Spectral Content in ^{3}H and ^{14}C Decays: A Review of Five Experiments

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Abstract

We conduct a generalized spectral analysis of previously published data to re-examine new reports of annual and monthly periodicities in the decay of ^{3}H and ^{14}C. We find no common spectral content in two pairs of simultaneously measured ^{3}H and ^{14}C samples, suggesting fluctuations over the nearly nine year experiment are systematic effects rather than evidence of solar influence on decay rates. Direct comparisons to three other ^{3}H experiments with anomalous results also suggest the presence of systematic effects rather than the appearance of new physics.

1. Introduction

In 2001, E. Falkenberg reported the results of an experiment purporting to show an annual variation in the β- decay of ^3H.[1] Falkenberg noted this variation to be roughly in phase with the changing distance between the Earth and Sun ($1/R_{Earth\text{-}Sun}$) and speculated that such variations could be driven by an unknown interaction with solar neutrinos. These claims bear resemblance to recent reports of periodic variations in ^3H decays by Veprev & Murostev[2] and Lobashev et al.[3] as well as in other isotopes reported by Jenkins et al.[4,5] Like Falkenberg, these works all suggest changes in the Earth's solar neutrino flux may be responsible for the observed variations.

Although Refs [1-3] propose similar phenomenology to explain anomalous ^3H observations, direct comparisons of the data are problematic. Unlike Falkenberg, who only analyzed the annual oscillation, Veprev & Murostev report the presence of diurnal and ~30 day variations but did not acquire enough data to consider an annual periodicity. Conversely, Lobashev et al. reports only a ~6 month variation, but did not sample frequently enough to investigate diurnal or ~30 day variations.

While Jenkins et al. have not reported specifically on ^3H, their collective work describes a number of common periodicities (including ~30 day, ~6 month, and annual variations) in several isotopes, though the presence of diurnal variations has not been observed. Moreover, the analyses conducted by Jenkins et al. rule out models which propose simple $1/R_{Earth\text{-}Sun}$ variations as the sole casual factor in their observations.[6]

Owing to the importance of ^3H and similar isotopes as calibration standards as well as a growing recognition of the importance of spectral content in making precise measurements of half-lives,[7] it is surprising that broader reviews of previously published data remain uncommon. We believe an ideal review of such data would necessarily require comparisons of high quality measurements acquired on different types of equipment across a common time period.

In this work, we search for periodic variations in ^3H and ^{14}C decay rates from four datasets acquired by Jaubert et al. in the course of a nine-year study of quality control techniques for Liquid Scintillation Counters (LSCs).[8] Although the measurement period only partially overlaps with the data presented in Refs [1-3], the data were independently and simultaneously acquired on multiple LSCs. Additionally, we also present a reanalysis of Falkenberg's data[1] for unreported spectral content, specifically looking for ~30 and ~60 day variations similar to those identified in Ref [2-3].

2. Description of Data Analyzed

Among the LSCs evaluated by Jaubert et al.[8] were two custom designed detectors as well as three commercial detectors (a Wallac 1414 Guardian and a Wallac 1220 Quantulus, referred to as "Guardian" and "Quantulus" LSCs). For the commercial LSCs, a set of calibrated ^3H and ^{14}C reference samples were used as comparisons against manufacturer performance specifications. In the course of running these diagnostics, the authors compiled robust pairs of ^3H (7.2 years) and ^{14}C (8.4 years) measurements. During this time, drifts in the detector responses were noted. These drifts were not always monotonic, raising the possibility that the authors were observing periodic oscillations in the decay rate of ^3H and ^{14}C. A representative sample of these data are shown in Figure 1, with plots for all data fully reproduced from Ref [8].

