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W. A. Beyer
C. Qualls*

*Visiting Staff Member. University of New Mexico, Albuquerque, NM 87106.

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by

W. A. Beyer and C. Qualls

ABSTRACT

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The event times are observed of a mixture of two kinds of radioactive atoms, each producing alpha particles. The probability of observing an event is p , $0 < p < 1$, and of missing the observation is $1 - p$. A maximum likelihood statistical method is given for estimating the size of the two populations. A second discussion assumes that the populations themselves are samples from parent populations. Bayesian methods are given for estimating parameters of the parent populations. An example is presented of an experiment involving the search for particle-bound polynutron systems.

I. INTRODUCTION

Suppose one is given a mixture of two populations, denoted by I and II, of atoms in which there are N_1 atoms of type I and N_2 atoms of type II. Suppose that each population undergoes radioactive decay with known constants λ_1 and λ_2 ; that is, the probability that an atom of type i will have decayed before time $t > 0$ is $1 - e^{-\lambda_i t}$ if, at time $t = 0$, the atom has not decayed. During time period $(0, \bar{t})$, the time intervals of decays of the mixture are recorded (that is, the centers of time intervals in which decay events occurred). It is given that the probability of observing a decay that occurs is p ($0 < p < 1$). The problem is to estimate N_1 and N_2 . In this report we give a maximum likelihood estimate of N_1 and a Bayesian probability distribution of μ , when it is assumed that N_1 is Poisson distributed with parameter μ and N_2 is uniformly distributed over some finite interval. An application is made to an experiment by Turkevich et al.¹ in which an estimate of the number of polyneutrons produced is required.

II. MAXIMUM LIKELIHOOD ESTIMATE OF POPULATION SIZES

Suppose the time interval $(0, \bar{t}]$ is subdivided by cut points $\{t_i | i = 0, 1, \dots, k\}$ where $0 = t_0 < t_1 < \dots < t_k = \bar{t}$. Put $q = 1 - p$ and $(t_{i-1}, t_i] \equiv I_i$. Let y_i , $1 \leq i \leq k$, be the number of events (decays) that are observed in interval i . Suppose first that only one type of atom is present at $t = 0$ and that the number of such atoms is N . Then the probability of observing the vector $y = (y_1, \dots, y_k)$ is given by

$$P(y|N) = \frac{N!}{y_1! y_2! \dots y_k! y'!} \prod_{i=1}^k \left[p \left(e^{-\lambda t_{i-1}} - e^{-\lambda t_i} \right) \right]^{y_i} \left(q + p e^{-\lambda \bar{t}} \right)^{y'}, \quad (1)$$

where $y' = N - \sum_{i=1}^k y_i$. In Eq. (1) the multinomial coefficient represents all ways that the set of decays of atoms can be distributed among the $k + 1$ cells I_i , $i=1, 2, \dots, k$, and the cell I_{k+1} that contains all the unobserved decays. Decays are not observed either because the atoms decayed in $(0, \bar{t}]$ but were not observed or because they decayed after $\bar{t}$. The second factor in Eq. (1) is the product of the probabilities of observing the y_i events in the interval I_i . The final factor in Eq. (1) is the probability of observing the y' events in the cell I_{k+1} .

Now consider the case where there are two kinds of atoms present. Let $\tilde{z}$ be the k -vector of observations. Then

$$P(\tilde{z} | N_1, N_2) = \sum_{\substack{\tilde{x} \geq 0 \\ \tilde{x} + \tilde{y} = \tilde{z}}} \sum_{\substack{\tilde{y} \geq 0}} P(\tilde{x} | N_1) P(\tilde{y} | N_2). \quad (2)$$

Put

$$\sum_{i=1}^k y_i = n_1, \\ \sum_{i=1}^k x_i = n_2,$$

$$n_1 + n_2 = n,$$

$$q_j = q + pe^{-\lambda_j \bar{t}}, \quad j = 1, 2$$

and

$$p_{ji} = p \left(e^{-\lambda_j t_{i-1}} - e^{-\lambda_j t_i} \right), \quad j = 1, 2, \quad 1 \leq i \leq k.$$

Also put

$$Q_{\tilde{z}}(n_1) = \sum_{\substack{\Sigma y_i = n_1 \\ 0 \leq y_i \leq z}} \prod_{i=1}^k \frac{p_{1i}^{y_i} p_{2i}^{z_i - y_i}}{y_i! (z_i - y_i)!}$$

for $\max(0, n - N_2) \leq n_1 \leq \min(n, N_1)$ and $Q_{\tilde{z}}(n_1) = 0$ otherwise. Then Eq. (2) can be written as

$$P(\tilde{z} | N_1, N_2) = \sum_{n_1 = \max(0, n - N_2)}^{\min(n, N_1)} \frac{N_1!}{(N_1 - n_1)!} \frac{N_2!}{(N_2 - (n - n_1))!} \times q_1^{N_1 - n_1} q_2^{N_2 - (n - n_1)} Q_{\tilde{z}}(n_1) \quad (3)$$

for $N_1 + N_2 \geq n$.

