Exploratory Assessment of *In Situ* Measurements of Radioactivity for Single Source

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Abstract: Radioactive measurements in the decay process of any radioactive sample can be predicted by radioactive decay law. This predication is based over its average behavior. In actual practice, the radioactive measurements show fluctuations about the average value. For any radioactive sample, there is an exact number which disintegrates in any given unit of time fluctuates around the average value. In counting applications, it is important to estimate this fluctuation because it indicates the repeatability of results of a measurement. This will identify it by using periodogram analysis that depicts the periodicity in the radioactive decay of the given sample of Americium-241. Statistical distributions that the given sample followed with goodness-of-fit tests are examined. Maximum-Likelihood Estimator (MLE) has been used to find the population parameters. The randomness in radioactive decay has been verified by non-parametric method. These statistical analyses are based over the amount of internal fluctuation in the given radioactive source that is consistent with the predictions obtained. These measurements are obtained by measuring the decay of 300 counts per 10 sec. of Americium-241 using a Geiger Muller (GM) Counter in the teaching laboratory, at the Department of Physics, University of Karachi, Karachi, Pakistan.

Keywords: Americium-241, Radioactive measurements, Nuclear Statistics, GM counter.

INTRODUCTION

The application of counting statistic in nuclear measurements is usually used to determine whether a particular counting system is functioning normally. The abnormal amounts of fluctuation can be detected, which could indicate malfunctioning of some portion of the counting system [1].

The two principal Americium isotopes (²⁴¹Am and ²⁴³Am) are both byproducts of the nuclear industry (Schulz, 1976). The Americium-241 emits Alpha and Gamma radiations with energies 5.39-5.55 MeV and 0.026-0.060 MeV, respectively, with a half-life of 458 years, is the daughter product of Plutonium-241 and has relatively high radio toxicity [2].

Americium-241 ($t_{\frac{1}{2}}$ = 433 years) formed by beta decay has more uses than any other isotopes, e.g. in nuclear gauges for numerous applications, static eliminators, smoke detectors and in many other applications. In addition, ²⁴¹Am has been used as a target material in nuclear reactors to produce ²⁴²Am, which has a number of potential uses, including its

decay to Plutonium (²³⁸Pu). The long-range availability of Americium looks attractive since increasing quantities of ²⁴¹Am and ²⁴³Am could be expected from Plutonium recycle either in light-water power reactors or fast breeder reactors. As greater quantities of these isotopes become available at reasonable prices, it can be expected that new and expanded uses will be found for them [3].

The transuranium elements, Americium (Am), Neptunium (Np), Plutonium (Pu) and Curium (Cm), are the products of nuclear weapons tests and nuclear fuel cycle operations particularly from reprocessing plants [2]. The α -ray emitter Americium occurs in fire alarms of older types [4].

There are different ways for the measurement of radioactivity radiations such as Gas-Filled Detectors, Geiger Mueller counters, Proportional counters, Electronic dosimeters, Chemical Dosimeters, Highpurity Germanium (HPGe) detectors, Cadmium Zinc Telluride (CZT) crystal detector, Organic and Inorganic Scintillators, Calorimetry, Cerenkov Detectors, slowneutron detectors and Bubble Detectors. Geiger Mueller (GM) counters are very convenient and reliable radiation monitors, providing both visual and audible responses. A GM tube as a probe is extremely sensitive. It can detect alpha and beta particles when

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fitted with a very thin window. It is efficient for betaparticle counting, but is less efficient for detection of gamma radiation. The GM counters essentially measures individual particles, its readings are therefore usually given in counts per second (IAEA, 1973). Geiger Mueller counters are very expedient and trustworthy radiation monitors, providing both visual and audible responses. They usually come operational with a removable shield that covers a thin window to enable the detection of beta and alpha particles in addition to gamma rays [5].

In this paper, various statistical techniques have been used including descriptive statistics, histogram, best discrete and continuous distributions, P-P plot, periodogram, box plot and goodness of fit. The vital aim of this paper is to assess the in situ measurements of radioactivity for a single source of Americium-241.

METHODOLOGY

The data set of Americium-241 containing 300 observations was obtained from the GM counter. The number of counts recorded each of 10 second interval was clearly not uniform. Therefore, the aim here is to find the most accurate value of the counting rate. The situation is that from a small number of actual observations (300 readings), we are interested to estimate the results of an infinite number of measurements called the parent population. For that purpose, a best-fit distribution was selected that reflect the probability that would be obtained if we were to make an infinite number of measurements. From the method of maximum likelihood, it would be possible to obtain the population parameters [6].

Radioactive decay is in fact random process that obeys the Poisson distribution; however, the true mean is never known and can never be found from a finite number of measurements [7]. The exact statistical information of an experiment is contained in the population distribution. This investigation adopted a technique of MLE to find the population parameters of the given sample [8].

It is desirable when data are obtained during an experiment to be able to determine if the recording system works well or not. A number of tests have been devised for checking the reliability of the results. If the data fail the test, the experimenter should be on the lookout for trouble. Some possible reasons for trouble are the following: Unstable equipment may give inconsistent results, e.g., spurious counts generated by a faulty component of an instrument. A large data point that is far away from the average called the "outlier" may divert the actual shape of the distribution [7].

A discontinuous random variable is defined as the number of times a decay event takes place in a continuous period of time. A non-parametric Runs test is implemented here to verify the randomness in the radioactive decay of the given source [9-10].