In contrast to the use of a scintillating counter system with calibrated samples, the Falkenberg ^{3}H data was acquired over 1.5 years by monitoring the photocurrent of an unknown "phosphorescent material containing tritium" with an unspecified photodiode arrangement.[1] The observed photocurrent is attributed to the combined result of tritium decay, the degradation of the phosphorescent material, and "other degradation effects." In his analysis, Falkenberg initially presumes a constant ^{3}H half-life and that the degradation of the phosphorescent material is best matched by an "arbitrary" doubly exponential function (χ^2_{red} = 3.40, 57 d.o.f.). These degradation effects are estimated to constitute > 85% of the observed photocurrent.

While no details about the strip of phosphorescent material (or "other degradation effects") used in the Falkenberg experiment are provided, many types of luminescence decays are characterized by the Becquerel decay law: ~ $(1+Bt)^{-n}$.[9,10] Indeed, modeling the background luminescence data in Ref [1] (as provided in [11]) with a Becquerel-like function (B = 0.0013, n = 0.7) yields a considerable improvement in fit statistics (χ^2_{red} = 2.47, 57 d.o.f.) over the original fit (see Figure 2). As a result, the spectral analysis in the next section is performed on the Becquerel background model instead of that reported by Falkenberg for the residuals.

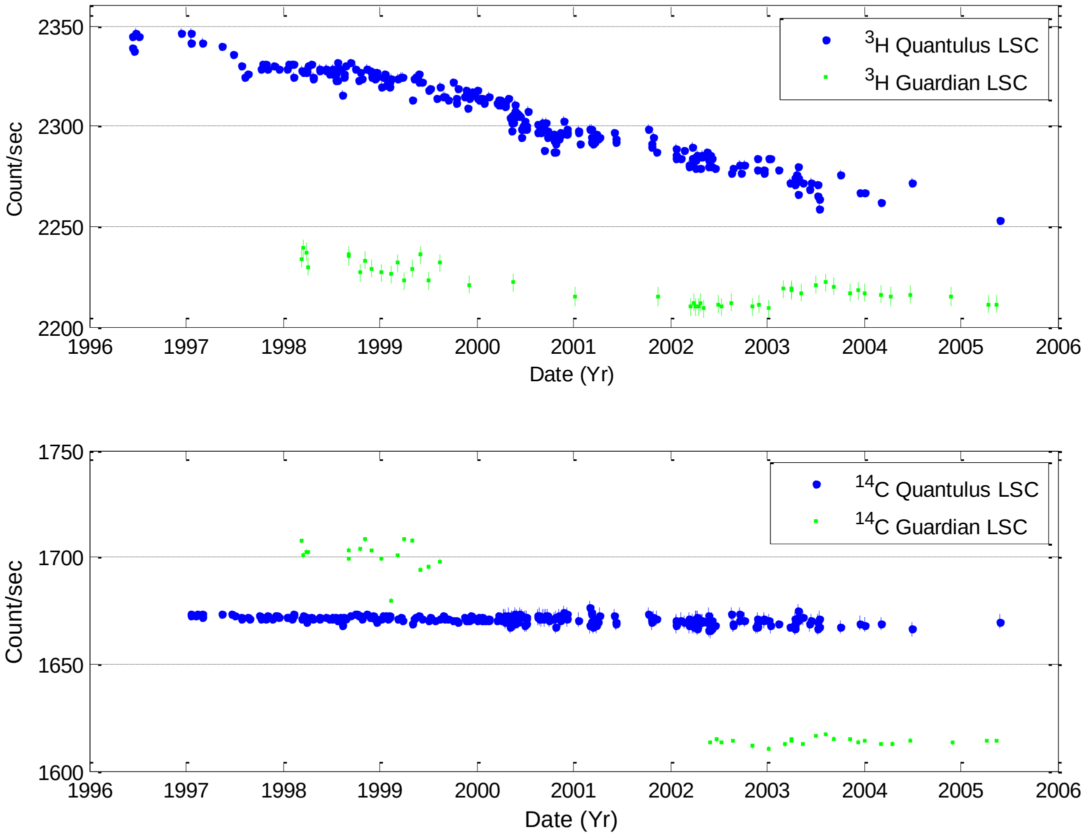

Figure 1. Sample count rates of ^{3}H (top) and ^{14}C (bottom) calibration standards as measured on the Wallac and Quantulus LSCs.[8]

