

Review Article

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The Half-life of Radionuclides Measurements and Determination Aspects: Review



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Abstract

This paper challenges the traditional assumption that radioactive decay rates are constant, a cornerstone of nuclear physics. While conventional theory and extensive experiments affirm the stability of decay constants, emerging evidence suggests that external influences, such as solar activity and neutrino emissions, could cause measurable fluctuations. The review critically evaluates the case for constant decay rates, citing experimental confirmations and theoretical coherence. Simultaneously, it highlights compelling anomalies, including variations in reported half-life measurements and correlations between solar flare events and altered decay rates in isotopes like Mn-54, Am-241, and Thorium. Through numerical analyses, the potential ramifications for radiometric dating are illustrated, emphasizing inaccuracies in age estimates when solar activity is overlooked. Additionally, the paper investigates the effects of other neutrino sources, such as nuclear reactors and proton cyclotrons, with experiments near cyclotrons providing robust evidence of neutrino-induced decay rate changes. This study concludes that the presumption of immutable decay rates requires critical reconsideration. The findings call for further investigation to elucidate the mechanisms underlying these phenomena, refine experimental approaches, and develop updated models for radiometric dating. Such advancements will deepen our understanding of nuclear physics and enhance its practical applications.

Keywords: Radioactivity; Decay; Gamma; Beta; Radiation; Nuclear; Cyclotron; Solar flare

Introduction

Decay constants, or half-lives, have been extensively studied and measured using various experimental techniques, and these measurements have consistently yielded reliable results within the uncertainties. These values have been instrumental in numerous applications, including geochronology, radiometric dating, and nuclear power generation. In general, arguments opposing changes in decay constants are often related to the principles of nuclear physics and the well-established theories and experimental evidence supporting them. Here are some common arguments against changes in decay constants:

Conservation of Energy: The decay of radioactive isotopes follows the principles of conservation of energy. Any proposed changes in decay constants would need to account for the energy balance in the decay process and should not violate this fundamental principle.

Experimental Verification: Decay constants have been measured and verified through numerous experiments over the years. These measurements are based on well-established

techniques and principles of nuclear physics. Any proposed changes in decay constants should be supported by robust experimental evidence.

Theoretical Consistency: Any changes in decay constants would need to be consistent with the existing theoretical framework of nuclear physics. Theories such as quantum mechanics and quantum field theory have successfully explained various phenomena in nuclear physics, and any modifications to decay constants should be compatible with these theories.

Natural Constants: Decay constants are considered fundamental constants of nature. Any changes in these constants would have far-reaching implications for other areas of physics and would require extensive reevaluation of various physical theories and models.

Pommé et al arguments opposing changes in decay constants:

In the field of nuclear physics, the stability of decay constants is a fundamental assumption that underpins our understanding of radioactive decay processes. The paper by Pommé and Pelczar [1] titled "Empirical decomposition and error propagation of medium-term instabilities in half-life determinations" delves into the arguments opposing changes in decay constants. The authors contend that extensive experimental evidence and theoretical consistency support the notion of constant decay constants over time. They emphasize the remarkable stability of decay constants across diverse experiments and the lack of compelling evidence to suggest variations. Furthermore, the paper emphasizes the compatibility of current decay constants with well-established theoretical models in nuclear physics. Any proposed modifications to these constants would need to align with the theoretical framework and account for the significant implications they would have on nuclear processes and phenomena. Through this examination, the authors provide a compelling case against alterations in decay constants and advocate for the prevailing understanding based on empirical measurements and theoretical coherence [1].

In the paper "On the claim of modulations in radon decay and their association with solar rotation" [2] Pommé argues the reliability of the findings in long-term decay measurements of gamma radiation from the discharge of radon from a Ra-226 source in a sealed container at the Geological Survey of Israel (GSI) laboratory [3-4]. It is argued that GSI radon measurements are unsuitable for studying decay constants because environmental factors like solar irradiance and rainfall strongly influence the data. Periodic changes in weather conditions can affect radon gas flow leading to daily and seasonal variations in gamma radiation counting. However, these inconsistent counts add up to at least a full day (as indicated therein on Figure 4). It is apparent from the analysis that changes in Rn-222 gamma radiation count rate are influenced solely by external factors in non-isolated measurement systems. However, this is not valid in fully stabilized systems. Also, periodical oscillations of decay rates sum up to the regular decay constant. It is undeniable that such delicate measurements must be conducted under carefully stabilized conditions to collect long-term, comparable counts. Pommé's group publications present strong conclusions that oppose any measured data indicating changes in decay constants. They commonly base their conclusions on the practice of averaging counts over long periods, like a few days. This approach can flatten unique signals

that require explanation. The alternative approach to making extensive decay rate measurements under careful conditions with several detection systems facing several different radioisotopes is presented in this paper.

