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Evidence against solar influence on nuclear decay constants

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ABSTRACT

The hypothesis that proximity to the Sun causes variation of decay constants at permille level has been tested and disproved. Repeated activity measurements of mono-radionuclide sources were performed over periods from 200 days up to four decades at 14 laboratories across the globe. Residuals from the exponential nuclear decay curves were inspected for annual oscillations. Systematic deviations from a purely exponential decay curve differ from one data set to another and are attributable to instabilities in the instrumentation and measurement conditions. The most stable activity measurements of alpha, beta-minus, electron capture, and beta-plus decaying sources set an upper limit of 0.0006% to 0.008% to the amplitude of annual oscillations in the decay rate. Oscillations in phase with Earth's orbital distance to the Sun could not be observed within a 10^{-6} to 10^{-5} range of precision. There are also no apparent modulations over periods of weeks or months. Consequently, there is no indication of a natural impediment against sub-permille accuracy in half-life determinations, renormalisation of activity to a distant reference date, application of nuclear dating for archaeology, geo- and cosmochronology, nor in establishing the SI unit becquerel and seeking international equivalence of activity standards.

1. Introduction

The exponential-decay law is one of the most famous laws of physics, already carved in stone since the pioneering work of Ernest Rutherford [1], Maria Skłodowska-Curie [2] and others. It has withstood numerous tests [3–5] demonstrating that the de-

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cay of a radionuclide can be characterised solely by a single decay constant – or equivalently by the half-life – which is invariable in space and time. However, observations of periodic oscillations in measured decay rates of radioactive sources [6–13] have been heavily debated in the last decade [6–25]. Controversy arose at two levels: (i) at the observational level, with experimental data sets showing significant differences in stability of decay rates with time, and (ii) at the interpretational level, either ascribing the observed modulations to instabilities in the detection system, or advocating new physics to explain variability in the decay constants.

As much as the instability claims attract interest as inspiration for new physical theories and applications [14,15], if true they would have major implications on traceability and equivalence in the common measurement system of radioactive substances. Variability of decay constants at permille level would limit the precision by which a half-life value could be assigned to a radionuclide, as well as the accuracy by which the SI-unit becquerel could be established through primary standardisation [26] and international equivalence demonstrated through key comparisons and the Système International de Référence (SIR) [27]. The implications at metrological level would eventually affect science built on the decay laws, from renormalisation of activity to a reference date for nuclear dosimetry to precise nuclear dating for geo- and cosmochronology.

At the heart of this controversy are the metrological difficulties inherent to the measurement of half-lives [28–30]. From a metrological point of view, it is obvious that instruments, electronics, geometry and background may vary due to external influences such as temperature, pressure, humidity and natural or man-made sources of radioactivity. Claims of variability of half-lives on the basis of deviations from an exponential decay curve can only be considered when the instrumental effects have been fully compensated and/or accounted for in the uncertainty budget. Jenkins et al. [9] claim to have done so before proposing their hypothesis that permille sized seasonal variations of decay rates of ^{226}Ra and ^{36}Cl are caused by solar influences on their decay constants [6–8]. Evidence has been collected to demonstrate instabilities in the decay of other radionuclides [10,11] and by means of time-frequency analysis periodicity at shorter and longer term than 1 year have been claimed [11–13]. However, this interpretation is being challenged by the publication of data sets confirming a close adherence to exponential decay with residuals in the 10^{-5} range [16,18,20,21,23].

Authors of both convictions expressed the need for collecting evidence for different radionuclides measured with different detection techniques [7,11,13,18,23]. At national metrology institutes (NMIs) taking responsibility for establishing the unit becquerel, mono-radionuclide sources are kept and regularly measured for standardisation purposes as well as for determining half-lives. In addition, gamma-ray spectrometry laboratories keep records of quality control measurements on their spectrometers which provide useful information on long-term trends in activity measurements of a reference source. In this work, the hypothesis that decay constants vary through solar influence in phase with Earth–Sun orbital distance has been tested through the analysis of a unique collection of activity measurements repeated over periods of 200 days up to four decades at 14 laboratories distributed across the globe.

