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Measuring radioactive half-lives via statistical sampling in practice

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Abstract – The statistical sampling method for the measurement of radioactive decay half-lives exhibits intriguing features such as that the half-life is approximately the median of a distribution closely resembling a Cauchy distribution. Whilst initial theoretical considerations suggested that in certain cases the method could have significant advantages, accurate measurements by statistical sampling have proven difficult, for they require an exercise in non-standard statistical analysis. As a consequence, no half-life measurement using this method has yet been reported and no comparison with traditional methods has ever been made. We used a Monte Carlo approach to address these analysis difficulties, and present the first experimental measurement of a radioisotope half-life (^{211}Pb) by statistical sampling in good agreement with the literature recommended value. Our work also focused on the comparison between statistical sampling and exponential regression analysis, and concluded that exponential regression achieves generally the highest accuracy.

editor's choice

Introduction. – The decay constant λ of radioactive isotopes is a fundamental quantity in radiation metrology, which finds application in nuclear medicine, power generation, nuclear forensics, geochronology, basic nuclear physics and astrophysics. Such constant, or the corresponding half-life $\hat{\tau} = \ln(2)/\lambda$, is widely regarded as independent of all the physical or chemical conditions [1], although variations have been detected in case of electron-capture and internal-conversion decay when the nuclear decay is coupled to the atomic environment [2,3]. In these cases, small changes can be induced by pressure, temperature, or electric fields, which effectively modify the electron density at the nucleus [4]. More drastic changes have been observed with ionized atoms, in which case decay modes possible in neutral atoms may become hindered or forbidden [5].

Despite the apparent simplicity of half-life measurements, half-lives derived from different data sets are often discrepant and for the majority of the radionuclides the spread of experimentally determined half-life values is larger than expected from the claimed accuracy [6,7]. Besides the need to resolve discrepancies, research toward improving half-life measurements may have important consequences in several areas of physics, which involve high-precision half-life measurements. Examples include the study of the coupling between the nuclear decay and the atomic electron cloud [4], and the study of superallowed β-decays, which sets stringent limits on the possible scalar current contribution to the weak interaction (see *e.g.*, ref. [8]). In addition, deviations from the exponential law for the decay of quantum systems are possible and were predicted in conformity to quantum electrodynamics for time intervals very short or very long compared to the mean half-life [9–11]. Experimental evidence of non-exponential decay were reported in atomic and molecular physics for the spontaneous emission of optical photons [11] and quantum tunneling [12]. Claims of nuclear radioactivity exhibiting a periodic decay component, possibly due to geophysical or astrophysical effects, were controversial and later disproved within a precision range of 10^{-6} to 10^{-5} [13,14]. The discovery of non-exponential nuclear decays may have extraordinary implications for example in the case of radioisotope dating.

In the context of studying new data analysis techniques for half-life measurements, this manuscript addresses the technique of statistical sampling for half-life measurements, which was first proposed in ref. [15] where the distribution of half-life estimates from pairs of activity measurements was shown unexpectedly to be a nearly perfect Cauchy distribution centered on the half-life. The probability density function (pdf) of the half-life distribution was determined in ref. [16] where it was also shown how a Cauchy distribution emerges from it. Reference [16] suggested that statistical sampling may have an advantage in the measurement of long-lived isotopes where activity measurements may be difficult to record at regular times. This is because the statistical sampling only deals with frequency of events rather than their time order. In the context of studying deviations from exponential law, ref. [17] pointed out that approaches based on statistical sampling often readily show effects such as periodic fluctuations (including detector instabilities [18]) as peak displacement and non-zero skewness of the half-life distribution. However, due to data analysis difficulties, no half-life measurement using this method have yet been reported. In this manuscript we report a statistical analysis based on a Monte Carlo (MC) approach that allows the application of the method in practice, and a quantitative comparison with the exponential regression analysis.