Reliable findings that point out radioactive decay's constant changes may reveal theoretical gaps and, therefore, can lead to scientific progress.

Methods

Accuracy of Half-Life Data for Long-Lived Radionuclides

In the RANC-2023 conference (3rd International Conference on Radioanalytical and Nuclear Chemistry 7–12 May 2023 / Budapest, Hungary) D. Schumann presented a plenary talk titled: "How accurate are half-lives of long-lived isotopes?". In this fascinating scientific talk, D. Schumann covered a long list of radioisotopes that were measured over the decades in several laboratories. The surprising conclusion of the talk was that, in some cases, the isotope half-life varies from one checkup to another.

In the paper by S. Heinitz, I. Kajan, and D. Schumann, the published uncertainties of half-lives for long-lived isotopes (greater than one year) are presented in Figure 1 [5]. Ten isotopes exhibit 10% to 20% uncertainty in their half-life. Specifically, La-137 has a 34% uncertainty, and Dy-154 has a 50% uncertainty. Table 1 provides a comprehensive list of isotopes, including their half-life data, relative error, and the number of measurements repeated over the years. It is evident that the decay constants for certain radionuclides cannot be precisely determined at this time. The detectors used for count rate measurements, such as NaI(Tl) scintillators for gamma rays and PIPS detectors for alpha particles, are quite common. Therefore, the challenges are not due to limitations in instrumentation. In the paper's conclusions, the authors stated their objective: "Our general intention is to raise awareness within the nuclear data community about the urgent need for reliable decay data and the difficulties associated with conducting new measurements." However, the paper did not explain the reasons behind the discrepancies observed in half-life measurements.

Table 1: Gamma-ray emission sources are used to track the effects of solar flares. The decay processes are listed alongside the gamma emission energies and their corresponding half-lives, as can be found in the literature.

Radioactive Source	Decay Reaction	Main Gamma Energy Line (keV)	Nominal Half-life	Comments
⁵⁴ Mn	$^{54}Mn \rightarrow e_k \rightarrow ^{54}Cr + v$	835	312 days	2 different experiments
⁵⁷ Co	$^{57}Co \rightarrow e_{k} \rightarrow ^{57}Fe + v$	122	272 days	
⁶⁰ Co	$^{60}Co \rightarrow ^{60}Ni + \beta^- + \stackrel{-}{v}$	1173, 1332	5.3 years	
¹³⁷ Cs	$^{137}Cs \rightarrow ^{137m}Ba + \beta^- + \nu$	662	30 years	
²²⁶ Ra	Radium-226 decay chain	Many, up to 2158	1600 years	
natTh	Thorium-232 decay chain	Many, up to 2615	14.5 Gy	
²⁴¹ Am	$^{241}Am \rightarrow ^{237}Np + \alpha$	59.5	432.2 years	
⁴⁰ K	$^{40}K \rightarrow e_k + ^{40}Ar + v$	1460	1.25 Gy	

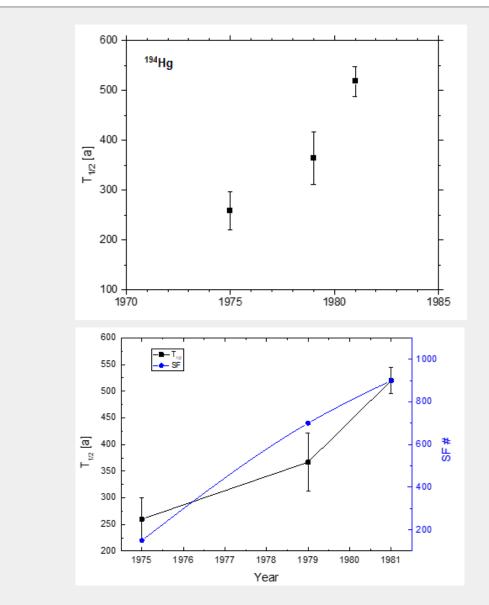


Figure 1: a. Overview of the measured half-life of Hf-194 with the weighted mean as provided by Nuclear Data Sheets (similar to figure in [5]). b. The half-life of Hg-194, as measured [12], is compared to the trend of solar flare occurrences during the same period.

