On the Claim of Modulations in ^{36}Cl Beta Decay and Their Association with Solar Rotation

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Abstract Recently, claims were made by Sturrock *et al.* (*Astropart. Phys.* **42**, 62, 2013), Sturrock, Fischbach, and Scargle (*Solar Phys.* **291**, 3467, 2016; [arXiv](#), 2017) that beta decay can be induced by interaction of the nucleus with solar neutrinos and that cyclic modulations in decay rates are indicative of the dynamics of the solar interior. Transient modulations in residuals from a purely exponential decay curve were observed at frequencies near 11 a^{-1} and 12.7 a^{-1} in repeated activity measurements of a ^{36}Cl source by Alburger, Harbottle, and Norton (*Earth Planet Sci. Lett.* **78**, 168, 1986) at Brookhaven National Laboratory in a period from 1984 to 1985. Sturrock *et al.* have speculatively associated them with rotational influence on the solar neutrino flux. In this work, more accurate ^{36}Cl decay-rate measurements – performed at the Physikalisch-Technische Bundesanstalt Braunschweig in the period 2010–2013 by means of the triple-to-double coincidence ratio measurement technique – are scrutinised. The residuals from an exponential decay curve were analysed by a weighted Lomb–Scargle periodogram. The existence of modulations in the frequency range between 0.2 a^{-1} and 20 a^{-1} could be excluded down to an amplitude of about 0.0016%. The invariability of the ^{36}Cl decay constant contradicts the speculations made about the deep solar interior on the basis of instabilities in former activity measurements.

Keywords Half-life · Decay constant · Radioactivity · Solar cycle · Neutrino

1. Introduction

The validity of the exponential decay law has been questioned in a series of recent articles by authors claiming that periodical oscillations in repeated radioactivity measurements are indicative of variability in the decay constants, whereas others have refuted these claims on metrological and theoretical grounds. More specifically, it was asserted that beta decay is

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induced by interaction of solar or cosmic neutrinos with the nucleus and that modulations in the neutrino flux cause changes in decay rates in the 10^{-3} range. It was speculated that precise measurements of beta decay could be used as an alternative to expensive and elaborate solar neutrino flux monitoring experiments. An overview of the discussion can be found in articles by Pommé *et al.* (2016, 2017a, 2017b, 2017c, 2018), which contradict the hypothesis of variability of decay constants – thus endorsing the validity of the exponential decay law – on the basis of a large suite of accurate decay-rate measurements provided by national radionuclide metrology institutes from all over the world.

In recent work, Sturrock *et al.* (2013), Sturrock, Fischbach, and Scargle (2016, 2017) claim to have observed modulations in beta decay with frequencies of the order of $11\text{--}13\text{ a}^{-1}$, which they speculatively associate with internal solar rotation. In particular, they observed a prominent ‘transient cycle’ of 12.7 a^{-1} (28.76 days period, 0.07% amplitude) in spectrograms formed from countings of ^{36}Cl and ^{32}Si decays in a gas-flow proportional counter at BNL (Alburger, Harbottle, and Norton, 1986) and found them indicative of the synodic rotation rate of the radiative zone of the Sun, since it matches its helioseismologically determined period of 28.7 days (Schou *et al.*, 1998; Komm *et al.*, 2003). However, it is questionable whether all experimental uncertainties were under control in the aforementioned ^{36}Cl and ^{32}Si decay measurements (Pommé, 2015, 2016).

Sturrock, Fischbach, and Scargle (2016, 2017) also linked the 12.7 a^{-1} period with an insignificantly small oscillation in the *Super-Kamiokande* (SK-I) (Yoo *et al.*, 2003) neutrino flux measurements and concluded that modulations in ‘neutrino-induced’ beta decays can provide information about the deep solar interior. In the ^{36}Cl data, an additional cycle appears at an 11 a^{-1} period, which was tentatively associated with rotation of the tachocline between the core and radiative zone. Interestingly, the cycles are called ‘transient’ because they do not persist in time, since they were present in the period 1984–1985, but disappeared in 1986–1987. Sturrock, Fischbach, and Scargle (2016) speculated that the neutrino flux had been modulated by the variable solar magnetic field, in which a primary synodic period of 26.84 days and a secondary period of 28.5 days have been identified (Gough, 2017).