The maximum likelihood principle asserts that the "best" estimate for N_1 and N_2 is the pair (N_1, N_2) that maximizes $P(\underline{z} | N_1, N_2)$ for the observed $\underline{z}$. For a discussion of the maximum likelihood principle, see Good² and de Finetti.³

The fact that the quantity $Q_{\underline{z}}(n_1)$ is the coefficient of s^{n_1} in the polynomial

$$\phi(s) = \prod_{i=1}^k \frac{(p_{1i}s + p_{2i})^{z_i}}{z_i!}$$

is the basis of a fast algorithm for computing $P(\underline{z} | N_1, N_2)$.

To obtain bounds on the value of (N_1, N_2) , say $(\bar{N}_1, \bar{N}_2)$, that maximize $P(\underline{z} | N_1, N_2)$ for a given $\underline{z}$, it is necessary that

$$P(\underline{z} | \bar{N}_1 - 1, \bar{N}_2) - P(\underline{z} | \bar{N}_1, \bar{N}_2) \leq 0$$

and

$$P(\underline{z} | \bar{N}_1, \bar{N}_2 - 1) - P(\underline{z} | \bar{N}_1, \bar{N}_2) \leq 0.$$

A calculation gives

$$P(\underline{z} | \bar{N}_1 - 1, \bar{N}_2) - P(\underline{z} | \bar{N}_1, \bar{N}_2) = \left[-\frac{\bar{N}_1}{\bar{N}_1 - \sum x_i} \left(q + pe^{-\lambda_1 \bar{t}} \right) + 1 \right] \\ \times P\{\underline{z} | \bar{N}_1 - 1, \bar{N}_2\} ,$$

so that

$$\bar{N}_1 \leq \frac{\sum x_i}{p \left(1 - e^{-\lambda_1 \bar{t}} \right)} \leq \frac{\sum z_i}{p \left(1 - e^{-\lambda_1 \bar{t}} \right)}.$$

Likewise,

$$\bar{N}_2 \leq \frac{\sum z_i}{p \left(1 - e^{-\lambda_2 \bar{t}} \right)}.$$

Also,

$$\bar{N}_1 + \bar{N}_2 \geq \sum_{i=1}^k z_i.$$

III. GOODNESS-OF-FIT THEORY FOR MAXIMUM LIKELIHOOD ESTIMATE

In this section we discuss testing the null hypothesis that $N_1 = 0$ based on the model discussed in Sec. II. Under this hypothesis, the unknown number N_2 of times of decay are independent random variables with identical exponential distributions having parameter λ_2 . Denote these random times by $y_1, y_2, \dots, y_{N_2}$. Let Σ be the set of all subsets of the indices $\{1, 2, \dots, N_2\}$. Consider $\sigma \in \Sigma$ as specifying which decays are observed in $(0, \bar{t}]$. The complement σ^c indicates which decays are not observed either because they are missed with probability q or because they occur after $\bar{t}$. Since the probability of observing the decay during $(0, \bar{t}]$ for any given type II atom is $p \left(1 - e^{-\lambda_2 \bar{t}} \right)$, we have a probability measure on the 2^{N_2} points of Σ given by

$$P(\sigma) = \left(p - p e^{-\lambda_2 \bar{t}} \right)^n \left(q + p e^{-\lambda_2 \bar{t}} \right)^{N_2 - n},$$

where $n = |\sigma|$, the number of indices in σ .

Now the random number N of observed times of decay, denoted by $x_1, x_2, \dots, x_N$, can be viewed as having been selected from $Y_1, \dots, Y_{N_2}$ by the random element $\sigma \in \Sigma$. Consequently, the conditional cumulative distribution function of $x_1, x_2, \dots, x_N$, given σ , is

$$\begin{aligned} F(x_1, x_2, \dots, x_n | \sigma) &= \prod_{i=1}^n P\left(Y_{\sigma_i} \leq x_i | Y_{\sigma_i} \leq \bar{t}\right) \\ &= \prod_{i=1}^n \frac{1 - e^{-\lambda_2 x_i}}{1 - e^{-\lambda_2 \bar{t}}} \quad \text{for } 0 < x_i \leq \bar{t}, 1 \leq i \leq n, \end{aligned}$$

where $n = |\sigma|$ and σ_i is the i^{th} member of σ with the members of σ considered to be ordered. The conditional probability density function of $x_1, x_2, \dots, x_N$, given $N = n$, is

$$\begin{aligned} f(x_1, x_2, \dots, x_n | n) &= \sum_{|\sigma|=n} f(x_1, x_2, \dots, x_n | \sigma) P(\sigma | n) \\ &= \left[\prod_{i=1}^n \lambda_2 e^{-\lambda_2 x_i} / \left(1 - e^{-\lambda_2 \bar{t}} \right) \right] \sum_{|\sigma|=n} P(\sigma | n) \end{aligned}$$

for $0 < x_i \leq \bar{t}$ and $1 \leq i \leq n$. Note that $\sum_{|\sigma|=n} P(\sigma | n) = 1$.