The statistical sampling method. – The radioactive decay is a random process described by the discrete binomial distribution or the Poisson distribution in the limiting case of large number of atoms. Under this assumption the decay rate (or activity) A as a function of time takes the well-known form

$$A(t) = A(t_0)e^{-\lambda(t-t_0)}, \tag{1}$$

where t_0 is an arbitrary reference time. A typical half-life measurement by radiometric techniques consists of a finite series of n activity measurements $A_{i=1,2...,n}$ each A_i determined from the number of decay in a time interval Δt. Hereafter we assume that the duration of such experiment is $n\Delta t$. The half-life is then estimated fitting data with eq. (1). Alternatively, the statistical sampling method consists in estimating half-life values from any subset of the possible $n(n-1)/2$ pairs of activity measurements accordingly to

$$\tau_{ij} = \frac{t_{ij}\ln 2}{\ln(A_i/A_j)} \qquad (t_{ij} = t_j - t_i > 0). \tag{2}$$

The distribution of these half-life estimates τ (hereafter half-life distribution) was studied in ref. [16], which reported the distribution's pdf in the assumption that activity measurements at different times are independent Poisson variate with high mean count:

$$f(\tau) = \sqrt{\frac{2}{\pi}}\,\frac{\ln 2}{n(n-1)}\sum_{i=1}^{n-1}\sum_{j=i+1}^{n}\frac{\left(\frac{t_{ij}}{\tau^2}\right)\exp\left(\frac{t_{ij}\ln 2}{\tau}\right)}{\sigma_{ij}} \times\exp\left\{-\frac{1}{2\sigma_{ij}^2}\left[\exp\left(\frac{t_{ij}\ln 2}{\tau}\right)-\frac{\mu_i}{\mu_j}\right]^2\right\}, \tag{3}$$

where μ_n is the number of decays recorded in the n-th counting time interval Δt, and

$$\sigma_{ij} = \sqrt{\frac{\mu_i}{\mu_j^2}\left(1+\frac{\mu_i}{\mu_j}\right)}. \tag{4}$$

In ref. [16] it was shown that when $\mu_n = \mu_0 e^{-\lambda t_n}$, and under the conditions:

a) $t_{ij} \ll \tau$ and $t_{ij} \ll \hat{\tau}$,

b) $\mu_0 \gg 1$,

c) $n \gg 1$,

the exact pdf can be approximated by a sum of Cauchy functions of different scale parameters all centered on the true half-life $\hat{\tau}$:

$$f(\tau) \approx \sum_{i=1}^{n-1}\sum_{j=i+1}^{n}\frac{t_{ij}}{\hat{\tau}^2}\,\frac{\exp\left(\frac{t_{ij}\ln 2}{\hat{\tau}}\right)}{1+\frac{\mu_0}{\tau^4}t_{ij}^2(\tau-\hat{\tau})^2}. \tag{5}$$

A further step of approximation, detailed in ref. [16], collapses the double sum of eq. (5) into the single Cauchy distribution:

$$f(\tau) \approx \left\{\pi\gamma\left[1+\left(\frac{\tau-\hat{\tau}}{\gamma}\right)^2\right]\right\}^{-1} \tag{6}$$

with approximate scaling factor γ:

$$\gamma = \frac{6\,\hat{\tau}^2}{n\ln 2\sqrt{\pi\mu_0}}. \tag{7}$$

Equations (3)–(7) provide the theoretical ground of our work.

Monte Carlo simulations. – A decay experiment with a source of N atoms, and consisting of activity measurements A_i determined at t_i from the number of decays in the time interval $[t_i - \Delta t/2, t_i + \Delta t/2]$, was simulated using the function *RandomVariate* [19] of Mathematica 11 [20,21] set to produce pseudo-random variate from the binomial distribution with N trials and success probability P_i:

$$P_i = \lambda\int_{t_i-\Delta t/2}^{t_i+\Delta t/2} e^{-\lambda(t-t_0)}\mathrm{d}t, \tag{8}$$

where t_0 is the starting time of the experiment. For simplicity and without loss of generality, simulations reported in the following were performed with $N = 10^7$, $n = 40$, $\hat{\tau} = 10$ a.u. (arbitrary units) and, unless otherwise stated, $\Delta t = 0.2$ a.u.

