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Serial Statistics: Is Radioactive Decay Random?

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Based on more than 10^8 counts obtained from γ emissions arising from cobalt-60 and cesium-137 nuclei, *serial* statistical tests—the sum of squares of 0,1 standardized slopes of linear regressions and the sum of squares of the closely related 0,1 standardized correlation coefficients—exhibit significant deviations from the theoretic (random) expectation as a function of differences in the source environment. On the other hand, more conventional, *nonserial* statistical tests—the χ-square goodness-of-fit and index of dispersion tests—derived from the same data are indistinguishable from those expected for random events. These serial discrepancies raise a substantial question as to the randomness of the detected emissions and, insofar as emissions and decay events are appropriately interrelated, the independence of the events themselves.

Introduction

Recently, Anderson, employing nonserial index of dispersion tests, reported that, under certain conditions, β radiation emitted by carbon-14-labeled organic submonolayers is not properly described by the Poisson distribution.[1] In contrast to generally accepted nuclear theory, the implication of this work is that the events themselves are thus not independent under those particular conditions.

Since it is unlikely that the causal factor for such anomalous statistical behavior is the formation of interactions *only* under those specific monolayer conditions cited, the possibility exists that such interactions, as shown by detected emissions being other than random, would generally be present also in nonmonolayer configurations. In order to test this possibility, a large number of sequential count totals arising from detected γ emissions of cobalt-60 and cesium-137 sources held under several different environmental conditions have been examined using a variety of statistical tests.

Historically, *nonserial* statistical methods have been employed to test experimental observations of radioactive emissions and thereby the adequacy of the thesis of independence of radioactive decay events. Primarily these have been the *chi-square* (χ^2) test which permits testing of the hypothesis that an observed frequency distribution is of the same population as a theoretic one and the *index of dispersion* (s^2/m) which is the ratio of the observed variance to the best estimate of σ^2, *i.e.*, the mean for Poisson distributions. Each test measures only specific parameters of the observed distributions and, in general, these parameters are not identical for the different tests.

Applied to radioactive counting, conformance of observed distributions with the expectation using a single statistical test has, in the literature, generally been taken as proof that the underlying assumption of independence has been verified. All that can reasonably be concluded, however, from conforming results of a single test, (*e.g.*, *P*'s of >0.05 or 0.01) is that the results are not inconsistent with the thesis of randomness and, insofar as emissions are directly related, of the independency of the events themselves.

If a series of numbers (such as radioactive counts) are, in fact, random, then each statistical test which measures at least one property of randomness must consistently show conforming results with an appropriately high frequency; the population of the counts must be, within accepted probability limits, of the same population as theory would predict and as would result from randomly generated numbers themselves. In the absence of artifact, consistently nonconforming results as shown by even one valid test are thus sufficient to raise serious questions as to the validity of the thesis of randomness of what is actually measured and to render the generality of this thesis untenable.

The earlier published evidence shows that the observed distributions, primarily of α emissions, were not differentiated from the expectation, *i.e.*, the Poisson.[2] At least one exception has already been noted.[3] Berkson has more recently reevaluated some of the earlier work using the s^2/m test; this evidence does not now appear as compelling as was originally thought.[4] Further, in studying times-between-α-emissions arising from ^{241}Am, Berkson, employing nonserial tests primarily, did not observe significant differences between the observed and expected distributions, although he noted what seemed to be a high correlation among consecutive times in one case. He concluded: "... *can we consider this examination a fairly definitive establishment of the randomness of such emissions? I do not think so ... I had the impression that a quite extreme departure from randomness might be operative without its being detected by these statistical tests* ..." (italics added).

Statistically, radioactive counts obtained as a sequential series of count totals (counts per unit time) may be considered as a set of *nonserial* totals for purposes of calculating the χ^2 goodness-of-fit test or the index of dispersion.

Such sequential counts may also be considered as *serial* values with respect to time, permitting the derivation of statistical indices such as, for example, the linear correlation coefficients or the slopes of linear regressions. In such series, each correlation coefficient is a measure of the linear correlation that exists within the set between counts and time while the slope statistic is a measure of the change in "mean" within each set also with respect to time. Since the population mean does not sensibly change due to radioactive decay for isotopes of such long half-lived species as ^{60}Co and ^{137}Cs during counting periods of less than 15 min as are described herein, the theoretic distribution of the slope is the distribution of the *apparent* slopes observed for finite samples and is a function of the mean and of the number of counting periods in each data set. The statistics, derived from the linear regression analysis, may thus be compared as is reported herein with well defined theoretic distributions in order to obtain probabilities that the observed distributions are of the same population as are those expected theoretically.