So we have reduced the problem of testing whether $N_1 = 0$ to considering the data $x_1, x_2, \dots, x_N$ conditioned on the event $[N = n]$ as independent, identically distributed observations drawn from the distribution

$$F(x) = \frac{1 - e^{-\lambda_2 x}}{1 - e^{-\lambda_2 \bar{t}}} , \quad 0 < x \leq \bar{t} .$$

This test can be done using the Kolmogorov-Smirnov statistic

$$D_n = \sup_{0 < x \leq \bar{t}} | F_n(x) - F(x) | ,$$

where F_n is the empirical distribution function constructed from the data. We reject the null hypothesis that $N_1 = 0$ at a level of significance α if

$D_n > d_{n,\alpha}$ where $d_{n,\alpha}$ is defined by $P(D_n > d_{n,\alpha}) \leq \alpha$.

Finally, the unconditional test also satisfies α by the computation

$$\begin{aligned} P(D_N > d_{N,\alpha}) &= \sum_n P(D_n > d_{n,\alpha} \mid N = n) P(N = n) \\ &\leq \alpha \sum_n P(N = n) = \alpha . \end{aligned}$$

IV. A BAYESIAN ESTIMATE

A second mathematical model of the two-population problem will now be discussed. This model, which is used later, assumes that each of the population sizes is a random variable. The second population size N_2 is a random variable uniformly distributed on L to $L + \eta$. The first population size N_1 is selected at random from a Poisson distribution with parameter μ , with "efficiency" γ , $0 < \gamma < 1$, so that N_1 is a random variable with the distribution

$$\frac{e^{-\gamma\mu} (\gamma\mu)^k}{k!}, \quad k = 0, 1, 2, \dots,$$

where γ is given. The Poisson parameter is obtained from considering a compound Poisson distribution. See Feller,⁴ p. 171, problem 27, and p. 288.

Assume these distributions for N_1 and N_2 are superimposed upon the random experiment described in Sec. II. If the random experiment is repeated ℓ times with vector outcomes $\tilde{z}_1, \tilde{z}_2, \dots, \tilde{z}_\ell$, then the probability of obtaining the vectors $\tilde{z}_i$ for a given μ is given by

$$f(\tilde{z}_i, 1 \leq i \leq \ell | \mu) = \frac{1}{(\eta+1)^\ell} \prod_{i=1}^{\ell} \sum_{N_1^i=0}^{\infty} \sum_{N_2^i=L}^{L+\eta} P\left(\tilde{z}_i | N_1^i, N_2^i\right) \times e^{-\gamma_i \mu} \frac{(\gamma_i \mu)^{N_1^i}}{N_1^i!}. \quad (4)$$

Now suppose one assumes that μ has a probability distribution $p(\mu)$ on $(0, \infty)$. Then it follows from Bayes' formula that the posterior distribution of μ for given vectors $\tilde{z}_i$ is

$$g(\mu | \tilde{z}_i, 1 \leq i \leq \ell) = \frac{f(\tilde{z}_i, 1 \leq i \leq \ell | \mu) p(\mu)}{\int_0^\infty f(\tilde{z}_i, 1 \leq i \leq \ell | \bar{\mu}) p(\bar{\mu}) d\bar{\mu}}. \quad (5)$$

If one puts $p(\mu) = 1/a$ for $0 < \mu < a$ and $p(\mu) = 0$ for $\mu > a$ and then takes the limit as $a \rightarrow \infty$, one obtains

$$g(\mu | \tilde{z}_i, 1 \leq i \leq \ell) = \frac{f(\tilde{z}_i, 1 \leq i \leq \ell | \mu)}{\int_0^\infty f(\tilde{z}_i, 1 \leq i \leq \ell | \bar{\mu}) d\bar{\mu}}, \quad (6)$$

provided the denominator in Eq. (6) converges.