Median and truncated mean of the half-life distribution. – If the half-life distribution eq. (3) was a perfect Cauchy (as eq. (6)), the half-life $\hat{\tau}$ would coincide with the sample median or the sample truncated mean [22,23]. The sample mean and sample variance would be undefined along with any moments of order greater than or equal to one. The half-life distribution, however, presents fundamental differences from the Cauchy distribution (see fig. 1) such as: i) the distribution tails are not heavy, which implies that the sample mean and variance are defined, ii) the distribution is in principle asymmetric, and iii) the distribution has a singularity at the half-life estimate $\tau = 0$. Despite these differences, simulations (see fig. 1) showed that in many cases of practical interest the median of the half-life distribution still provides an accurate estimate of the true half-life. These cases are defined by the parameter space $(n, \mu_0, \hat{\tau})$ of narrow distributions that do not involve the $\tau = 0$ singularity. Notice that the parameters n, μ_0, $\hat{\tau}$ determine the width of the Cauchy-like distribution qualitatively as in eq. (7). Therefore, one can easily see that the method of extracting half-life as the distribution median suffers in the limits of low initial activities and long half-lives. To estimate the uncertainty of a half-life measurement, we followed the steps of a) determine the median $\hat{\tau}_m$ of the experimental half-life distribution, b) run a large number of pseudo experiments each with half-life $\hat{\tau}_m$, and simulating the same number of decay events as in the real experiment, c) for each pseudo experiment determine the median of the half-life distribution, d) determine the standard deviation of the pseudo-experimental medians, which we assumed to be the statistical uncertainty of the experimental half-life. The same procedure was also used for the half-life measurement by truncated mean. The uncertainties reported in fig. 1 were determined using this procedure; another quantitative comparison between the median and the regression analysis is reported in the following sections describing the experimental measurement of the half-life of the radionuclide ^{211}Pb.

Least-square fit of the half-life distribution. – The least-square (LS) fit of the pdf to the half-life distribution is the most comprehensive way to study the half-life distribution and extract the half-life. However, because activity data are repeatedly used, entries into the half-life distribution are not independent (see fig. 2). As a consequence, i) the weights needed for the LS method are non-trivial to determine, and ii) simulations showed that the uncertainty estimated from the covariance matrix as square root of the diagonal elements largely underestimates the half-life uncertainty. Dealing with these issues is not a matter of standard statistical analysis, we addressed them using a MC approach described in the following.

Determination of weights. A large number of simulated experiments were performed with half-life $\hat{\tau}$ and initial activity $\mu_0/\Delta t$, and the resulting half-life distributions were binned in histograms of identical bin width. The histograms comparison allowed to determine the content distribution and the standard deviation σ of each bin. Figure 3 shows the content distribution of one bin chosen arbitrarily as an example, and σ as a function of the average bin content. We used the standard deviations σ computed from simulations to determine the weight $w = 1/\sigma^2$ for each bin needed for the LS fit.

Results of the LS fit for simulated decays. The unnormalised probability function resulting from the LS method agreed very well with simulations (see fig. 4). Deviations

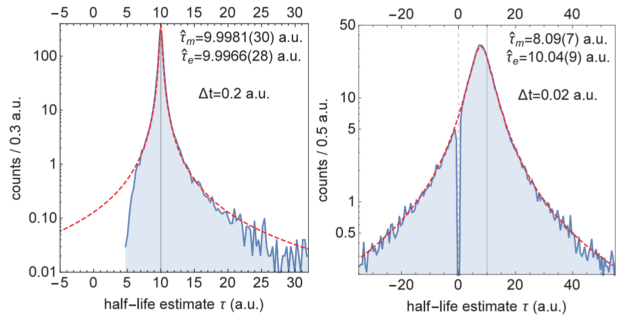

Fig. 1: (Colour online) Average of 100 simulated half-life distributions in case of (left) $\Delta t = 0.2$ a.u. and (right) $\Delta t = 0.02$ a.u. Such average is necessary to highlight the shape of the low-count distribution tails. For each simulated experiment a half-life was determined from exponential regression and statistical sampling by median. It is possible to compare the two methods after determining the average of the half-lives from exponential regression ($\hat{\tau}_e$) to the average of the half-life distribution medians ($\hat{\tau}_m$). Uncertainties were determined as standard deviation of the mean (sdom). When the half-life distribution assumes values across the singularity at $\tau = 0$ the measurement by median is affected by a significant systematic error. Otherwise, the median is a good estimator of the half-life despite the half-life distribution asymmetry. A Cauchy distribution (dashed red) is superimposed to guide the eye, with parameters of $\gamma = 0.182$ and $\hat{\tau} = 9.997$ a.u. (left) and $\gamma = 4.166$ and $\hat{\tau} = 7.835$ a.u. (right).