3. Spectral Analysis Method

Using ^{3}H and ^{14}C count rate data from both Quantulus and Guardian LSCs8 and the residuals of the Bacquel-fitted ^{3}H photocurrent from (Figure 2), a spectral analysis was performed in a manner similar to our previous work.[6] In particular, a Lomb-Scargle analysis is first performed to probe for statistically significant frequency content. Since all datasets posses a non-uniform but not random sample rate, we estimate the upper frequency limit for each set of data using the median Nyquist frequency. This analysis yields a Power Spectral Density (PSD) that can be used to make direct estimates of frequencies and phases for any periodic variations within the data (Figure 3). When the residuals to the data are distributed normally, the standard technique for addressing whether a particular peak in the PSD is deterministic is to estimate the False-Alarm Probability (FAP).[12] In this paper we define a statistically significant peak as one with a FAP of less than 0.05.

Peaks from the Lomb-Scargle analysis are further tested for potential aliasing as well as subjected to additional significance testing with a 10,000 trial “shuffle test”.[13] This methodology was selected as it does not rely on the interpolation or re-sampling of the data nor do the final estimates of significance depend upon an assumed form for the underlying distribution of measurements. We further include the criteria that the power, P(n), of each peak in the actual data exceeds 95% of those formed by 100,000 random shuffles of the data.

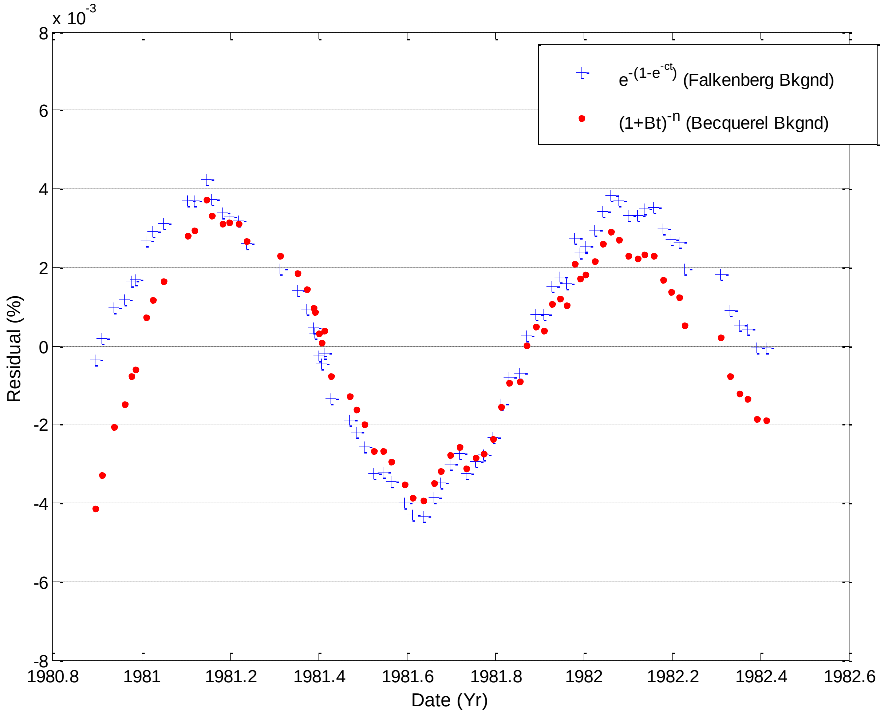

Figure 2. Comparison of ^{3}H residuals formed by background model in Ref [1] ($\boldsymbol{\chi^2_{red} = 3.40}$) to those formed by a Becquerel-like (χ^2_{red}= 2.47) model.