Several research groups are currently working on locating radionuclides with inconsistent half-life measurements over the years [6,7]. As a result, new measurement systems have been established to determine and report the final half-life constant for these radionuclides [8-11]. The half-life measurements of the isotope Hg-194 have shown significant changes over the years. In the Heinitz et al paper [5], Figure 5 illustrates these changes over decades. Specifically, Figure 1a on the right side of the graph displays measurements from 1975 to 1985, during which the half-life of Hg-194 varied from around 250 years to 520 years, showing a significant variation by a factor of 2. The recommended

half-life value of Hg-194 was set to be 450 ± 50 a (uncertainty of 11%). Taking these measured half-life values [12], we performed a comparison of their changes to the 11-year solar cycle and depicted the results in Figure 1b. The graph of solar flare activity clearly shows a trend in half-life change. Our comparison suggests that solar activity and its related particle emissions impact the half-life measurements at the Earth's surface.

Solar Flares

Solar flares are powerful bursts of energy and radiation that originate from the Sun's surface. These events are closely associated with sunspots, which are darker, cooler regions on the Sun's photosphere. Sunspots are caused by intense magnetic activity beneath the Sun's surface. The occurrence of solar flares varies over an approximately 11-year solar cycle. During the solar maximum phase, the Sun's magnetic field is more active, leading to a higher frequency of sunspots and solar flares. Conversely, during the solar minimum phase, the Sun is less active, and fewer sunspots and flares occur. Solar flares release a vast amount of energy across the electromagnetic spectrum, including X-rays, ultraviolet radiation, and visible light. These energetic particles can disrupt Earth's atmosphere, affecting satellite communications, radio transmissions, and power grids. In extreme cases, solar flares can even cause geomagnetic storms that can damage infrastructure and disrupt essential services. The underlying cause of solar activity is the thermonuclear fusion process that takes place within the Sun's core. This process involves the conversion of hydrogen into helium, releasing immense amounts of energy in the form of heat and light. The Sun's magnetic field plays a crucial role in channeling this energy and producing solar flares.

The solar cycle has various effects on space weather and terrestrial phenomena. Increased solar activity during solar maximum can lead to more frequent and intense solar flares, coronal mass ejections (CMEs), and other solar events. These phenomena can impact Earth's magnetosphere and ionosphere and can cause geomagnetic storms, auroras, and disruptions to satellite communications and power grids. During solar flares, the flux of several particles increases significantly. These particles are primarily accelerated and ejected from the Sun's atmosphere during the intense energy release of a solar flare. Some of the particles whose flux increases during solar flares include electrons, protons, neutrons, alpha particles, and some other ions. The study and understanding of the solar cycle have implications for space weather forecasting, satellite operations, and our overall comprehension of the Sun's behavior. Several scientific organizations continuously monitor and study the solar cycle using various instruments and techniques, such as ground-based observatories, space-based telescopes, and spacecraft dedicated to solar observations, in order to better understand this intriguing phenomenon.

The solar X-ray flare phenomenon is thought to be related to the particle transfer loop from the Sun to the corona [13]. The sun's corona plays a crucial role in the solar flare phenomenon. Solar flares are intense bursts of electromagnetic radiation and high-energy particles that originate from the Sun's surface. The corona, the outermost layer of the Sun's atmosphere, is where solar flares occur. The intense magnetic energy stored in the corona is released during a flare, resulting in a sudden and powerful burst of energy and light [14]. The Geostationary Operational

Environmental Satellites (GOES) play a crucial role in observing and measuring solar phenomena, such as solar flares. Positioned in geostationary orbits, GOES satellites continuously monitor specific Earth regions, providing real-time data on various space weather aspects, including X-ray flux from the Sun. Solar flare sizes are categorized as A, B, C, M, and X, and are measured in X-ray flux units of 10⁻⁷ to 10⁻⁴ W.m⁻². The information collected by these satellites is essential for predicting space weather events that can impact communication systems, navigation, and other technologies. The Sun's energy is primarily generated through nuclear fusion, where hydrogen is converted into helium in its core. This process not only produces immense amounts of light and heat but also emits various particles into space. Among these are solar neutrinos, nearly massless particles that provide crucial insights into the Sun's internal processes. High-energy protons and electrons make up a significant part of the solar wind when they reach Earth.