We now prove this convergence. In the following, $\sum_{1,2}$ is defined by

$$\sum_{1,2} = \sum_{N_1=0}^{\infty} \sum_{N_2=L}^{L+\eta} . \quad (7)$$

One has for each a , $0 < a < \infty$, and for some j , $1 \leq j \leq \ell$,

$$\begin{aligned} & \int_0^a f(z_{\sim 1}, 1 \leq i \leq \ell | \bar{\mu}) d\bar{\mu} \leq \int_0^a f(z_j | \bar{\mu}) d\bar{\mu} \\ &= \frac{1}{\eta + 1} \int_0^a \sum_{1,2} P(z_j | N_1^j, N_2^j) e^{-\gamma_j \mu} \frac{(\gamma_j \mu)^{N_1^j}}{N_1^j!} d\mu \\ &\leq \frac{1}{\gamma_j} \sum_{1,2} P(z_j | N_1^j, N_2^j) \\ &= \frac{1}{\gamma} \sum_{\substack{1,2 \\ N_1 + N_2 \geq n}} \sum_{n_1=\max(0, n-N_2)}^{\min(n, N_1)} \frac{N_1!}{(N_1 - n_1)!} \frac{N_2!}{[N_2 - (n - n_1)]!} \\ &\quad \times q_1^{N_1 - n_1} q_2^{N_2 - (n - n_1)} Q_{\tilde{z}}(n_1) \end{aligned}$$

(suppressing the index j)

$$= \frac{1}{\gamma} \sum_{N_2=L}^{L+n} \sum_{n_1=\max(0, n-N_2)}^n \frac{N_2!}{[N_2 - (n - n_1)]!} q_1^{-n_1} q_2^{N_2 - (n - n_1)} Q_{\tilde{z}}(n_1)$$

$$x \sum_{N_1=n_1}^{\infty} \frac{N_1!}{(N_1 - n_1)!} q_1^{N_1} < \infty,$$

because $0 < q_1 < 1$. So

$$\int_0^a f(z_i, 1 \leq i \leq \ell | \bar{\mu}) d\bar{\mu}$$

is bounded and monotonically increasing as $a \rightarrow \infty$ and therefore has a limit.

V. DESCRIPTION OF EXPERIMENT

The objective of an experiment recently performed by Turkevich et al., partly diagrammed in Fig. 1, was to measure the number of particle-bound polynutron systems x_n (for short: polyneutrons) produced in the reaction

The target may be, in various experiments, boron carbide, lead, or uranium. The following discussion depends on restricting attention to polyneutrons with $x \geq 4$.

The detection of polyneutrons in this experiment is somewhat complicated. The target is partly surrounded by a lead detector of ~ 4 kg of metallic lead, 52% being ^{208}Pb . The polyneutrons that are produced may react with ^{208}Pb in the lead detector according to

The ^{212}Pb then undergoes the following decay chains with indicated half-life times and, in the second case, the probabilities are given for the two branches:

Fig. 1.
Decay chain in experiment.

and

After exposure of the target to protons, the lead detector is dissolved. The solution containing ^{212}Pb and ^{212}Bi at the location in Fig. 1 marked (*) is called a "cow." A few mg ($\sim 10^{19}$ atoms) of ^{209}Bi are added to the solution and the mixture of ^{209}Bi and ^{212}Bi , with tracer amounts of ^{207}Bi (long-lived gamma emitter), are removed chemically from this mixture. The removal process is called "milking." A fraction $1 - \gamma$ of the atoms of the mixture is lost during the milking. The fraction γ is called the efficiency and can be determined by recovery of the macroscopic amounts of ^{209}Bi added or from recovery of the gamma radioactivity of ^{207}Bi . In the process of milking, atoms of ^{212}Pb and its daughter ^{212}Bi are introduced by contamination from the atmosphere, reagents, and glassware. These two contamination products are in radioactive equilibrium. The isolated sample of bismuth, marked (**) in Fig. 1, is put in an alpha counter and the 8.78-MeV alpha-decays, selected with good discrimination from other energy alpha particles, are observed for a period of time. Because of counter geometry, the fraction of alpha-decays recorded is about 0.3. After a milking, the number of ^{212}Bi atoms in the "cow" again increases from zero to some number in radioactive equilibrium with the ^{212}Pb present.

Because of the short half-life of ^{212}Po and because the 6.08 MeV alpha-decays are not counted, the reactions (9) to (11) can be replaced in this study by

Thereafter, ^{212}Bi refers to those ^{212}Bi with 8.78 MeV α -particle progeny.

Consider the sample marked (**) in Fig. 1. This sample contains ^{212}Bi and ^{212}Pb . There are two sources for the ^{212}Bi . One source is the chain which commenced with the reaction $x_n + ^{208}\text{Pb}$. The other source is the contaminate ^{212}Bi in decay equilibrium with its mother ^{212}Pb . It follows that for each decay of the mother contaminate ^{212}Pb there is a decay of the daughter contaminate ^{212}Bi . Thus the decay of the ^{212}Bi having the x_n as an ancestor appears to have a half-life of 60.6 min and the decay of the contaminate Bi appears to have a half-life of 10.6 h. (For further discussion, see Appendix A). This implies that $\lambda_1 = 0.01144$ decays/min and $\lambda_2 = 0.00109$ decays/min.

The objective is then to determine from the time history of the alpha-particle count, the number of (apparent) 10.6-h ^{212}Bi atoms present in the sample and the number of 60.6-min ^{212}Bi atoms present in the sample.