Other serial statistical tests may also be employed to test the adequacy of the thesis of randomness. Thus autocorrelation coefficients for the same data sets as have been examined herein have been calculated and have been compared to approximate theoretic distributions, distributions which are considerably less rigorously derived than are those of the linear regression cited. Largely confirming the linear regression statistics, the analyses based on the autocorrelation coefficients are summarized in the supplemental notes.[5]

Experimental Section

Detection and Counting Equipment. Counts for the various statistical analyses were all produced by detection of γ emissions from small sources (*i.e.*, less than 5 μCi per source of ^{60}Co or ^{137}Cs) positioned more than 3 cm from the detector using in sequence a sodium iodide crystal detector and photomultiplier tube, preamplifier, amplifier, single channel analyzer, and a multichannel analyzer operated in the multiscale mode thereby permitting automatic accumulation of data sets of 1024 separate and precisely timed consecutive count totals (0.4 sec/channel for all ^{60}Co series and 0.8 sec/channel for all ^{137}Cs runs). The multichannel analyzer analog-to-digital converter was bypassed in the arrangement with the result that the equipment resolution time loss was insignificant at the relatively low levels of count rate involved (in excess of 1500 counts/sec in only 5 of the 262 series and less than 2600 cps in all runs).

The detector–PMT assembly, whose case was electrically grounded, was surrounded by lead bricks on top and on the sides, all positioned inside a styrofoam box to decrease to a minimum any possible counting fluctuations due to ambient temperature variations within each 409.6- or 819.2-sec period as well as to reduce the effect of background. Experimentally the variations of the ambient were observed to be less than 0.1 °C/15-min period during several tests with the result that the variations within the enclosure must have been even less. Except for loss of power due to line interruptions which occurred two or three times over the 10 months of the project, both the high voltage and the main power were kept "on" over the period of the project. Further to ensure maximum stability of the detector–PMT output, a highly stabilized high-voltage supply was employed.[6]

The basic multichannel analyzer was made by Nuclear Data and the auxiliary equipment by Canberra Industries.[7] The equipment was demonstrated on several occasions (prior to the start of the tests, approximately midway during the 10-month period of the tests, and following completion of the tests) to record "standard" counts properly in each channel using similar "window" limits for the particular input signal and bypassing only the detector–PMT system. When these "standard" tests were run using pulses based ultimately on a crystal oscillator, the input/output counts per channel did not vary at all over the 1024 channel sequences. At similar count rates to those employed during the radioactive counting, no drift whatsoever was observed under this test condition. These checks demonstrate a high likelihood that this portion of the equipment (preamplifier, amplifier, SCA, and multichannel analyzer portions) made no random or nonrandom contribution to the recorded counts.

The output of the PMT was also examined repeatedly during the course of the work using a visual oscilloscope presentation. No apparent variation in the form of the output was observed during these tests (*cf.* note 8).

For ^{137}Cs, the SCA baseline was set in the valley below the photopeak and the window was set at a point above the peak so that no sensible loss of signal occurred but also so that substantially only photopeak signals were counted. In the case of ^{60}Co, both photopeaks were similarly included within the baseline–window setting but the sum peak was excluded. Both baseline and window settings were verified frequently; no significant long term drift was noted.[8]

The memory-retained counts were typed out by teletypewriter and simultaneously punched into paper tape (ASR-33 terminal). The paper tapes were subsequently converted into series of punched cards using a computer program that automatically flagged those numbers which did not conform to the standard pattern of six digits (*e.g.*, 000544). The few errors thus defined were corrected by comparison with the teletyped record and new cards were punched. In only two cases, zero values were observed both in the paper tape and in the teletyped records; these two particular sets of 1024 counts each were discarded and are not included in the statistical summaries.

Following preparation of the punched cards, the data were analyzed using an IBM 360/30 computer and Fortran IV programs (double precision where indicated) that were prepared specifically for the purpose. Each punched card was individually verified to be in sequence prior to analysis of the serial statistics.

No human-error-producing step was thus encountered between the actual decay events and the derivation of statistical properties, as shown by the several tests. The average count total per unit time (*i.e.*, channel) was approximately 400 with very few data sets having more than 600 or less than 200 counts per unit time.

Radioactive Sources. All radioactive sources used in the work reported herein were prepared by evaporative deposition (aided by a heat lamp above) of mildly acid chloride solutions (generally of 10 μl or less) onto 3003 aluminum foil or onto clear adhesively surfaced plastic film substrates (acetate of 0.13-mm thickness) followed by covering of the crystalline residues with another small section of adhesively surfaced plastic. In one source, two self-adhesive films were used to encapsulate an aqueous solution of ^{137}CsCl (*ca.* 20 μl). Prior to counting, the source substrates were trimmed to *ca.* 3 × 3 cm size with the actual radioactive portion centered in the sandwich.

Prior to deposition of the radioactive solutions, the aluminum foil was freed of prior adsorbents by heating at 320° in the laboratory atmosphere for a period of at least 20 min as has been described elsewhere.[1]

Following preparation, sources which had been deliberately subjected to an environment other than ambient were never used a second time at ambient nor were they used other than in sequential series at other than ambient.