Nuclear Fusion in the Sun

The primary fusion process that powers the Sun and other main-sequence stars is the p-p chain. It involves a series of nuclear reactions where protons combine to form helium nuclei, releasing energy in the process. The p-p chain comprises several stages or branches, and the specific reactions are contingent on the temperature and density conditions within the stellar core. The p-p chain reactions take place in the Sun's core, where temperatures and pressures are extremely high. The core temperature reaches about 15 million degrees Celsius, allowing protons to overcome their electrostatic repulsion and come close enough for the strong nuclear force to bind them together. The p-p chain reaction is fully listed in many textbooks, website sources, and articles. The article by Borexino Collaboration [15] presents the p-p chain reaction and the CNO cycle reactions, along with their solar neutrino flux spectrum (the reaction formulae are colored to show the neutrino source in Figure 1). In the p-p chain three reactions emit neutrinos: pp-v, pep-v, hep-v, and . The total neutrino flux from the p-p chain is about 66×10⁹ cm⁻² s⁻¹ (based on Table 2 in ref. Borexino Collaboration [15]). The CNO cycle adds about 0.7×109 cm⁻² s⁻¹ to the neutrino flux from the Sun [16]. Neutrino oscillation vs distance might contribute to a full mixing of the three neutrino flavors. Hence, only one-third of the flux consists of electronneutrinos, so $\sim 3 \times 10^{10} \, \text{cm}^{-2} \, \text{s}^{-1}$ arrive on Earth. The sun's corona is an interesting and important part of our star; however, it is not a significant source of neutrino emissions. Most solar neutrinos come from the sun's core. During solar flares, the Sun's activity is greatly heightened, leading to an increase in the flux of neutrinos. We note that the undisturbed arrival of a high flux of neutrinos may be linked to changes in radioactive decay rates.

Table 2: Beta(-) emission sources are utilized to monitor the effects of solar flares. The decay processes are listed alongside the maximum emission energies and their corresponding half-lives, as documented in the literature.

Radioactive Source	Decay Reaction	Maximal Energy (keV)	Nominal Half-life
^{nat} Th	Thorium-232 decay chain	2250	14.5 Gy
⁹⁰ Sr- ⁹⁰ Y	$^{90}Sr \rightarrow ^{90}Y + \beta^{-} + \bar{\nu} \rightarrow ^{90}Zr + \beta^{-} + \bar{\nu}$	546, 2826	28.8 years
⁶⁰ Co	$^{60}Co \rightarrow ^{60}Ni + \beta^- + \stackrel{-}{\nu}$	317	5.3 years

Recent Results and Discussion

Solar Flare Effect

The primary finding regarding the impact of solar flares on gamma radiation count rates was published by Jenkins and Fischbach in 2009 [17]. In their paper, the authors highlighted a rapid change in the half-life of Mn-54, which was observed in connection with several strong solar flares. Based on these findings, Fischbach and Jenkins registered their patent for a novel method for the detection of neutrinos [18]. Systematic experiments with gamma radiation sources were conducted for long-term measurements using various radionuclides by Orion's research group. All tested sources are detailed in Table 1. In all these measurements, a NaI(Tl) detector was used; details can be found in the methods section of each reference.

Notably, a careful repetition of the Mn-54 measurement indicated a four-day delay between the solar flare event and the subsequent decrease in radiation [19]. Three different beta (-) sources were used to monitor the effects of solar flares, as outlined in Table 2. For all these measurements, a plastic scintillator was

utilized. More details can be found in the methods section of the referenced paper [20].

Table 3 summarizes the average percentage decrease in radiation counts observed for various radioactive sources during solar flare events. The primary classifications of solar flares that impacted these measurements are listed, along with references to detailed results. Notably, K-40 showed no discernible effect. The percentage of count dips varied depending on the source type. Co-60 showed no effect, while Cs-137 exhibited an almost negligible change in its count rate. The most affected sources were Mn-54, Am-241, and Thorium, with count dips of approximately 0.9%. Table 4 summarizes the average percentage decrease in radiation counts observed for different radioactive sources during solar flare events. The main classifications of solar flares that affected these measurements are provided, along with references to detailed results. The measurements of the beta source primarily indicated smaller responses. In addition, the Co-60 source showed no response with its gamma counts, but it demonstrated a clear response with its beta counts [21].