The lead detector, labeled (*) in Fig. 1, was milked sequentially three times in the experiment reported by Turkevich et al. Each of the samples was placed under a counter for a length of time. Table I gives the time histories of the 8.78-MeV events. The values are the centers of 15-min intervals in which an event was registered. Time is recorded in minutes from the beginning of the counting.

TABLE I

TIME HISTORIES OF 8.78-MeV EVENTS FOR THE THREE MILKINGS

	<u>I</u>	<u>II</u>	<u>III</u>
	22	53	8
	235	98	52
	250	114	177
	359	221	318
	406	304	349
	545	315	364
	792	488	456
	1157	719	487
		796	502
		1010	517
		1086	793
			884
			914
			1083
Number of events	8	11	14
Termination of counting	1500	1350	1500
Efficiency γ of milking	0.47	0.68	0.45

VJ. APPLICATION OF MAXIMUM LIKELIHOOD ESTIMATION TO THE EXPERIMENT

In this section we apply the mathematical model of Sec. II to the data given in Sec. V to obtain joint estimates of N_1 and N_2 .

The maximum likelihood estimates of N_1 , N_2 , together with the maximum value of $P\{z|N_1, N_2\}$ given by Eq. (2), are presented in Table II for each of the three milkings. Here N_1 is the estimated number of ^{212}Bi atoms with x_n as ancestor present after milkings and at the start of measurement ($t = 0$), and N_2 is the estimated number of contaminate ^{212}Bi atoms present at $t = 0$. The column in Table II marked "combined data" is calculated by assuming there is a milking in which the counts are obtained by interleaving the counts from each of the milkings; thus: 8, 22, 52, 53, 98, 114, 177, 221, 235, etc. The time of termination of the counting in the combined data is $\bar{t} = 1350$. Note that the efficiency γ does not appear in these calculations because N_1 refers to atoms after milking.

The estimates of N_1 indicate that a small number of polyneutrons might have been present in the milkings. Of course these estimates are subject to statistical fluctuations and it would be desirable to place upper and lower bounds on these estimates. This is mathematically difficult, but the goodness-of-fit result of Sec. III does say something about the lower bound. In Sec. VII a different model is used to obtain an upper bound.

Applying the Kolmogorov-Smirnov goodness-of-fit test⁵ when one assumes $N_1 = 0$ gives the results listed in Table III. This table indicates that the model of Sec. II with no polyneutrons present ($N_1 = 0$) is consistent

TABLE II
VALUES OF (N_1, N_2) THAT MAXIMIZE $P(z|N_1, N_2)$

	I	II	III	Combined Data
N_1	1	2	2	4
N_2	48	68	85	200
$P(z N_1, N_2)$	2.9×10^{-12}	7.0×10^{-15}	1.1×10^{-17}	4.6×10^{-29}

TABLE III
K-S GOODNESS-OF-FIT RESULTS

	<u>I</u>	<u>II</u>	<u>III</u>	<u>Combined</u>
Kolmogorov-Smirnov Statistic D_n	0.194	0.169	0.179	0.116
Critical values for $\alpha = 0.10$	0.411	0.352	0.314	0.21

with the data from the experiment. Consequently $N_1 = 0$ is no doubt the lower bound one would obtain by any reasonable procedure.

It seems reasonable that the data are also consistent with the model of Sec. II where N_1 and N_2 are the maximum likelihood estimates. This statement is based on two arguments. First, estimates of N_1 are close to $N_1 = 0$, and therefore the fit should be similar. Second, it is reasonable to expect the full model where the number N_1 of polynutrons is not restricted to zero to fit the data as well as the restricted model does.

VII. APPLICATION OF BAYESIAN ESTIMATION TO THE EXPERIMENT

The second mathematical model (Sec. IV) is applied to estimating the number of ^{212}Bi atoms having x_n as an ancestor as follows. As is stated in Sec. IV, the ^{212}Bi atoms present in the lead detector are the daughters of ^{212}Pb atoms. Hence the number of ^{212}Bi atoms having x_n as an ancestor present at any given time is a random variable. We wish to show that this random variable is Poisson distributed. Suppose initially w atoms of type ^{212}Pb and zero daughter atoms ^{212}Bi are present in the lead detector. Let λ_1 be the decay constant of the ^{212}Pb and λ_2 the decay constant for ^{212}Bi . What is the probability for a single atom ^{212}Pb to have decayed to ^{212}Bi and not further before time t ? Divide the interval $(0, t)$ into sub-intervals of length $\Delta\tau$ each. Then the probability that the above occurs is

$$\sum_{\text{all intervals}} \left[e^{-\lambda_1 \tau} - e^{-\lambda_1 (\tau + \Delta\tau)} \right] e^{-\lambda_2 (t - \tau - \Delta\tau)}$$

$$\rightarrow \lambda_1 \int_0^t e^{-\lambda_1 \tau} e^{-\lambda_2 (t - \tau)} d\tau, \quad \text{as } \Delta\tau \rightarrow 0,$$

$$= \frac{\lambda_1}{\lambda_2 - \lambda_1} \left(e^{-\lambda_1 t} - e^{-\lambda_2 t} \right).$$