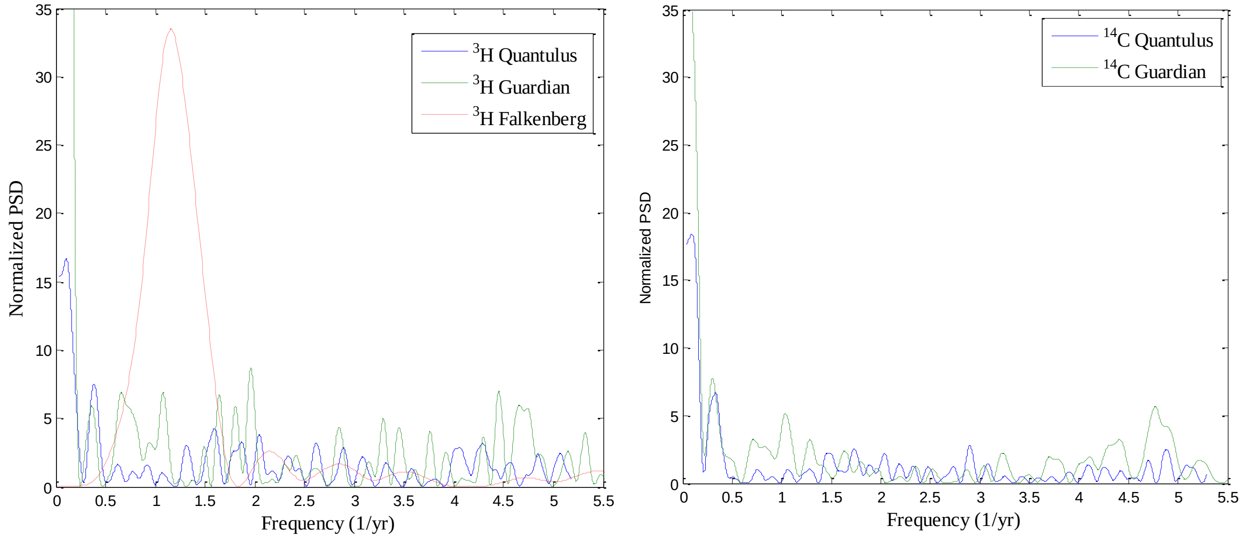

Figure 3. Raw Power Spectral Density (PSD) for the ^{3}H and ^{14}C data presented in Ref [1,8]. When additional considerations such as FAP, aliasing, and significance testing are considered, relatively few peaks are statistically significant. Table 1 summarizes the peaks which are likely to be deterministic components of the data.

4. Analysis Results

The results of the Lomb-Scargle analysis are summarized in Figure 3 and Table 1. Table 1 includes only those peaks considered statistically significant and that do not result from aliasing. The Falkenberg ^{3}H residuals yield only a single peak at 1.15 yr^{-1} which may be identified as the annual periodicity. Fitting a sinusoid with this frequency to the data yields an oscillation amplitude of 0.003% ± 0.002% and a phase shift of 57.4 ± 3.5 days corresponding to approximately 26 February. This is in general agreement with similar findings reported by Falkenberg (f = 1 yr^{-1}, φ = 47 days).[1]

Table 1. Summary of Lomb-Scargle Analyses for the ^{3}H and ^{14}C data in Ref [1,8]. The median Nyquist frequency for each set of data is shown as well as all the shuffle test probability for all statistically significant (shuffle test probability < 0.05, FAP < 0.05) peaks returned from the Lomb-Scargle Analysis.