Table 3: Gamma radiation measurement results: each line consists of the source, the class of solar flare affecting it (both minimum and maximum), the average observed dip, and the reference.

Radioactive Source	Solar Flare Class	Average Counts Dip (%)	Reference
⁵⁴ Mn	X1, X3	-1.0	[17]
⁵⁴ Mn	C1.5, M3.9	-0.6	[19]
⁵⁷ Co	C1.9, M4.4	-0.23	[19]
⁶⁰ Co	M1, X5	0.00	[21]
¹³⁷ Cs	C8.9, M1.4	-0.05	[21]
²²⁶ Ra	C, M3.4	-0.34	[22]
^{nat} Th	B,C, M3.9	-0.93	[23]
²⁴¹ Am	В, С	-0.9	[24]

Table 4: Beta(-) radiation measurement results: each line consists of the source, the class of solar flare affecting it (both minimum and maximum), the average observed dip, and the reference.

Radioactive Source	Solar Flare Class	Average Counts Dip (%)	Reference
^{nat} Th	M3, X2	-0.17	[20]
⁹⁰ Sr- ⁹⁰ Y	M1, X1.6	-0.14	[21]
⁶⁰ Co	M1, X5	-0.18	[21]

Radio-geological Dating Impact

Several widely-used geological radiometric dating methods, including those relying on the U-235, U-238, Th-230, and Th-232 decay chains, may be influenced by solar activity, specifically solar flares. This raises concerns about the accuracy and reliability of these methods. Given the Sun's evolutionary nature and its variable solar activity over time, it is plausible that the half-lives of radioactive isotopes, crucial for dating geological layers, might be affected. A Monte Carlo simulation was employed to quantify the potential impact of intense solar flares on the Th-232 decay constant over a 200-million-year period. The simulation suggests, for example, that a 200-million-year-old geological layer could be as young as 186 million years due to these solar effects. These findings necessitate a reassessment of geological radiometric dating methods. To ensure accurate age determinations, corrections must be applied to account for the influence of solar flares [23]. The decay of Radium-226 influenced by solar activity was simulated using the Monte Carlo technique. By analyzing measured count dips and correlating them with the magnitude and frequency of solar flare events, the simulation recorded a 6% variation in the half-life of Radium-226 compared to the standard value [22]. The evaluation indicated that careful estimations should be considered for several cases of radio-geological dating applications [24].

Nuclear Reactor Effect

Anti-neutrino flux is emitted from a nuclear reactor as a result of neutron decay. An experiment conducted near the High Flux Isotope Reactor at Oak Ridge National Laboratory aimed to expose three gamma radiation sources to anti-neutrinos to examine their effect on radioactive decay [25]. The gamma radiation was measured using NaI(Tl) scintillation spectrometers in one-hour live-time intervals in front of each source: Mn-54, Na-22, and Co-60. The measurements were analyzed during repeated On/Off cycles of the reactor. In this series of experiments, no significant effects were observed.

There are several points to address along this trail:

- **i.** If an anti-neutrino is not identical to a neutrino, differences in their interactions could influence the observed effects. Further investigation into this aspect may be necessary, as anti-neutrino effects might differ or even be nonexistent.
- **ii.** As noted in Table 3, Co-60 gamma detection showed no effect from strong solar flares. Na-22 has not been tested

previously, so its response remains unknown and requires further experimental validation.

- **iii.** The observed 4-5 day delay in the Mn-54 response underscores the need to consider potential time lags in the analysis.
- **iv.** The statistical analysis in this study is thorough and offers a strong basis for assessing the significance of the results. The comprehensive approach ensures that the conclusions are well-supported by the data.

Future research should further explore the differences between neutrinos and antineutrinos, investigate the response of Na-22 to solar flares, and refine the analysis to account for time-dependent effects.