Call the last quantity $f(t)$. The probability of i atoms at t being in the form ^{212}Bi is then

$$\binom{w}{i} [f(t)]^i [1 - f(t)]^{w-i}.$$

For w large, $f(t)$ small, and $w f(t)$ moderate, this is approximated by the Poisson distribution (Feller,⁴ p. 153)

$$\frac{[w f(t)]^i}{i!} e^{-w f(t)}.$$

(Also, cf. Feller,⁴ p. 424, problem 11, where it assumed that w is constant). Therefore, it is reasonable to assume the number N of ^{212}Bi atoms with x_n as ancestor has a Poisson distribution $e^{-\mu} \mu^k / k!$, $k = 0, 1, 2, \dots$, where $\mu = w f(t)$ is a parameter to be estimated. Note that μ refers to the number of atoms present before the milkings.

It is assumed that the time between milkings is short enough that μ does not change and long enough that the amount of ^{212}Bi reaches a steady state before each of the three milkings. It will be further assumed that the number of contaminate atoms N_2^i is uniformly distributed between 30 and 120. (A small number of subsequent dry-run experiments where no polynucleotides were created shows that this uniform distribution assumption on N_2 is not unreasonable.) For a given μ , the probability of producing three given vectors $\tilde{z}_i$, $i = 1, 2, 3$ of observations is then

$$f(\tilde{z}_1, \tilde{z}_2, \tilde{z}_3 | \mu) =$$

$$\frac{1}{91^3} \prod_{i=1}^3 \sum_{N_1^i=0}^{\infty} \sum_{N_2^i=30}^{120} P(z_i | N_1^i, N_2^i) e^{-\gamma_i \mu} (\gamma_i \mu)^{N_1^i} / N_1^i! . \quad (12)$$

For the three milkings and corresponding counts reported in Sec. V, the value of μ that maximizes Eq. (12) (maximum likelihood value) is given by $\mu = 1.59$ and the value of Eq. (12) is 2.66×10^{-17} .

To compare this estimate of $\mu = 1.59$ to the estimates given in Table II for the first mathematical model, it is necessary to multiply by the efficiencies for each of the three milkings. The results rounded to the nearest integer are not inconsistent with the results in Table III.

The estimates of Sec. IV and App. B give the final estimate

$$\int_0^{\infty} f(z_1, z_2, z_3 | \mu) d\mu \approx 3.48 \times 10^{-13}.$$

Figure 2, the graph of the probability distribution $g(\mu | z_1, z_2, z_3)$ for μ [Eq. (6)] for vectors z_i determined experimentally as discussed in Sec. V, shows that the probability that $0 \leq \mu \leq 17.3$ equals 0.9. One may interpret this as a 90% interval estimate of μ .

Fig. 2.

The probability distribution $g(\mu | z_1, z_2, z_3)$ as a function of μ . A 90% confidence interval is (0, 17.3).

VIII. CONCLUSIONS

A mixture of two populations of radioactive atoms is observed for a finite time by observing their alpha particle decays. The problem is that of estimating the sizes of these two populations, given the two decay constants. With some given probability, alpha particle emission events may occur, but not be observed. Two methods of estimation are presented: maximum likelihood estimation and Bayesian estimation.

In the maximum likelihood estimation, a formula [Eq. (2)] is derived, which expresses the probability $P\{\tilde{z}|N_1, N_2\}$ of a vector $\tilde{z}$ of observations for given population sizes N_1 and N_2 of the two populations. In the Bayesian estimation, the second population size N_2 is assumed to be a random variable uniformly distributed on the positive integers between L and $L + n$. The first population size N_1 is assumed to be selected from a Poisson distribution

$$e^{-\gamma\mu} \frac{(\gamma\mu)^k}{k!}, \quad k = 0, 1, 2, \dots,$$

where μ is uniformly distributed in a generalized sense on $(0, \infty)$ and γ , $0 < \gamma < 1$, is an efficiency that depends on the experiment. The probability distribution on μ , for a given set of ℓ experiments with observation vectors $\tilde{z}_1, \tilde{z}_2, \dots, \tilde{z}_\ell$ is given by Eq. (6).