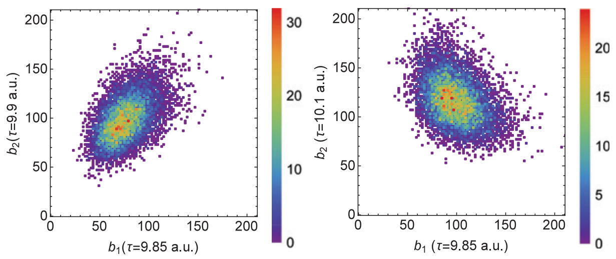

Fig. 2: (colour online) Left: histograms of 10^4 simulated half-life distributions were constructed. For each histogram the content of two bins b_1 and b_2 corresponding to the frequency of occurrence of the two half-life estimates τ_1 and τ_2 with $\tau_1 < \tau_2 < \hat{\tau}$, were plotted against each other in a 2D histogram. Right: same as in the left panel, but for $\tau_1 < \hat{\tau} < \tau_2$. The tilt of the ellipsoids implies that the frequency of occurrence of half-life estimates shorter (longer) than $\hat{\tau}$ are directly correlated to each other, and they are anticorrelated to frequency of occurrence of half-life estimates longer (shorter) than $\hat{\tau}$.

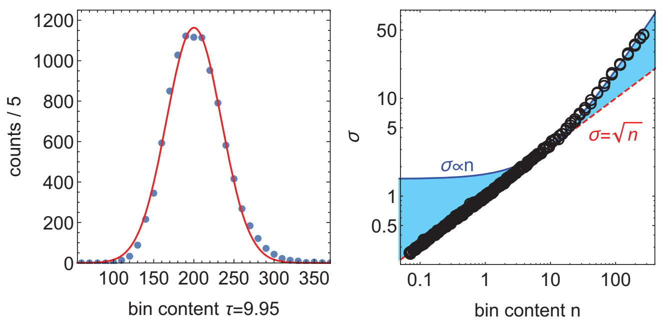

Fig. 3: (Colour online) 10^4 simulated half-life distributions were binned into as many histograms. The comparison of these histograms allowed the determination of: i) the content distribution of the i-th bin b_i, ii) the mean content of each bin $\bar{b}_i$, and iii) the bin's content standard deviation σ_i. The left panel shows the distribution of the content of the bin b_0 chosen arbitrarily as an example, and relative to the half-life estimate $\tau_0 = 9.95$ a.u. The fit of this distribution with a normal function (solid red) show a small deviation from normality. Right: the plot of σ_i as a function of $\bar{b}_i$. As opposite to the case of independent data that follows Poisson statistics, in general $\sigma_i \neq \sqrt{\bar{b}_i}$.

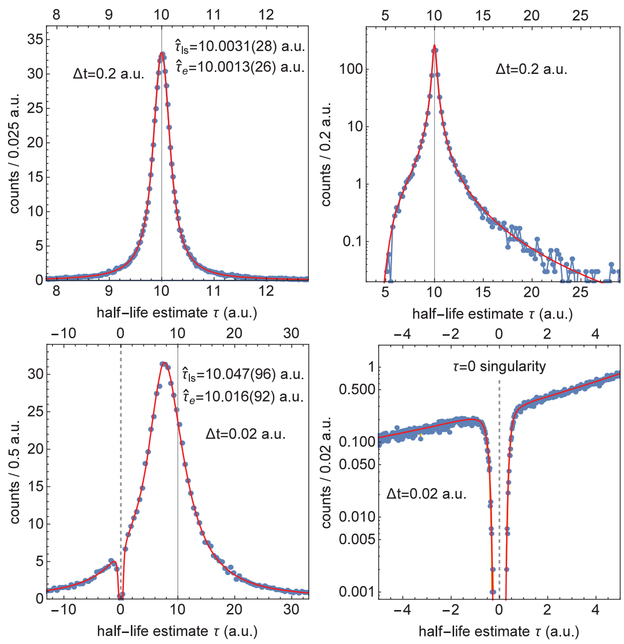

Fig. 4: (Colour online) LS fit of the average half-life distribution of 100 simulations using the unnormalized probability density (red solid line) and weights obtained from 10^3 MC simulations. The figure shows the case of top: $\Delta t = 0.2$ a.u. in linear (left) and logarithmic (right) scale, and bottom: $\Delta t = 0.02$ a.u., reporting for both cases the mean half-life obtained from the LS fits ($\hat{\tau}_{ls}$) and from exponential regression ($\hat{\tau}_e$) with their sdom. The pdf could account for all the features of the half-life distribution including the asymmetry and the $\tau = 0$ singularity (bottom right). In both cases, the LS fit provided half-lives in agreement with the exponential regression.

