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# STUDY OF THE TIME DEPENDENCE OF RADIOACTIVITY

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Abstract. The activity of a $^{137}$Cs source was measured using a germanium detector installed deep underground in the Gran Sasso Laboratory. In total about 5100 energy spectra, one hour measuring time each, were collected and used to search for time variations of the decay constant with periods from a few hours to 1 year. No signal with amplitude larger than $9.6 \times 10^{-5}$ at 95 % C.L. was detected. These limits are more than one order of magnitude lower than the values on the oscillation amplitude reported in the literature. The same data give a value of $29.96 \pm 0.08$ years for the $^{137}$Cs half life, which is in good agreement with the world mean value of $30.05 \pm 0.08$ years.

Keywords: radioactivity, beta decay, Gran Sasso.

## 1. Introduction

Interest in the time dependence of the radioactive nuclei decay constant has increased strongly in recent times [1], since various experiments began to report evidence of this effect. In particular, the constancy of the activity of a $^{226}$Ra source was measured using an ionization chamber in [2]. An annual modulation of amplitude 0.15 % was observed, with the maximum in February and the minimum in August. The activity of a $^{152}$Eu source was measured using a Ge(Li) detector, and an even larger annual modulation (0.5 %) was detected. Alburger et al. [3] measured the half-life of $^{32}$Si, which is interesting for many applications in Earth Science. Data collected over a period of four years show annual modulation, with an amplitude of about 0.1 %. In [1, 4] the existence of a new and unknown particle interaction was put forward to explain the yearly variation in the activity of radioactive sources [2, 3]. The authors correlate these variations with the Sun-Earth distance. However, the possibility that anti-neutrinos affect $\beta^+$ decay was excluded in a recent reactor experiment [5]. A paper by Jenkins et al. [1] triggered strong interest in the subject. Old and recent data have been analyzed, or reanalyzed, to search for periodic and sporadic variations with time. Some of the measurements and analysis confirm the existence of oscillations [6, 7], whereas others contradict this hypothesis [8–10]. For example, [6] presents the time dependence of the counting rate for $^{60}$Co, $^{90}$Sr and $^{90}$Y sources, measured using Geiger Müller detectors and, for a $^{239}$Pu source, measured with silicon detectors. While beta sources show annual (and monthly) variation with amplitude of about 0.3 %, the count rate from the Pu source is fairly constant. Many authors have called for more dedicated experiments to clarify whether the observed effects are physical or are due to systematic effects not taken into account. We therefore performed a dedicated experiment using a $^{137}$Cs source. Time dependence of the order of 0.2 % with a period of 24 hours and 27 days has already been reported [11] in the decay constant of $^{137}$Cs measured with a germanium detector during a 4-month experiment. The special feature of our experiment is the set-up installed deep underground in the INFN Gran Sasso Underground Laboratory. The laboratory conditions are very favorable: the cosmic ray flux is reduced by a factor of $10^6$ and the neutron flux by a factor of $10^3$ with respect to above ground. As a consequence, we do not have to take possible time variations of these fluxes into account, since their contribution to the counting rate is completely negligible. Moreover, the laboratory temperature is naturally constant, the maximum variation being of a few degrees Celsius in the course of the year.

## 2. The measurements

The set-up is installed in the STELLA low background facility (SubTErranean Low Level Assay) located in the underground laboratories of LNGS. The $^{137}$Cs source, embedded in a plastic disk 1″ in diameter and 1/8″ in thickness, had activity of 3.0 kBq at the beginning of the measurements (June 6, 2011), and it is firmly fixed to the copper end-cap of the germanium detector in order to minimize variations in the relative positions of the source and the detector. As a matter of fact, Monte Carlo simulations indicate that a variation of 1 micron in the source-detector distance would cause a variation of $5 \times 10^{-5}$ in the counting effciency. The Germanium detector, a p-type High Purity Germanium with 96 % relative effciency, is pow-

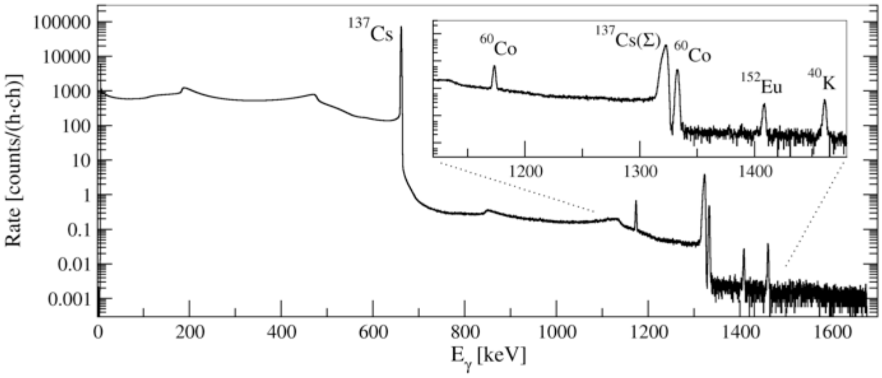


FIGURE 1. Measured γ-ray spectrum of the $^{137}$Cs source after 5124 hours. A weak activity from $^{60}$Co, $^{152}$Eu and $^{40}$K contaminants is observed at high energy (see inset). The $^{40}$K line is also evident in the background spectrum. The peak at 1.323 MeV results from the accidental coincidence of two 661.6 keV γ-lines when two $^{137}$Cs decays occur within the pulse pair resolution of the spectroscopy amplifier.