Cyclotron Effect

After several studies, as presented above, that indicated that solar flares can temporarily alter the decay rates of radioactive nuclides, it becomes necessary to study radioactive decay changes in systems positioned near neutrino emission sources. Proton cyclotrons are widely used for medical isotope production due to their consistent and frequent supply to medical imaging institutes. A concise list of radionuclides produced in a medical cyclotron can be found in [26] (see Table 1). Orion's research group carried out a series of experiments using a NaI(Tl) detector, which was positioned next to two cyclotrons (details can be found in the methods section of [26]). Four radioactive sources, each at a certain time, were placed facing the detector over several days, and gamma counts were taken. The operation log of the cyclotron was compared to the measured gamma radiation counts. It is important to note that during the measurements, there were a few solar flares, as the years 2019 to 2020 represented a low point in the solar activity cycle. If solar flares had occurred more frequently, they could have interfered with the effect being examined.

The study utilized 18 MeV proton cyclotrons as a controlled neutrino source to measure changes in the decay rates of four radioisotopes: Rn-222, Thorium, Am-241, and Co-57. It was observed that during periods of increased neutrino flux from the cyclotron, there was a decrease in the gamma radiation count rates for all four radioisotopes. This data is summarized in Table 5. Measurements of gamma radiation influenced by cyclotrons provide strong evidence that neutrinos affect radioactive decay rates. It is highly advised to avoid half-life measurements near proton accelerators due to their neutrino emission.

Table 5: Gamma radiation measurement results affected by cyclotrons [26].

Radioactive Source	Counts Dip Range (%)
⁵⁷ Co	-0.95 to -1.63
²²⁶ Ra	-0.48 to -1.3
natTh	-0.97 to -1.07
²⁴¹ Am	-0.21 to -0.38

Conclusions

This paper has examined the fundamental assumption of constant decay rates in radioactive isotopes, which is a key principle in nuclear physics and has various applications, such as radiometric dating. While the established theoretical framework and numerous experiments support the idea of stable decay constants, there is increasing evidence suggesting that external factors, particularly solar activity and neutrino emissions, may cause measurable variations. This challenges the traditional view and calls for a reevaluation of our understanding of radioactive decay. For instance, electric resistivity, as defined by Ohm's law, is typically considered a constant. However, the resistivity of a device is significantly influenced by temperature and can change as a result of heat variations.

The core arguments for constant decay rates, rooted in conservation laws, experimental verification, and theoretical consistency, remain important. However, discrepancies in reported half-life measurements for certain isotopes, as highlighted by [5], raise concerns about the precision and reliability of existing data. This underscores the need for continued and rigorous experimental efforts to refine these measurements and address the observed inconsistencies. The investigation into the influence of solar flares on decay rates provides compelling evidence for external modulation. Studies by Jenkins and Fischbach [17] and subsequent experiments by Orion's research group have demonstrated correlations between solar flare events and changes in the decay rates of specific isotopes, most notably Mn-54, Am-241, and Thorium. While some isotopes, such as K-40, Co-60, and Cs-137, exhibited minimal or no response, the observed effects on others cannot be dismissed. The varying degrees of response among different isotopes suggest a complex interaction mechanism that warrants further investigation.

The potential impact of these findings on radio-geological dating is significant. If solar activity, which changes over geological timescales, influences decay rates, then radiometric dating methods that depend on these isotopes may produce inaccurate age estimations. Specifically, focusing on Th-232 and Radium-226 illustrates the potential magnitude of these errors. This highlights the need to incorporate adjustments for solar activity into dating models. This has profound implications for our understanding of the Earth's history and the timing of geological events. Beyond solar flares, the paper explored the influence of other neutrino sources on decay rates. Experiments conducted near nuclear reactors, while not showing conclusive effects from anti-neutrinos, raise important questions about the potential differences in

interaction between neutrinos and anti-neutrinos. Furthermore, experiments utilizing proton cyclotrons as controlled neutrino sources provided strong evidence for neutrino-induced changes in decay rates. The observed decreases in gamma radiation counts during periods of increased neutrino flux from the cyclotron further support the hypothesis that neutrino emissions can modulate radioactive decay. This finding has practical implications for experimental design, suggesting that half-life measurements should be conducted away from proton accelerators to minimize potential interference.

In conclusion, this paper presents a compelling case for revisiting the assumption of strictly constant decay rates. The evidence presented, linking solar activity and neutrino emissions to variations in decay rates, calls for a paradigm shift in our understanding of radioactive decay. Future research should focus on elucidating the underlying mechanisms responsible for these observed effects, refining experimental methodologies, and developing corrected models for radiometric dating. This will not only enhance our fundamental understanding of nuclear physics but also improve the accuracy and reliability of various applications that rely on radioactive decay.

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