USE OF DAILY DETECTOR VERIFICATION DATA FOR ISOTOPICAL HALF-LIFE DETERMINATION

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Abstract. In this work, the possibility of using data from the daily detector verification routines performed at the Neutron Activation Laboratory of IPEN to determine precise values for the half-lives of ^{57}Co and ^{60}Co was evaluated. For this purpose data from 4 of the laboratory's detectors, some spanning for more than 9 years, were inspected, separated in consistent groups, and then analyzed using a robust least-squares fit procedure in order to determine the half-lives. The results allowed for a discussion on the possibilities and limitations of the use of these data for the determination of half-lives.

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INTRODUCTION

The precise and accurate determination of half-lives of long-lived radionuclides is of great importance, as these are often employed as standard sources for the calibration of detector systems. Thus, as there are small discrepancies in the values found in the literature, new measurements are frequently made with the aim of updating the compiled value, making it more precise and reliable.

There are two ways of determining long half-lives [1]. The first method consists on measuring the specific activity of a radionuclide precisely, what implies in measuring simultaneously both the mass of the sample and its activity – see eq. 1, where $A(t)$ is the sample activity, λ is the isotope decay constant ($= \ln 2/T_{1/2}$) and N_0 is the number of nuclei of the radioactive species in the sample;

$$\frac{A(t)}{N_0} = \lambda \cdot e^{-\lambda \cdot t} \tag{1}$$

The other method consists in following the activity (or the count rate, assuming that the detector system is the same and remains stable during the measurement) of a radioactive sample over a long period of time, so that it is possible to properly fit the radioactive decay (eq. 2).

$$A(t) = A_0 \cdot e^{-\lambda \cdot t} \tag{2}$$

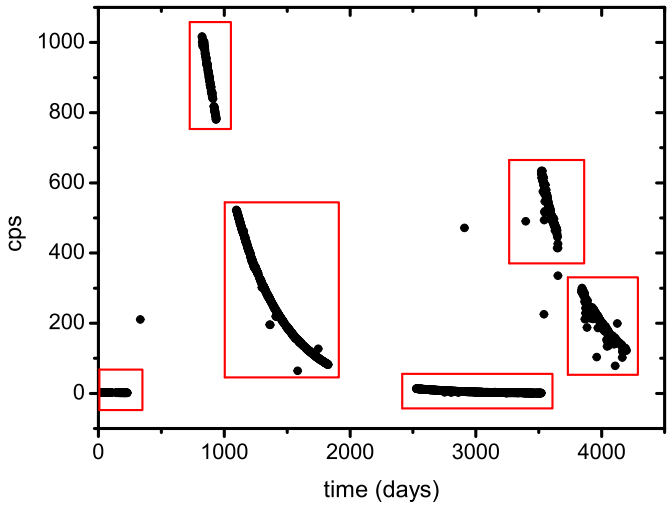

FIGURE 1. Example of the subset separation for one of the detectors and the ^{57}Co decay (worst case scenario).

Whereas the first method requires the precise determination of the concentration of an element as well as the isotopic abundance of the radioactive isotope, the second requires the follow-up of the activity of the same sample in the same detector for a period of some half-lives (and that may mean several years for some isotopes). Nevertheless, radiation detection laboratories often do a daily detector verification by measuring a radioactive source every day (or every few days) in each detector and so, assuming that the detectors remain stable, the results of these measurements could in principle be used for the determination of the half-lives of the isotopes in that source.

EXPERIMENTAL PROCEDURE

To test the reliability and precision of the half-life results obtained from detector verification data, the results of the daily verifications of 4 detectors from the Neutron Activation Analysis laboratory of IPEN-CNEN/SP, some spanning from 1999, were analyzed. In this lab, a mixed $^{57+60}$Co source is measured for 600 s and the peak position, resolution, count rate and uncertainty are registered for both the 122 keV peak from ^{57}Co and the 1332 keV peak from ^{60}Co. These data are manually transcripted to the logbook of the detector system, and later are typed into an electronic spreadsheet; as this procedure is quite error prone, many outliers are found in the data and the obvious ones were manually removed from the datasets. The data for each detector were then separated into smaller consistent subsets, as during this long period of time (more than 10 years) there have been many "configuration changes" (i.e., change of the radioactive source, changes in the detector's operational parameters, and so on) – a worst case scenario is shown in Fig. 1, where the gross outliers had not been removed yet.