ered at a nominal bias of 3500 V and is surrounded by at least 5 cm of copper followed by 25 cm of lead to suppress the laboratory gamma ray background. Finally, the shielding and the detector are housed in a polymethylmetacrylate box flushed with nitrogen at slight overpressure, which works as an anti-radon shield.

The signal from the detector pre-amplifier goes first to an Ortec amplifier (mod.120-5F), where it is shaped with 6 µs shaping time, and then to a Multi Channel Analyser (Easy-MCA 8k Ortec, 0.21 keV/channel). The busy and pile-up signals from the amplifier are also sent to the MCA. The pile-up rejection circuit of the amplifier is able to recognize two signals if they are separated in time by at least 0.5 µs. In order to minimize the noise, the whole set-up, detector and electronics modules, is powered through a 3 kVA AC–AC isolation transformer. Finally, temperature and atmospheric pressure are the two important parameters that we have to monitor. Temperature is continuously measured with a Pt100 sensor located in the shielding very close to the Cs source, whereas atmospheric pressure is measured in a nearby hall of the underground laboratory. The temperature at the position of the electronics modules is also continuously monitored, since it can affect the performance of the electronics.

### 2.1. Data taking

Spectra from the MCA are collected every hour (real time provided by the MCA), except while the detector is being refilled with liquid nitrogen. These refilling interruptions occur twice a week and last less than 2 hours. The quality of the measurement is monitored by checking the energy resolution at the Cs line (661.6 keV). Its value, $1.79 \pm 0.02$ keV, is stable, thus proving that the electronic noise does not change with time. This is also confirmed by the counting rate in the lowest MCA channels, below 7 keV, where the noise is dominant and the rate remains constant with time. The position of the Cs peak is also constant, within 0.21 keV, proving the stability of the electronics. finally, we monitored the dead time, provided by the MCA. It changed smoothly from the initial value of 5.10 % to 4.98 % as a consequence of the decreased activity of the Cs source.

### 2.2. Energy spectrum

The spectrum measured with the present set-up is shown in Fig. 1. Modest signals from $^{40}$K, $^{60}$Co and $^{152}$Eu are visible in the energy spectrum above 1 MeV. The $^{40}$K line is also present in the background spectrum, while the $^{60}$Co and $^{152}$Eu activities are related to the source. Their contribution to the total count rate, estimated by Monte Carlo simulation, is $1.7 \times 10^{-2}$ Hz for $^{60}$Co and $2.7 \times 10^{3}$ for $^{152}$Eu and $^{40}$K. The total count rate above the threshold of 7 keV is of about 700 Hz. The intrinsic background, i.e. the shielded detector without the Cs source, was measured over a period of 70 days. Thanks to the underground environment and the detector shielding, the intrinsic background is very low, down to about 40 counts/hour above the 7 keV threshold (0.01 Hz). The signature of the $^{137}$Cs source is given by the outstanding peak at 661.6 keV and by the peak at 1.323 MeV due to the sum of two 661.6 keV gamma rays too close in time to be recognized by the pile-up circuit.

## 3. Results

The data discussed here were continuously collected over 217 days, from June 6, 2011 to January 9, 2012. The activity of the source can be calculated for any interval of time multiple of the hour. In particular, Fig. 2 shows the activity per 4 days as a function of time, i.e. the integral from 7 keV to 1.7 MeV of the MCA spectra collected during 4 days and corrected for the dead time. A total error equal to the linear sum of the statistical uncertainty ($6.4 \times 10^{-5}$ relative) and the fluctuations of the dead time ($2.8 \times 10^{-5}$ relative) was assigned to each data point. The decrease in activity is due to the source decay. The continuous line in Fig. 2 is the exponential fit obtained with the mean