were only apparent in simulations when very low-activities were involved, when the normal approximation to the Poisson statistics (in which the pdf was derived) was not fulfilled. Agreement was also found in the case where the $\tau = 0$ singularity was concerned when, as discussed in the previous section, the median cannot be used as an estimate of the half-life. Notice that despite the apparent agreement between simulations and the pdf, the reduced χ^2 is not guaranteed to hold meaning as a goodness-of-fit parameter because the number of degree of freedom in case of non linear models is in general undetermined [24]. Alternative methods to address the goodness-of-fit are beyond the scope of this manuscript, however, the results of the statistical sampling could be compared precisely with the results of the exponential regression analysis. Figure 5 shows such comparison for a set of 10^3 simulations. For both methods the average half-life converged to the true half-life, which implies the two methods achieved the same trueness. However, the sample of half-lives resulting from the statistical sampling had a slightly larger standard deviation, *i.e.*, the method is less precise than the regression analysis. These conclusions did not change either when substantial interruptions in the experiment were introduced or when the sampling time of activity measurements was chosen randomly. The speculated advantage of the statistical sampling consisting in dealing with frequency of half-life estimates rather than time ordered activities is, therefore, not substantiated by this work.

The comparison between the statistical sampling and the exponential regression method showed two other main differences. In the first case the correlation between $\hat{\tau}$ and μ_0 is very small —likely a consequence of the almost perfect symmetry of the half-life distribution. As an example, in case of 10^7 decay and half-life $\hat{\tau} = 2000$ a.u., the correlation factors determined from the covariance matrix were 0.7 and 0.07 for the exponential regression and the statistical sampling fit, respectively. In addition, the sensitivity of the statistical sampling method to μ_0 is much smaller than for the exponential fit. In the case of the previous example, the measurement of μ_0 lead to $66.96(3)\times10^4$ s^{-1} and $77(20)\times10^4$ s^{-1} for exponential regression and the statistical sampling, respectively.

Test with experimental data: half-life of the radionuclide ^{211}*Pb.* The lead isotope ^{211}Pb is a naturally-occurring β^- emitter, member of the ^{235}U decay chain. It was measured recently at the National Physical Laboratory (NPL) by α-particle counting of twelve samples of ^{211}Pb in equilibrium with its α-emitting progeny ^{211}Bi and ^{211}Po [25]. The α-particle background was measured prior to each sample measurement and in each case it was less than $0.1\,\mathrm{s}^{-1}$. This was negligible compared to the source decay rate that over the measurement time of 10 half-lives decreased from $\approx 10^4$ to 10 s^{-1}. In this work we analysed the same data as in ref. [25] using both the exponential regression and the statistical sampling methods. Table 1 reports the results of the analysis for each of the twelve sources. The weighted average of the 12 measurements resulting from exponential regression