The data points from each subset were then fitted using the exponential decay function 2 using a robust fitting procedure [2] that is essentially a combination of the least-squares fitting procedure with the normalized residuals averaging technique [3] and aims

TABLE 1. Half life values obtained for the decay of ^{57}Co using each of the statistical tools compared to the ENSDF tabulated value [4].

Method	Half Life (days)	Z-Score
LRSW	273.2 (5)	2.9
NR	273.40 (11)	13.3
RT	273.77 (10)	17.4
ENDSF [4]	271.74 (6)	

to do a proper outlier treatment while fitting. Briefly, what is done is to iteratively fit the chosen function to the data and, in each iteration, data points with residues larger than 5 ($P < 0.0001\%$) are removed and data points with a residual between 3 and 5 have their uncertainties adjusted so as to have their residual reduced to 3 – this procedure is iterated until convergence is reached (i.e., until the fit χ^2 doesn't change more than 0.01 between consecutive iterations).

Finally, the results obtained for each subset were analysed together using three different statistical tools [3]: Limitation of Relative Statistical Weight (LRSW), Normalized Residuals (NR) and Rajeval Technique (RT).

RESULTS

The results obtained after applying the three statistical tools for ^{57}Co and ^{60}Co are shown in Tables 1 and 2, respectively. These results show that, while for the shorter-lived ^{57}Co the procedure gives precise ($\sim 0.05\%$) results, with an uncertainty close to that found in the literature, the accuracy is not so good, with Z-Scores greater than 10 when compared to the ENSDF compilation value [4]; as for the longer-lived ^{60}Co, the precision ($\sim 1\%$) is very far from the one needed for the intended application. Two possible explanations arise for these behaviours: first, the separation of the data into subsets is a delicate and error-prone task – Fig 2 shows a case where the analysis of the decay curve alone would not suffice to do a proper separation, but the analysis of the uncertainties make it clear that further subdivision is needed –; also, for longer half-lives, the excessive separation into smaller datasets required because of source-detector instabilities does not allow the fitting procedure to establish the half-life with an adequate uncertainty.

CONCLUSIONS

The results presented in this study show that, while in theory the determination of the half-lives of nuclides used in daily detector verification data would be possible, there are many issues that undermine the precision and accuracy of the half-life values obtained. In particular, the data fitting procedure, which is still in development, needs further enhancements in the way it treats outliers. Also, the assumption that the detectors remain stable over time isn't fulfilled by the detector systems used, so the data needs to be split

TABLE 2. Half life values obtained for the decay of ^{60}Co using each of the statistical tools compared to the ENSDF tabulated value [5].

Method	Half Life (days)	Z-Score
LRSW	1808 (24)	-4.9
NR	1982 (23)	2.5
RT	1968 (19)	2.3
ENDSF [5]	1925.3 (3)	

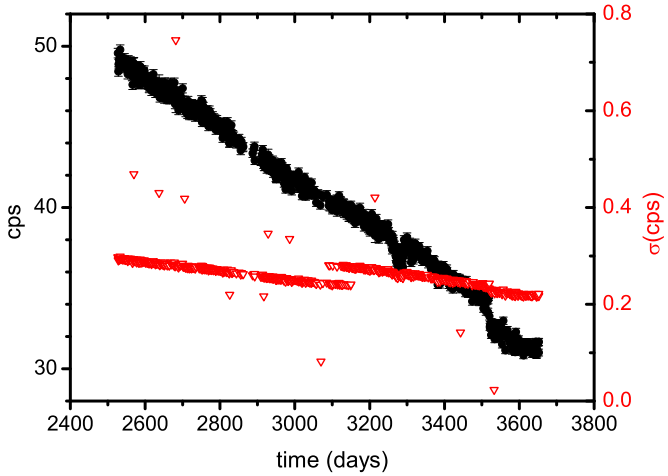

FIGURE 2. Decay curve for one of the subsets of the same detector shown in Fig 1, now for the decay of ^{60}Co – the black dots show the count rate and the red triangles are the associated uncertainties.

in consistent subsets and this procedure needs to be undertanken with great care, as it became clear that in some cases the datasets would need to be split even further, with the inconsistencies visible only when the uncertainties are analysed, too. Finally, for the purpose of determining half-lives of several years (as it is the case of ^{60}Co), only consistent sets spanning for some years should be used, as for shorter subsets the fitting procedure will not determine the half-life with an acceptable uncertainty.

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