These two models are applied to experimental results in a search for particle-bound polynutron systems reported by Turkevich et al. The two models gave consistent answers. The maximum likelihood estimate of N_1 was $N_1 = 4$. The Bayesian estimate μ is 1.59 for the combined three experiments and 90% of the posterior distribution of μ is in the interval $(0, 17.3)$. It is emphasized that the maximum likelihood estimate of $N_1 = 4$ is of the total number of polynutron indicators obtained in all the milkings at the box marked (**) in Fig. 1, while the Bayesian estimate $\mu = 1.59$ is of the mean number of polynutron indicators available to any one of the three milkings at the box marked (*) in Fig. 1. To compare the estimates of the two models in the report, one can consider the combined estimate of $N_1 = 4$ to be an estimate of the parameter $\mu(\gamma_1 + \gamma_2 + \gamma_3)$. Knowing the efficiencies γ_i , the maximum likelihood model can then be used to estimate $\mu = 2.5$, while the Bayesian model estimates $\mu = 1.59$.

Some open mathematical problems should be mentioned: (1) extend the goodness-of-fit results of Sec. III, which were restricted to the case $N_1 = 0$, to the full model and (2) determine if a confidence interval for N_1 can be derived without using Bayesian techniques.

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APPENDIX A

DECAY OF PARTICLES INITIALLY IN RADIOACTIVE EQUILIBRIUM

The proof of the statement on p. 10, Sec. V, is as follows. Recall that there are two chains in the sample, both of which involve the decay of ^{212}Bi .

In chain 1, the amounts of ^{212}Pb and ^{212}Bi present initially are such that they are in radioactive equilibrium. In chain 2, only ^{212}Bi is present initially. Let $\text{Pb}^j(t)$ and $\text{Bi}^j(t)$, $j = 1$ and 2 , be the amount of ^{212}Pb and ^{212}Bi present in chain j . In chain 1,

$$\frac{d \text{Pb}^1}{dt} = -\lambda_1 \text{Pb}^1$$

and

$$\frac{d \text{Bi}^1}{dt} = \lambda_1 \text{Pb}^1 - \lambda_2 \text{Bi}^1.$$

In chain 2,

$$\frac{d \text{Bi}^2}{dt} = \lambda_2 \text{Bi}^2$$

or

$$\text{Bi}^2(t) = \text{Bi}^2(0) e^{-\lambda_2 t}.$$

In chain 1, because of initial equilibrium,

$$\frac{d \text{Bi}^1}{dt} (0) = 0 ,$$

so that

$$\text{Bi}^1(0) = \frac{\lambda_1}{\lambda_2} \text{Pb}^1(0) .$$

Now

$$\text{Pb}^1(t) = \text{Pb}^1(0) e^{-\lambda_1 t}$$

and

$$\frac{d \text{Bi}^1}{dt} = \lambda_1 \text{Pb}^1(0) e^{-\lambda_1 t} - \lambda_2 \text{Bi}^1$$

or

$$\frac{d \text{Bi}^1}{dt} + \lambda_2 \text{Bi}^1 = \lambda_1 \text{Pb}^1(0) e^{-\lambda_1 t}$$

or (suppressing the superscript "1")

$$\begin{aligned}
 \text{Bi}(t) &= e^{-\lambda_2 t} \lambda_1 \text{Pb}(0) \int_0^t e^{(\lambda_2 - \lambda_1)\tau} d\tau + e^{-\lambda_2 t} \text{Bi}(0) \\
 &= e^{-\lambda_2 t} \lambda_1 \text{Pb}(0) \left[\frac{e^{(\lambda_2 - \lambda_1)t} - 1}{\lambda_2 - \lambda_1} \right] + e^{-\lambda_2 t} \text{Bi}(0) \\
 &= \text{Bi}(0) e^{-\lambda_1 t} \left[1 + \lambda_1 t + \frac{(\lambda_1 - \lambda_2)(1 + \lambda_1 - \lambda_2)t^2}{2!} + \dots \right]. \quad (\text{A-1})
 \end{aligned}$$

Thus for small $\lambda_2 t$,

$$\text{Bi}^1(t) \cong \text{Bi}^1(0) e^{-\lambda_1 t}. \quad (\text{A-2})$$

A comparison of Eqs. (A-1) and (A-2) shows more precisely what is asserted in the statement that the decay of ^{212}Bi having x_n as an ancestor appears to have a half-life of 60.6 min and the decay of the contaminant ^{212}Bi appears to have a half-life of 10.6 h.