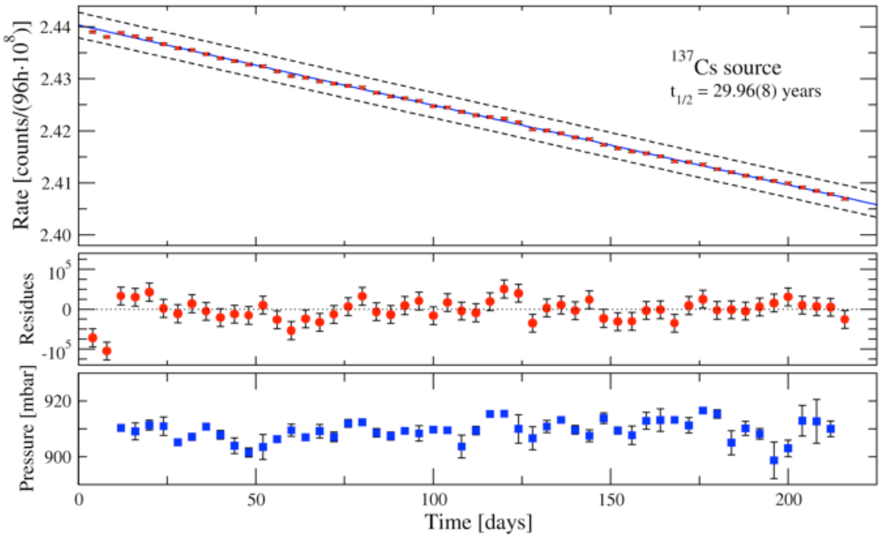


Figure 2. Detected activity of the $^{137}$Cs source. The dead-time corrected data are summed over 96 hours. The first two points correspond to the beginning of data taking, when the set-up was stabilizing, and they are not considered in the analysis. Dotted lines represent a 0.1 % deviation from the exponential trend. Residuals (mid panel) of the measured activity to the exponential fit. Error bars include statistical uncertainties and fluctuations in the measured dead time. Bottom panel: laboratory pressure as function of time.

life which minimizes the Chi Squared: 43.22 years (we take the year of 365.25 days of 86 400 seconds each). The reduced Chi Squared per degree of freedom is 1.02. During the 7-month period of data taking, the temperature at the Cs source increased smoothly by 0.4 K, with a few larger variations up to 0.7 K during one week. There is no effect on the data correlated with these temperature variations. In the same period, the pressure inside the laboratory varies in the range $\pm 10$ mbar. For these pressure changes we can also exclude any significant effect on the rate.

### 3.1. $^{137}$Cs half-life

The $^{137}$Cs half-life has been determined by many authors over a period of more than fifty years (for a critical review of available data, see [12]). Data collected in this measurement allow for a new and precise estimate of the $^{137}$Cs half-life. The data are fitted with an exponential function leaving two parameters free: initial decay rate and half-life. The resulting half-life is $10\,942 \pm 30$ days, to be compared with the recommended value of $10\,976 \pm 30$ days.

### 3.2. Time modulations

We searched for time variations with periods from 6 hours to 400 days. For short periods, up to 40 days, it is appropriate to use the discrete Fourier transform of the hourly data, after subtracting the exponential trend. For a longer period, we performed a Chi Squared analysis of the daily count rate, fitting with a superposition of the exponential decay function (with our mean lifetime estimate of 43.22 years) and a sine function of fixed period. The initial activity, the amplitude of the oscillation and its phase are left free. We scanned all the periods from 40 to 400 days, studying in particular the 1 solar year period. No significant improvement of Chi Squared per degree of freedom was observed in this range. Our analysis excludes any oscillation with amplitude larger than $9.6 \times 10^{-5}$ at 95 % C.L. In particular, for an oscillation period of 1 year the amplitude is $3.1(2.7) \times 10^{-5}$, well compatible with zero; and a limit of $8.5 \times 10^{-5}$ at 95 % C.L. on the maximum allowed amplitude is set independently of the phase [13].

## 4. Conclusions

The half-life of a $^{137}$Cs source has been estimated to be $10942 \pm 30$ days, in agreement with its recommended value. Moreover, from our measurements we can exclude the presence of time variations in the source activity, superimposed on the expected exponential decay, larger than $9.6 \times 10^{-5}$ at 95 % C.L. for oscillation periods in the range from 6 hours to 1 year. In particular, we exclude an oscillation amplitude larger than $8.5 \times 10^{-5}$ at 95 % C.L. correlated to the variation of the Sun–Earth distance. Data taking is now continuing to cover the one-year period.

## Acknowledgements

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## Discussion

**Smedar Bressler** — Is it possible to plan an experiment to modify artificially variations in the decay time of radioactive nuclei?

**Carlo Broggini** — To my knowledge, the only decay which can be modified is electron-capture decay, by changing the pressure at the place where the source is.

**Lawrence Jones** — I wondered whether radioactive decay time might also have a vertical height dependence; i.e. analogous to the gravitational redshift of electromagnetic radiation.

**Carlo Broggini** — I think so, since the effect is due to time dilation in the gravitational field. However, even for a source placed on the surface of the Sun the size of the relative effect, as compared to a source on Earth, would be of about $2 \times 10^{-6}$ only.

**Anatoly Petrukhin** — Did you try to search correlations between your effect and annual cosmic ray variation or any other source of radiation?

**Carlo Broggini** — We do not see any time dependence of the decay constant. In addition, the relative contribution of the background, as compared to the signal, is about $10^{-5}$ in our experiment. Even a 10 % variation of the background would be undetectable for us.