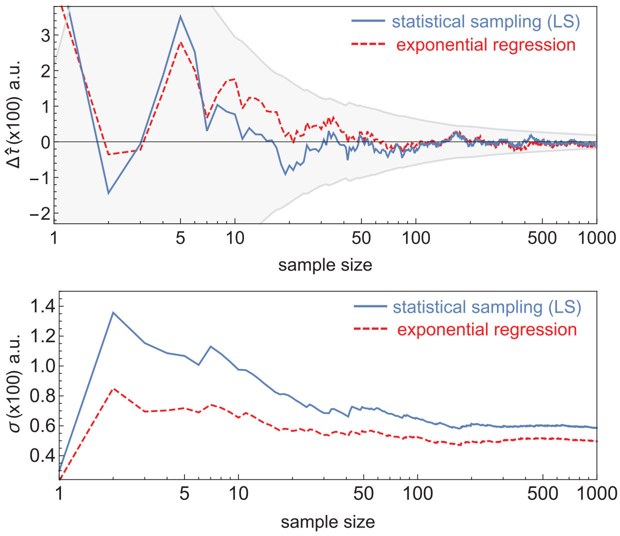

Fig. 5: (Colour online) Top: difference $\Delta\hat{\tau}$ between the true half-life $\hat{\tau}$ and the average half-life of simulated experiments measured by exponential regression $\hat{\tau}_e$ and statistical sampling (LS method) $\hat{\tau}_{ls}$ as a function of the sample size. $\Delta\hat{\tau}$ converged to zero for large number of experiments, and its fluctuations were within the expected uncertainty (shaded area) calculated as $\pm\sigma_{ls}$ where σ_{ls} was the sdom of $\hat{\tau}_{ls}$. Therefore, the trueness of the two estimators is comparable. Bottom: the standard deviations σ of half-lives as a function of the sample size for different methods are comparable, but the exponential fit achieves the best precision with smallest σ.

analysis 2169.77(15), in agreement with the statistical sampling results of 2169.94(17) s, 2170.01(17) s, and 2169.84(17) s is determined from the LS fit, median, and truncated mean, respectively. Both results are compatible with the recommended value 2170(2) s in ref. [25]. Notice that the uncertainty of the latter value includes an estimation of the systematic error, which is beyond the purpose of this work. In the case of the LS fit, the half-life needed to generate the simulated weights was the sample median, and the results of the fits are illustrated in fig. 6. For each source, the half-life determined by statistical sampling was in very good agreement with the result of the exponential fit. The agreement between the three statistical sampling methods is not surprising. In fact, this example fulfills the criteria highlighted in the previous section concerning narrow distributions and the singularity at $\tau = 0$. Uncertainties from MC simulation are compatible with uncertainties extracted from the covariance matrix in case of the exponential regression fit, but not in case of the statistical sampling, where uncertainty from the covariance matrix clearly underestimates the half-life uncertainty. Also in this case, with experimental data, we concluded that the statistical sampling method is less precise than the exponential regression analysis.

Conclusions. – This manuscript reports on our study of the statistical sampling method for the measurement of radioactive half-lives that allowed the first experimental

