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On the claim of modulations in radon decay and their association with solar rotation

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ABSTRACT

Claims were made by Sturrock et al. that radioactive 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. They analysed a series of measurements of gamma radiation associated with the emanation and decay of radon in a sealed container at the Geological Survey of Israel (GSI) laboratory. The integral count rates in the NaI detector showed strong variations in time of year and time of day. From time-series analysis, Sturrock et al. claim the presence of small oscillations at frequencies in a range between 7.4 a^{-1} and 12.5 a^{-1}, which they speculatively associated with rotational influence on the solar neutrino flux. In this work, it is argued that the GSI radon measurements are unsuited for studying the variability of decay constants, because the data are strongly influenced by environmental conditions, such as solar irradiance and rainfall. At the JRC and PTB, decay rate measurements of the radon decay chain were performed with ionisation chambers, gamma-ray spectrometers and an alpha spectrometer. No deviation from the exponential-decay law was observed. The existence of cyclic variations in the decay constants is refuted, as well as the concept of measuring solar rotation through radioactive decay.

1. Introduction

In 1900, Rutherford [1] described how activity emanating from thorium decayed away with a constant rate λ such that the ionisation current halved every minute. Today, the 'emanation' can be identified as the noble gas radon-220, and the above paper by Rutherford as the first in which the exponential-decay law is explicitly derived to describe the time dependence of radioactive decay. To the surprise of Rutherford, the exponential-decay law seemed valid only when measuring in a closed system, since *'the movement of the air caused by the opening or closing of a door at the end of the room opposite to where the apparatus is placed, is often sufficient to considerably diminish the rate of discharge'* [1]. More than a century later, measurement conditions are still the most critical issue in a debate concerning the validity of the exponential-decay law.

In the last decade, several papers [2–11] were published discussing repeated measurements of radioactive decay which showed cyclic deviations from exponential decay. Whereas the most obvious explanation is variability in the measurement conditions [12,13], a small group of authors suggested that the decay rate variations were caused by variability of the decay constants [2–11]. They claimed that radioactive decay can be induced by solar and cosmic neutrinos and that oscillations in decay rates are indicative of changes in the solar neutrino flux on Earth due to seasonal variations in Earth-Sun distance or processes in the solar interior. These claims have been refuted on metrological and theoretical grounds. There is overwhelming experimental evidence [14–28] that decay constants are free of monthly or annual oscillations within 0.001%–0.0001%. The most stable measurements of radioactivity [18,19,22–27] performed by primary standardisation [29] laboratories show no evidence to support the idea of neutrino-induced decay.

In spite of all the counterevidence, Sturrock et al. [6,11] still insist on having found compelling evidence of cyclic variations in the decay of radon and its progeny (see Fig. 1 for the decay scheme). They refer to a long-term experiment involving series of measurements at 15-min intervals of radiation associated with the emanation and decay of radon in a sealed container at the Geological Survey of Israel (GSI) laboratory [30] (see Section 2.1). The integral count rates in an enclosed gamma-ray detector show strong variations in time of year and time of day. Whereas most of the variability in the count rate appears to be of environmental origin,

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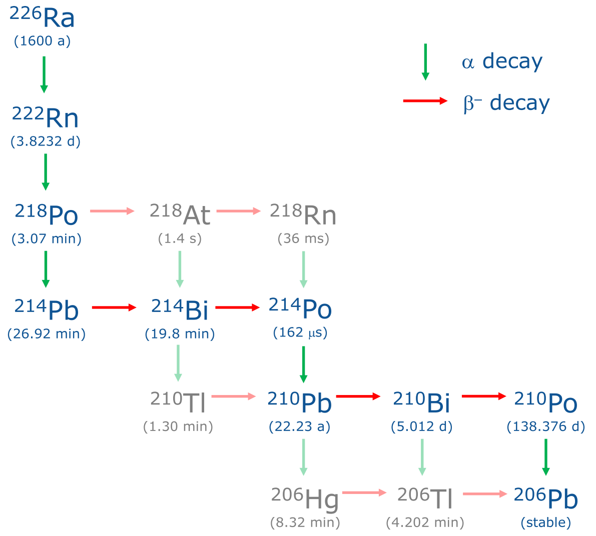

Fig. 1. Decay series of ^{226}Ra/^{222}Rn with decay modes and half-life values.

