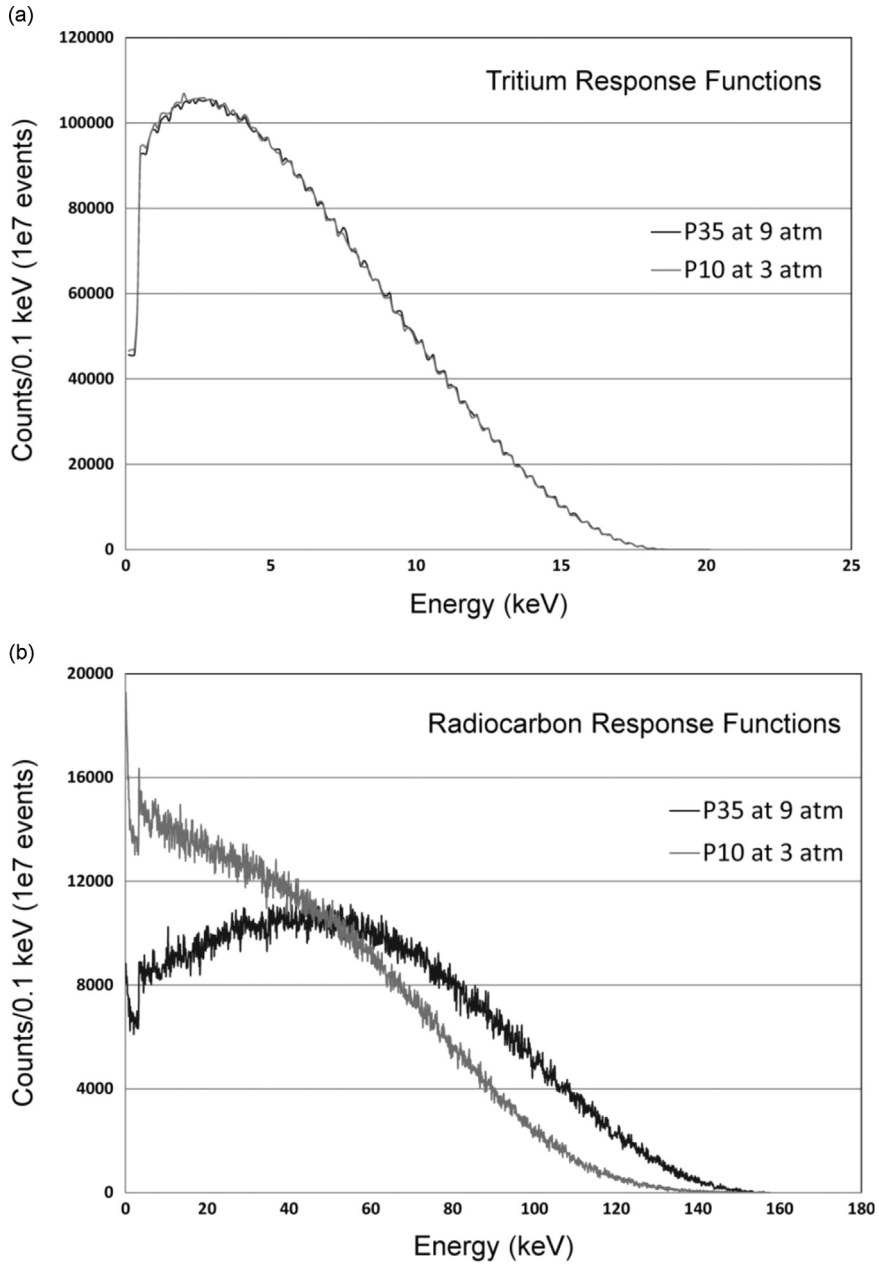

Fig. 1. EGS4 Monte Carlo results for (a) tritium and (b) radiocarbon beta decays. Two gas fills are simulated: P10 at 3 atm and P35 at 9 atm.

measured. Natural methane samples are the most simple to count via PC since they can be loaded directly into counters with minimal chemistry or sample preparation.

There are similar techniques used to measure radiocarbon: LSC, gas PC, and accelerator mass spectrometry (AMS). For radiocarbon analysis, AMS is the acknowledged leader in ^{14}C-dating [8]. However, proportional counters and LSC are also able to do ^{14}C measurements with less precision.

There exists a class of samples where the dual isotope measurement for ^{14}C/T is more desirable than preparing the sample for alternative methods. Specifically, natural methane samples that are easily loaded into a proportional counter without complex sample preparation chemistry. This class of samples could include swamp or marsh gases, dissolved methane from freshwater lakes, and atmospheric methane.

This paper concerns a dual tritium–radiocarbon measurement using ultra-low-background gas proportional counters that were developed at Pacific Northwest National Laboratory (PNNL) [1]. These proportional counters have a design volume of 100 cm^3 (STP) and are operable up to a pressure of 10 atm. Low backgrounds are achieved with a combination of ultra-pure materials [11], active and passive shielding (Seifert 2011), and operation in a shallow underground laboratory [3]. In this study, candidate gas fills are restricted to mixtures of argon and methane. PXX denotes such a mixture with XX% methane, e.g., P10 is 10% methane with the balance (90%) being argon. The methane component (i.e. the sample) could come from a natural methane source (most simple option) or could be prepared from an organic or water precursor, which requires a greater level of effort [18].

The simultaneous measurement of tritium and radiocarbon has been previously demonstrated [19], but this work was performed during a time of much higher atmospheric tritium content. We revisit this topic in light of the challenges now associated with a dual measurement decades after atmospheric nuclear testing has ceased and tritium and radiocarbon components are significantly diminished. It is necessary to optimize detector parameters in order