	Source	***Median Nyquist Freq Limit (yr^{-1})***		***Spectral Analysis Results***	
			freq (yr^{-1})	***P(n)***	***Prob (%)***
3H	Falkenberg	15.4	1.15	33.5	< 10^{-3}
	Guardian	5.3	0.05	130.8	< 10^{-3}
			1.96	8.73	1.3
	Quantulus	5.2	0.39	7.54	0.4
14C	Guardian	5.2		*None*	
	Quantulus	5.2		*none*	

While the Jaubert ^{3}H count rates also show some spectral content, neither the Guardian nor Quantulus LSCs indicate they have a statistically significant annual component. The most prominent peak of 0.05 yr^{-1} in the Guardian LSC is very low frequency content likely related to the overall monotonic drift of the data as shown in Figure 1. The 1.96 yr^{-1} peak in this same ^{3}H sample has a relatively high FAP, and its significance is confirmed by shuffle test in which the power of the actual data exceeds 98.7% of all random shuffles.

Similarly, the actual power of the 0.39 yr^{-1} peak of the Quantulus ^{3}H sample in Table 1 exceeds 99.6% of all random shuffles. There is no observed spectral content in the count rates of ^{14}C as measured in either the Guardian or Quantulus LSC which cannot be directly attributable to aliasing.

The pairs of ^{3}H and ^{14}C count rates presented in Ref [8] do not share any common periodicities which may be considered statistically significant. Additionally, neither ^{3}H sample in Ref [8] possesses spectral content in its decay rate similar to those observed by Falkenberg. Given that the two pairs of sources were measured during the same time period on different equipment under the same conditions, it would be reasonable to conclude that what little spectral content is observed is likely a product of systematic detector effects. Since new physics such as the solar neutrinos previously proposed would be expected to affect the samples and not the detectors, a sample run on two independent detectors should produce the same spectral content on both detectors. As a result, the lack of the reproduction of the frequency content for a given sample run on both the Quantulus and Guardian detectors indicates a detector source of the anomalies.

A further comparison which summarizes our results in relation to Ref [2] & [3] is shown in Table 2. Only the Guardian ^{3}H sample and Lobashev et al. ^{3}H result[3] show agreement with a shared periodicity of ~6 mo (~2 yr^{-1}). However, the Guardian ^{3}H periodicity is relatively weak and not replicated in the Quantulus ^{3}H sample or data provided in Ref [1] and [2]. Tellingly, a ~6 mo periodicity is also the only spectral component which is not addressed by Ref [2] and [3] as having a definitive solar or cosmological counterpart, although more recent work by Sturrock et al. has suggested such ~2 yr^{-1} frequency content is consistent with the solar Rieger periodicity.[14]

In Table 2 we deliberately provide the inclusive dates of the experiments considered. In a model such as the $1/R_{\text{Earth-Sun}}$, the annual spectral content would be expected to be stable over time. However, if a condition existed where the frequency content was not stable then comparisons such as those performed here would only be applicable for dates where the experiments overlap. For the pairs of measurements made by Jaubert this condition would hold but comparisons with the other experiments in Table 2 would not. We highlight this because frequency stability studies of these experiments may indicate the latter.

Table 2. Comparison of five independent ^{3}H datasets which have been examined for frequency content. Yes/No entries correspond to the appearance of spectral content within an approximate period specified at the top of the column. Datasets with sampling intervals which did not permit such a determination are left blank.

^{3}H Dataset	**Experiment Duration**		**Periodicities Observed in ^{3}H Decay Rate**				**Ref**
	Start	Stop	~1 da	~30 da	~6 mo	~1 yr	
Jaubert (Guardian)	Nov-96	Jan-06	-	-	Yes	No	This work
Jaubert (Quantulus)	Mar-98	May-05	-	-	No	No	This work
Falkenberg	Nov-80	May-82	-	No	No	Yes	This work
Lobashev et al	Feb-94	Feb-98	-	-	Yes	No	[3]
Veprev & Murostev	Aug-08	Dec-08	Yes	Yes	No	-	[2]

5. Conclusions

In conclusion, in a model with stable frequency content, the general lack of agreement in all five datasets shown in Table 2 is suggestive of systematic effects unique to each detector rather than evidence supporting a common cosmological source of influence which affects the decay rate of ^{3}H.

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