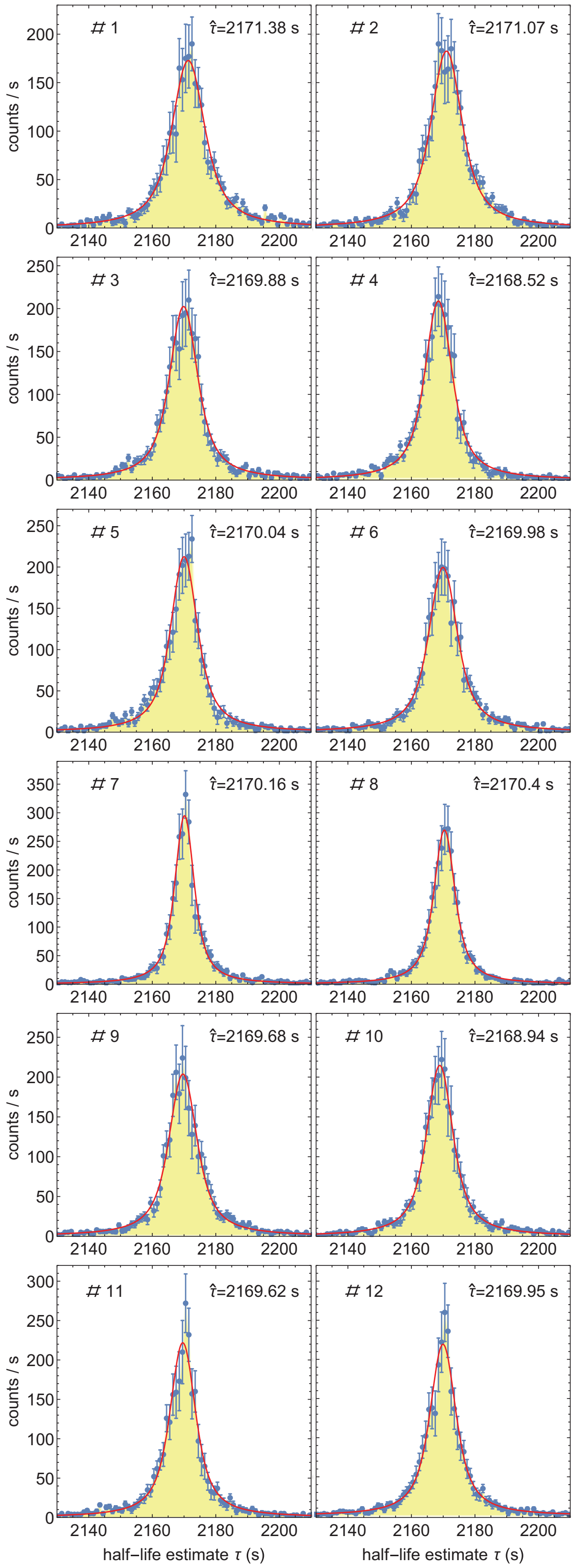

Fig. 6: (Colour online) Experimental half-life distribution of 12 sources of ^{211}Pb. The error bars shown were computed from simulation and used to determine weights (see text). The unnormalized probability function (red line) was fit to the experimental data.

Table 1: Half-life measurements of 12 different sources of ^{211}Pb using exponential regression and statistical sampling method. In the latter case the methods of median, truncated mean, and least squared fit were used. The uncertainty quoted from MC for the statistical sampling method is the one resulting from the median, which provide a conservative estimate or the uncertainty.

	Exponential Regression			Statistical Sampling				
Source #	$T_{1/2}$ (s)	σ (s) MC	σ (s) cov.	$T_{1/2}$ (s) LS	$T_{1/2}$ (s) median	$T_{1/2}$ (s) t.mean	σ (s) MC	σ (s) cov.
1	2168.63	0.61	0.52	2171.38	2171.67	2171.72	0.70	0.14
2	2170.60	0.57	0.49	2171.07	2171.09	2171.11	0.64	0.16
3	2170.22	0.51	0.44	2169.88	2169.99	2169.84	0.59	0.17
4	2169.16	0.52	0.45	2168.52	2168.39	2168.24	0.60	0.15
5	2169.92	0.62	0.54	2170.04	2169.79	2169.56	0.73	0.19
6	2169.88	0.56	0.49	2169.98	2170.03	2170.13	0.66	0.15
7	2169.01	0.43	0.38	2170.16	2170.29	2170.26	0.49	0.13
8	2170.36	0.40	0.36	2170.40	2170.46	2170.25	0.48	0.09
9	2170.03	0.47	0.42	2169.68	2169.74	2169.87	0.52	0.20
10	2169.89	0.50	0.45	2168.94	2169.11	2168.04	0.60	0.10
11	2169.54	0.51	0.45	2169.62	2169.68	2169.37	0.60	0.22
12	2170.06	0.52	0.46	2169.95	2170.05	2169.89	0.61	0.19
weighted mean	2169.77			2169.94	2170.01	2169.84		
uncertainty	0.15			0.17	0.17	0.17		

measurement of a radioactive isotope (^{211}Pb) by statistical sampling, and the first quantitative assessment of the method's performance in comparison with the exponential regression analysis. Correlation between entries of the half-life distribution is the main difficulty we encountered, which we addressed using a Monte Carlo approach. In particular, our analysis i) confirmed the validity of the probability density function determined in ref. [16] in a wider range of conditions than previously considered, ii) identified the conditions in which the median and the truncated mean of the half-life distribution can be used reliably to measure half-lives. These conditions are fulfilled in many cases of practical interest, offering a very simple mean to extract the half-life from experimental data with no need for any regression analysis. Our analysis also provided iii) a framework for the measurement of the half-life by statistical sampling using a least-square fit method. Based on such analysis, we concluded that the statistical sampling method is valid, and achieves the same trueness of the exponential regression. However, in comparison to the latter, the statistical sampling is slightly less precise. In addition, the suggested advantage of the statistical sampling in case of activity measurements recorded at irregular times was not recognized in this work; and the use of the method in the search for non-exponential decays was found to be difficult due to the lack of an obvious goodness-of-fit parameter. The method is, therefore, unlikely to replace exponential regression in any application requiring high precision. In the other cases, statistical sampling provides a valid alternative with a very intuitive visualization of the half-life that may be exploited in the future to some advantage.

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