Sturrock et al. [11] observe the presence of small oscillations (0.35% amplitude [private communication]) at frequencies near 12.7 a^{-1} and 13.7 a^{-1}, which they speculatively associated with synodic and sidereal rotation frequencies of the solar radiative zone. Other frequency pairs, at approximately 11.4 a^{-1} and 12.4 a^{-1} as well as 7.4 a^{-1}, 8.4 a^{-1} and 9.4 a^{-1}, have also been tentatively associated with obliquely rotating parts of the solar interior.

In this work, experimental evidence is provided to illustrate that the radon measurements by Steinitz et al. [30] are highly sensitive to environmental conditions, which are the most likely cause of variation in the apparent decay rates. As counterevidence, measurements of radon performed in more stable laboratory conditions are presented which show no significant oscillations in the decay rates of ^{222}Rn, its progeny or its precursor ^{226}Ra.

2. Radon decay by Steinitz et al

2.1. Experimental conditions

The experimental set-up at GSI has been described by Steinitz et al. [30]. A sealed tank (see Fig. 2) contains at the bottom a layer of ground phosphorite rock with ^{238}U/^{226}Ra, which produces radon (^{222}Rn) and its progeny. The gaseous radon emanates to the upper part of the tank, where a 2″ × 2″ NaI(Tl) detector and two alpha detectors are suspended in air. They detect gamma-rays or alpha particles preferentially emitted by nearby decaying radon atoms and short-lived progeny. The integral count rates in the NaI detector show strong variations in time of year and time of day [6,11,30]. Data from the alpha detectors also show systematic patterns [30], but no solar phenomena were inferred from them.

The set-up is installed in the garden of the GSI research institute, thus being susceptible to various weather conditions. The aim of the experiment was to mimic and study the large temporal variations of radon encountered in air in the geologic environment, which makes it unsuited as a tool to test the (in)variability of decay constants. Whereas a proper experimental design requires a closed system with fixed source-detector geometry and stable measurement conditions, the GSI set-up is exposed to environmental factors which not only affect the operation of the electronic components, but also create variations in transport of radon and its progeny towards the detector area. Both effects can induce potentially large changes in detection efficiency, which hamper conclusions about the variability of the decay constants.

Fig. 2. Front view of the tank with sensors at GSI [11], with indications of the level of the phosphorite rock at the bottom and the positions of the gamma (middle) and two alpha sensors. The covering hut is open to the south, such that the instrument is partially exposed to rainfall driven by southern wind and to light from the Sun and the adjacent building. The external parts of the electronics are covered with large polyethylene sheets (see bottom right picture) for protection.

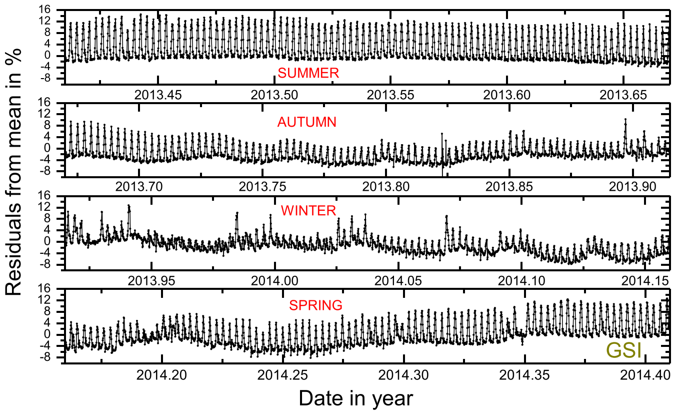

Fig. 3. Residuals from the average count rate in a NaI(Tl) gamma spectrometer at GSI [30] from radon emanation from a layer of rock in a closed vessel as a function of time in a period covering one year from 2013 to 2014.

2.2. Daily and annual oscillations

The measurand in the NaI(Tl) detector is the number of pulses recorded above a threshold corresponding to 50 keV energy. No spectral information is provided, but some similarity can be assumed with the BGO spectrum published by Zafrir et al. [31]: the main contributors to the spectra are gamma-ray emissions following the decay of ^{214}Pb and ^{214}Bi and to a lesser extent ^{226}Ra. At the low-energy side – i.e. near the threshold cut-off energy – there is a relatively high count rate mostly due the contribution from incomplete energy detections through Compton interactions, the 53.23 keV peak from the decay of ^{214}Pb decay, and possibly a small peak at 46.54 keV from ^{210}Pb. This makes the integral count rate sensitive to changes in amplification, threshold setting and electronic noise level.

In Fig. 3, an overview is presented of the residuals of the count rates relative to the mean, for a period of 1 year (2013–2014). Fig. 4 shows detailed plots of typical variations in the measured count rates for 3-day periods in spring, summer, autumn and winter. The