A. PENDIX B

ESTIMATE OF REMAINDER IN INFINITE INTEGRAL

Numerical integration is used to estimate the integral appearing in Eq. (6) when $\ell = 3$:

$$\int_0^\infty f(z_1, z_2, z_3 | \mu) d\mu.$$

The integral is estimated by

$$\int_0^\infty f(z_1, z_2, z_3 | \mu) d\mu \approx \sum_{i=1}^{\bar{i}} f(z_1, z_2, z_3 | \mu_i) \Delta\mu + \int_A^\infty f(z_1, z_2, z_3 | \mu) d\mu,$$

where $\Delta\mu = 0.1$, $\mu_i = 0.1i$ ($i = 1, 2, \dots, \bar{i}$), and $A = 0.1\bar{i} + 0.05$. Choose A so that $\int_A^\infty$ is small. A value for A is estimated as follows. Define

$$(P_2 P_3)^{\max} = \max_{N_1, N_2} P(z_2 | N_1, N_2) \max_{N_1, N_2} P(z_3 | N_1, N_2).$$

Define $\sum_{1,2}^i$ as in Eq. (7) but with N^i replacing N . Then

$$\begin{aligned} & \int_A^\infty f(z_1, z_2, z_3 | \mu) d\mu \\ &= \frac{1}{(\eta+1)^3} \int_A^\infty \prod_{i=1}^3 \sum_{1,2}^i P(z_i | N_1^i, N_2^i) e^{-\gamma_i \mu} \frac{(\gamma_i \mu)^{N_1^i}}{N_1^i!} d\mu, \\ &\leq \frac{(P_2 P_3)^{\max}}{\eta} \int_A^\infty \sum_{1,2}^1 P(z_1 | N_1^1, N_2^1) e^{-\gamma_1 \mu} \frac{(\gamma_1 \mu)^{N_1^1}}{N_1^1!} d\mu, \\ &\leq \frac{(P_2 P_3)^{\max}}{\eta} \sum_{1,2}^1 P(z_1 | N_1^1, N_2^1) \frac{1}{N_1^1!} \int_A^\infty e^{-\gamma_1 \mu} (\gamma_1 \mu)^{N_1^1} d\mu, \\ &= \frac{(P_2 P_3)^{\max}}{\eta} \sum_{1,2} P(z | N_1, N_2) \frac{1}{N_1! \gamma} \int_{\gamma A}^\infty e^{-x} x^{N_1} dx, \end{aligned}$$

(suppressing "1")

$$= \frac{(P_2 P_3)^{\max} e^{-\gamma A}}{\eta \gamma} \sum_{1,2} P(z|N, N_2) \frac{(\gamma A)^N}{N!} \left[1 + \frac{N}{\gamma A} + \frac{N(N-1)}{(\gamma A)^2} + \dots + \frac{N!}{(\gamma A)^N} \right],$$

(where $N = N_1$)

$$= \frac{(P_2 P_3)^{\max} e^{-\gamma A}}{\eta \gamma} \sum_{1,2} P(z|N, N_2) (\gamma A)^N \left[\frac{1}{N!} + \frac{1}{(N-1)! \gamma A} + \dots + \frac{1}{(\gamma A)^N} \right],$$

$$< (P_2 P_3)^{\max} \frac{e^{-\gamma A}}{\eta \gamma} \sum_{1,2} P(z|N, N_2) (\gamma A)^N \frac{(N+1)}{(\gamma A)^{r-1} (N-r+1)!},$$

{ where $r = \max(0, [N - \gamma A])$ }

$$= (P_2 P_3)^{\max} \frac{e^{-\gamma A}}{\eta \gamma} \sum_{1,2} \sum_{n_1=\max(0, n-N_2)}^{\min(n, N)} \frac{N!}{(N-n_1)!} \frac{N_2!}{[N_2 - (n-n_1)]!}$$

$$\times q_1^{N-n_1} q_2^{N_2 - (n-n_1)} Q_{\tilde{z}}(n_1) \frac{(\gamma A)^N (N+1)}{(\gamma A)^{r-1} (N-r+1)!}$$

$$= \frac{(P_2 P_3)^{\max} e^{-\gamma A}}{\eta \gamma} \sum_{N_2=L}^{L+1} \sum_{n_1=\max(0, n-N_2)}^n \frac{N_2!}{[N_2 - (n-n_1)]!}$$

$$\times q_1^{-n_1} q_2^{N_2 - (n-n_1)} Q_{\tilde{z}}(n_1) \sum_{N=n_1}^{\infty} \frac{(\gamma A)^{N-r+1} (N+1)!}{(N-r+1)!} \quad (B-1)$$

The infinite series is estimated as follows.

$$\begin{aligned}
& \sum_{N=n_1}^{\infty} \frac{q_1^N}{(N-n_1)!} \frac{(\gamma A)^{N-r+1}}{(N-r+1)!} (N+1)! \\
& \leq \frac{(\gamma A)^{\gamma A+2}}{[\gamma A]!} \sum_{N=n_1}^{\infty} (N+1)(N)(N-1)\dots(N-n_1+1) q_1^N \\
& \leq \frac{(\gamma A)^{\gamma A+2}}{[\gamma A]!} \sum_{N=n_1}^{\infty} (N+1)^{n_1} q_1^N.
\end{aligned}$$

The series $\sum_{N=n_1}^{\infty} (N+1)^{n_1} q_1^N$ is convergent because $0 < q_1 < 1$ and its remainder may be estimated.