There are some special techniques to detect periodicities in a time series and periodogram is one of them. A periodogram analysis is a way for estimating the coefficients of A_i , B_i in such a way that square of the sum of these amplitudes should be significantly greater than zero, otherwise it would possible that we are estimating the coefficients of those terms which are actually not existing in the model equation. Hence, after determining the Fourier coefficients we convert these coefficients into amplitudes and phase angles for each of the frequencies [11-15].

RESULTS AND DISCUSSION

In literature survey, radioactive decay is a random process and follows Poisson distribution, which is one of the discrete distributions. The probability of success for decaying a particular atom at a particular instant has always been very small and that would link this process to that distribution. However, this could be varied if the outliers introduce in the observations either by the malfunctioning of the instrument or by the operator's fault or any other source of error. The superficial inspection of Figures 1 and 3 reveals that the series of radioactive decay followed quite well a Poisson distribution. However, the goodness of fit test Kolmogrove-Simernov (K-S) fit а continuous distribution, called Log-Logistic (three parameters)

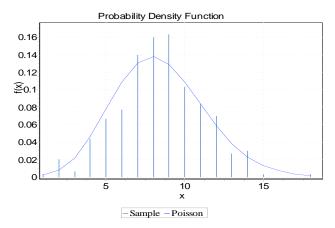


Figure 1: Best discrete distribution followed by radioactive decay of a given sample.

distribution (shown in Figure **2**) at 99% and 98% level of significance, whereas Anderson Darling (AD) accepts it for 95% level of significance. These results are shown in Table **1**.

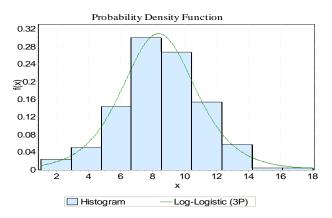


Figure 2: Best continuous distribution followed by radioactive decay of a given sample.

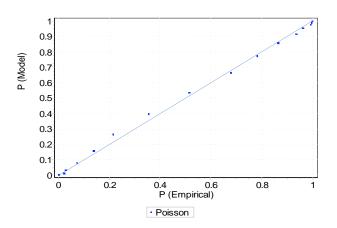


Figure 3: P-P plot examine the Poisson distribution.

Table 1: Details of Goodness of Fit for Log-Logistic (3P) Distribution

Test	Statistic	P-Value
Kolmogorov-Smirnov Test	0.08456	0.0258
Anderson-Darling Test	1.9685	

The number of radioactive atoms that disintegrate per unit time has some average value. It is therefore important to estimate this average value because the exact number that disintegrates per unit time will fluctuate around it. Further, this fluctuation indicates the repeatability of results of a measurement. For that purpose a periodogram has been established and the periodicity is defined with respect to the highest peak depicted by the periodogram of 2 counts per 10 seconds, as shown in Figure **4** and Table **2**.

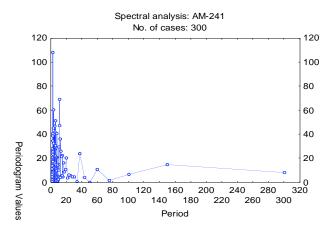


Figure 4: Periodicity in radioactive decay of a given sample.

Table 2: Periodogram Analysis Depicted for the Highest Peak

Variables	Values	
Frequency	0.45	
Period	2.18	
Cosine Coeffs.	-0.62	
Sine Coeffs.	-0.575	
Periodogram Value	107.99	
Density	52.00	

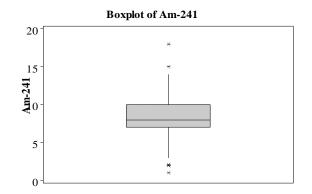


Figure 5: Outliers identification using Box Plot.

Box plot shown in Figure **5** identifies four outliers that could be responsible for deviation of the distribution from its actual shape. These are identified as 18, 15, 2 and 1 counts. A non-parametric Runs test with its configuration shown in Table **3** defines randomness in the given data series after being recognized its p-value of 0.306. The non-parametric test is implemented here because it is a distribution free method and could be reasonable where the exact distribution is ambiguous. The use of the Maximum Likelihood Estimator enables us to estimate the population parameters that are shown in Table 4. This has been notified that they are very close to the sample values. Actually, nuclear disintegrations obey Poisson statistics, but if the number of observed events is large, the Poisson distribution is sufficiently approximated by a normal distribution. Central-Limit Theorem defines that whatever be the shape of the sample, its population always follows a normal distribution. However the standard error increases by a factor of 0.004.

Table 3: Runs Test to Verify the Randomness in the **Radioactive Decay of a Given Sample**

Runs Test	Values
Runs above and below K	8.42667
The observed number of runs	142
The expected number of runs	150.833
Observations above K	145
Observations below K	155
P-value	0.306

Table 4: Sample and Population Statistic

Statistic	Sample Values	Population Values
Sample Size	300	300
Mean	8.42	8.43
Variance	7.32	7.69
Coeff. of Variation	0.321	0.328
Std. Error	0.156	0.160
Skewness	0.049	0.070

CONCLUSIONS

The relevance of statistics to nuclear counting data is obligatory to the accuracy with which measurements are made. The investigation in this communication recognized that the distribution followed by the radioactive decay is other than Poisson. This could happen by the outliers that are one of the sources of error in this account. The important investigation is the periodicity obtains in the radioactive decay of Americium-241 that is equivalent to one of the outlier as identified by the Box plot. This indicates that the

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radioactive decay of this source has some unusual behavior. By using Maximum-Likelihood Estimator (MLE) the population mean and variance are achieved. Runs test authenticates the presence of randomness in the series.

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