Kossert and Nähle (2014, 2015) demonstrated the invariability of the decay constants for ^{36}Cl and $^{90}\text{Sr}/^{90}\text{Y}$ by triple-to-double coincidence ratio (TDCR) measurements (Kossert *et al.*, 2015), directly rebutting previous claims for annual oscillations ascribed to seasonal changes in the solar neutrino flux due to variations in the Earth–Sun distance (see also Nähle and Kossert, 2015). However, no specific investigation of other frequencies was performed on their ^{36}Cl data set. Pommé *et al.* (2018) scanned decay curves of various nuclides for cyclic modulations in a frequency range between 0.08 a^{-1} and 20 a^{-1} and found no indications of non-exponential behaviour for α , β^{-} , β^{+} , and EC decaying nuclides alike. In this work, a specific analysis of the ^{36}Cl decay-rate measurements is performed in search of the modulations claimed by Sturrock, Fischbach, and Scargle (2016, 2017). They suggested that ‘a reasonable search band for the synodic modulation of beta-decay rates would seem to be $9\text{--}14\text{ a}^{-1}$, corresponding to periods in the range of 26–41 days’.

2. Analysis and Results

2.1. Measurements

The experimental work was described in detail by Kossert and Nähle (2014). Liquid scintillation vials with ^{36}Cl in solution were prepared in December 2009 using 15 mL Ultima

Figure 1 Relative difference from the mean value of measured ^{36}Cl decay rates by Alburger, Harbottle, and Norton (1986) at the BNL (applying 0.1% uncertainty on individual data) (*top*) and by Kossert and Nähle (2014) at the PTB (source #3 by TDCR) (*bottom*). The PTB values are an order of magnitude more precise.

GoldTM scintillation cocktail in 20 mL borosilicate glass vials with low potassium content. Source 3 was measured 66 times between December 2009 and April 2013 in the custom-built TDCR detector at the Physikalisch-Technische Bundesanstalt (PTB) Braunschweig; this has been described by Nähle, Kossert, and Cassette (2010). The activities and the corresponding counting rates of the sources were nearly constant because of the long ^{36}Cl half-life of $302(4) \times 10^3$ a. A small linear correction was applied to the dataset to compensate for increasing colour quenching in the scintillation cocktail with time. Since the TDCR method (Broda, Cassette, and Kossert, 2007; Kossert *et al.*, 2015) is a primary standardisation technique for radioactivity (Pommé, 2007), the PTB decay-rate measurements showed a far higher stability ($\sim 10^{-4}$) than the BNL gas-flow proportional counter measurements ($\sim 10^{-3}$) on which Sturrock *et al.* have based their claims.

2.2. Periodogram

The relative deviations of the measured ^{36}Cl decay rates from their average value are shown in Figure 1, both for the PTB and BNL data sets. The standard deviation of the PTB values ($\sigma = 0.009\%$) is 14 times lower than that of the BNL data ($\sigma = 0.13\%$). The residuals were scanned for cyclic modulations through a weighted Lomb–Scargle periodogram (LSP) (Lomb, 1976; Scargle, 1982), following the procedure described by Pommé *et al.* (2018). The calculated power density takes a value between 0 and 1, reflecting the relative reduction of the χ^2 of the data through the fit of a cycle with a certain angular frequency ω ; $p(\omega) = 1$ if the data match the fit function perfectly. For the PTB data set, the improvement of the χ^2 is less than 12% over a wide frequency range from 0.2 a^{-1} to 20 a^{-1} . The LSP reveals several peaks, none of which is particularly pronounced. The largest peak in the search band, at 13.1 a^{-1} , corresponds to an insignificantly small oscillation of 0.0038 (16) % amplitude (see Section 2.4). This suggests that there are no major cyclic modulations in the ^{36}Cl decay rate.

Figure 2 Weighted Lomb–Scargle periodograms of the 5-day averaged *Super-Kamiokande I* solar neutrino flux data (1996–2001) (top) and ^{36}Cl decay rate data measured at the BNL (middle) and the PTB (bottom). The indicated frequencies correspond to the most apparent peak in the SK-I data (9.43 a^{-1}) and two cycles (11.0 a^{-1} and 12.7 a^{-1}) observed in the BNL count rates.

In Figure 2, a zoom-in of the LSP is shown covering the search band between $9\text{--}14 \text{ a}^{-1}$. A direct comparison is made between the modulations in the SK-I neutrino flux data (top) and the BNL (middle) and PTB (bottom) ^{36}Cl decay-rate residuals. Three particular frequencies are indicated by a dashed line. The 9.43 a^{-1} cycle is the only one that may be statistically significant (Desai and Liu, 2016) in the SK-I neutrino flux data (with a fitted amplitude of 5.5 (17) %), but there is clearly no analogue in the ^{36}Cl decay-rate datasets. Conversely, the 11.0 a^{-1} and 12.7 a^{-1} cycles may have significant amplitudes in the BNL ^{36}Cl decay rates, but there are no matching modulations in the PTB decay data, nor in the SK-I neutrino flux data. In summary, there is no indication of synchronicity among the three data sets.