2. Measurements & analysis

Precise activity measurement series were performed for alpha decay (^{209}Po, ^{226}Ra series, ^{228}Th, ^{230}U, ^{241}Am), beta minus decay (^{3}H, ^{14}C, ^{60}Co, ^{85}Kr, ^{90}Sr, ^{124}Sb, ^{134}Cs, ^{137}Cs), electron capture (^{54}Mn, ^{55}Fe, ^{57}Co, ^{82,85}Sr, ^{109}Cd, ^{133}Ba), a mixture of electron capture and positron decay (^{22}Na, ^{65}Zn, ^{207}Bi), and a mixture of electron capture and beta minus/plus decay (^{152}Eu). More than 60 data sets were collected, some of which were performed over several decades. Some data sets excel in precision, others reveal vulnerability of different measurement techniques to external conditions. Characteristics of the data sets are summarised in Table 1.

The measurement techniques employed are as follows: ionisation current measurements in a re-entrant ionisation chamber (IC) or a hospital calibrator (HIC) [31,32], net area analysis of full-energy γ-ray peaks (and integral spectrum counting) by γ-ray spectrometry with a HPGe detector (HPGe) [33], particle counting in a planar silicon detector in quasi-2π configuration (PIPS) [34], X-ray counting at a small defined solid angle with a gas-filled proportional counter (PC) [35,36], live-timed β–γ anti-coincidence counting (LTAC) [37], triple-to-double coincidence counting with a liquid scintillation vial and three photodetectors (TDCR) [38], liquid scintillation counting (LSC) [38], particle and photon counting in a sandwich CsI(Tl) spectrometer (CsI) [39], internal gas counting (IGC) [40], and α-particle counting at a small defined solid angle with a large planar silicon detector (αDSA) [35,36]. An overview of standardisation techniques and their sources of error can be found in the special issues 44(4) and 52(3) of Metrologia [41,42] and references in [25,28].

Exponential decay curves were fitted to the data and the residuals were inspected for annual modulations. The data sets were first compensated for (1) the presence of occasional outlier values, (2) abrupt systematic changes in the detector response, e.g. due to replacement of the electronics or recalibrations of the instrument, and (3) systematic drift extending over periods of more than 1 year, e.g. due to gas loss from an ionisation chamber, uncompensated count loss through pulse pileup in a spectrometer, activity build-up from decay products in a source, etc. The residuals were binned into 8-day periods of the year and averaged to obtain a reduced set of (maximum) 46 residuals evenly distributed over the calendar year. To the averaged residuals, a sinusoidal shape $A\sin(2\pi(t+a)/365)$ has been fitted in which A is the amplitude, t is the elapsed number of days since New Year, and a is the phase shift expressed in days. The fitted amplitude values can be considered insignificant if they are of comparable magnitude as their estimated standard uncertainty (see Table 1).

3. Discussion

The controversy started with the interpretation [7,8] of $A \approx 0.15\%$ modulations in the decay rate measurements of a sealed ^{226}Ra reference source in an IC at the PTB between 1983 and 1998. The averaged residuals, shown in Fig. 1A, have a sinusoidal shape with amplitude $A = 0.083\ (2)\%$ and phase $a = 59$ days. An explanation through solar influence on the alpha or beta decay constants of nuclides in the ^{226}Ra decay series seems unlikely, since the residuals are out of phase with the annual variation of the inverse square of the Sun–Earth distance, $1/R^2$ (renormalised to 0.15% amplitude in the Figs. 1–2 of this work). The real cause is of instrumental nature, since the modulations were significantly reduced after changing the electrometer of the IC [22,25]. There is a remarkable correlation with average seasonal changes of radon concentration in air ($A = 16\ (2)\%$, $a = 57$ days) measured inside the laboratory from 2010 to 2016, but causality has not been proven.

At other institutes, annual modulations of smaller amplitude and different phase have been observed, which demonstrates the local character of the non-exponential behaviour. The data sets for ^{226}Ra show a different level of instrumental instability, but the most stable ^{226}Ra measurements prove invariability of its decay constant against annual modulations within 0.0025% to 0.005%. An