2.3. Fit of Sinusoidal Function

To assess the magnitude of the modulations, the data were binned in 50 sub-intervals of a cycle, and the amplitude and phase of the best-fitting sinusoidal function were determined. The dates were taken relative to a fixed reference date (1/1/1970), and the fit function was defined as

$$y(t) = A \sin(2\pi(t + a)/T) + C, \quad (1)$$

in which A is the amplitude, t is the elapsed number of days since the reference date, a is the phase shift expressed in days, T is the period of the cycle, and C is a small constant that compensates for a non-zero average value of the grouped residuals.

The binned ^{36}Cl decay-rate data and fitted sinusoids for cycles at frequencies of 9.43 a^{-1} , 11.0 a^{-1} , and 12.7 a^{-1} are shown for the BNL and PTB combined in Figures 3, 4, and 5, respectively. The corresponding amplitudes of the PTB data are 0.0012 (15) %, 0.0019 (16) % and 0.0029 (12) %, all of which are statistically indistinguishable from zero and significantly lower than the 0.07% (local maximum) effects in ^{36}Cl decay reported by Sturrock,

Figure 3 Residuals from exponential decay of ^{36}Cl sources at BNL (*top*) and PTB (*bottom*) binned into 50 subperiods of a 38.75-day cycle (9.43 a^{-1}) and a sinusoid function fitted to the data. The fitted amplitudes A are indicated in the corresponding graphs.

Figure 4 Residuals from exponential decay of ^{36}Cl sources at BNL (*top*) and PTB (*bottom*) binned into 50 subperiods of a 33.20-day cycle (11.0 a^{-1}) and a sinusoid function fitted to the data. The fitted amplitudes A are indicated in the corresponding graphs.

Fischbach, and Scargle (2016, 2017). This reflects the superior stability of the measurements in this work and puts to question why the claim of variability of the ^{36}Cl decay constant has been maintained at present day on the basis of old, unreliable measurements with an unstable instrument, whereas the precise measurements analysed in this work have been available

Figure 5 Residuals from exponential decay of ^{36}Cl sources at BNL (*top*) and PTB (*bottom*) binned into 50 subperiods of a 28.76-day cycle (12.7 a^{-1}) and a sinusoid function fitted to the data. The fitted amplitudes A are indicated in the corresponding graphs.

in literature since 2014 (Kossert and Nähle, 2014). The assertion that the cyclic events are ‘transient’ would be a far-fetched argument.

2.4. Heuristic Uncertainty

As observed with other nuclides (Pommé *et al.*, 2018), a simple heuristic relationship appears to hold between the uncertainty $u(A)$ of the amplitude (expressed in percent) and the relative standard deviation $\sigma(x)/\bar{x}$ of the set of N decay rates (decay-corrected and compensated for annual modulations):

$$u(A) \approx \frac{1.5}{\sqrt{N}} \frac{\sigma(x)}{\bar{x}}. \quad (2)$$

In this case, $N = 66$, and the relative standard deviation of the PTB decay-rate data is $\sigma(x)/\bar{x} = 0.0088\%$, which leads to $u(A) = 0.0016\%$. This value is in good agreement with the calculated uncertainties on the fitted amplitudes. Compensation of the PTB ^{36}Cl data set for the fitted annual oscillation of 0.0040 (16) % amplitude did not significantly alter the amplitudes of the fitted cycles in the search band.

For all decay curves studied in Pommé *et al.* (2018), it was found that the amplitudes of fitted cycles were within a range $u(A)/3 < A < 3u(A)$, which suggests that they result from random variations in the decay-rate measurements. This implies that no statistically significant cycle in the 9–14 a^{-1} search band could be observed in radioactive decay, the case of ^{36}Cl beta decay included.

3. Conclusions

The precise ^{36}Cl decay-rate measurements performed at the PTB were investigated for cyclic deviations from exponential decay at frequencies between 0.2 a^{-1} and 20 a^{-1} . The mea-

sured decay curve shows no evidence of deviations from the exponential-decay law beyond the measurement uncertainty. Periodograms reveal no pronounced oscillations. There is no indication for a cycle at 7.43 a^{-1} , corresponding to a hypothetical cycle in the SK-I neutrino flux measurements between 1996 and 2001, neither are there modulations at 11.0 a^{-1} and 12.7 a^{-1} down to a 0.0016% uncertainty on the amplitude. This contradicts claims by Sturrock *et al.* of ‘transient’ cycles in ^{36}Cl decay rates.

These conclusions are in line with similar research performed on other radionuclides, showing that amplitudes of modulations do not exceed three times their uncertainties. The hypothesis of neutrino-induced decay has been contradicted by experiment for α , β^- , β^+ , and EC decaying nuclides alike. There is no indication that solar neutrinos influence radioactive decay, therefore all speculative solar science deduced from variations in beta-decay rates should be regarded as unsubstantiated.

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