Environmental Conditions during Source Counting. (1) Nongrounded Aluminum Substrates. (a) ^{60}*Co.* Forty series (8 sources) were counted under "steady state" conditions in which the aluminum backing was insulated from electrical ground by wedging the source between two nested styrofoam cups (in turn rigidly positioned with respect to the detector crystal), the inner one having a small *ca.* 1-cm hole cut out on the lower side, the hole being positioned adjacent to the aluminum on the other side of which was the site of the radioactive salt. Thirty-three series (10 sources) were counted while the source, positioned as described above, was cooled with liquid nitrogen by keeping the nitrogen level in the inner cup well above the exposed aluminum backing. Another 16 series (4 sources) were counted with the source taped to a polyethylene sleeve in turn immersed in liquid nitrogen in a Pyrex Dewar. (No apparent change in the statistical behavior was noted in the two types of −196° cooling.) When the styrofoam cups were used they were, in turn, immersed to approximately 1 cm from the top of the outer cup in *ca.* 1 l. of water to prevent changes in count due to adventitious condensation of moisture. While cooled, counts were recorded immediately after the nitrogen had become quiescent and each half-hour thereafter but in no event maintaining the cooling for longer than four consecutive counting sequences (<2 hr per source). Twenty-two series (5 sources) were recorded during warmup from the −196° condition, in each case within 105 min of disappearance of coolent.

(b) ^{137}*Cs.* Fifty-four series (7 sources) were counted under "steady-state" conditions; 4 series (2 of the same sources) were recorded during the first 40 min of cooling to −196° while 4 more (the same 2 sources) were obtained during the first 40 min following disappearance of the liquid nitrogen from the styrofoam cups as in (1a) above.

(2) Grounded Aluminum Substrates. ^{137}*Cs.* Forty-one series (4 sources) were counted while the aluminum substrates were deliberately electrically grounded, in each case the source being maintained at ambient; 8 series (1 source) were counted while the source was held at −196° during 320 min.

(3) Plastic Encapsulated Sources. ^{137}*Cs.* Seventeen series (4 sources) were counted at ambient while 10 series (2 of the 4 sources) were counted during cooling to −196° in a Dewar during 135 min in each case; 13 series (1 source) were counted using the aqueous solution of CsCl.

Statistical Tests

The nonserial tests listed herein involved comparisons of observed distributions with the theoretic expectation using (1) the χ^2 goodness-of-fit test of frequencies of observed counts per unit time with frequencies of actual Poisson generated counts and (2) the index of dispersion (s^2/m) employing each group of 1024 sequential count totals as an independent set of data. In addition, the linear correlation coefficient and the regression slope (and, as recorded in the supplemental notes, the autocorrelation coefficients using lags of 1 through 320 in each set of data) have been calculated for each set of 1024 automatically time-sequenced counting series.[9]

No attempt was made directly to derive statistics from among the several sets (such as common means or cross correlations) since the starting points (in time) of each set were always initiated manually and somewhat arbitrarily. Once derived, statistical indices have been combined with others calculated similarly (as the sum of the squares) in order to examine overall probabilities for many sets of data observed under the homogeneous conditions listed above.

To facilitate analysis, linear correlation coefficients were converted to values (that would have an approximate 0,1 normal distribution if they were derived from random data) by means of the "z" transformation, *i.e.*, by calculating the hyperbolic arc tangent of each ($= 0.5 \ln \{(1 + r)/(1 - r)\}$), by subtracting the approximate theoretic mean ($= 0$), and by dividing by the approximate theoretic standard deviation ($= 1/(n - 3)^{1/2} = 0.031296$).[10] The sum of the squares—and the mean square (called $s^2/\hat{\sigma}^2$ in the tables)—was then calculated, separately, for the standardized hyperbolic arc tangents of the correlation coefficients. Each sum of squared standardized values is distributed (approximately) as χ^2 with the degrees of freedom equal to the number of independent data sets.

Since the regression slopes are theoretically normally distributed, they were 0,1 standardized merely by subtracting the theoretic mean ($= 0$) and by dividing by the best estimate of the theoretic standard deviation ($= \{m/\Sigma(t_i - t)^2\}^{1/2}$).[11] Since m is itself a variable, derived from a Poisson distribution, the resulting 0,1 standardized distribution is not, strictly speaking, normally distributed. However when the slope variance for each data set is accumulated by summation of the squared 0,1 standardized slopes, the summation within each homogeneous grouping is almost identical with the similar measure of the overall variance obtained by substituting the value $\{m(n - 1)\}$ for $\{\Sigma(a_i - m)^2\}$ followed by calculating the individual values by means of the "z" transformation, thus indicating that the deviation from the 0,1 distribution is relatively insignificant at least for the variance analysis employed herein.

Since independently (as well be seen in Table I) the index of dispersion (s^2/m) has been found to approximate

TABLE I: Non-serial Tests and Probabilities

Source (data sets)[a]	χ^2 Test (Poisson) χ^2/DF (D.F.)	P^b	Index of dispersion s^2/m (D.F.)	P^b
(1a) ^{60}Co: Steady State (40)	0.993 (3188)		0.997 (40920)	
: Low Temp (49)	0.982 (3610)		1.001 (50127)	
: Warmup (22)	0.990 (1226)		1.006 (22506)	
Combined (111)	0.988 (8024)	*0.217*	1.001 (113553)	*0.553*
(1b) ^{137}Cs: Steady State (54)	0.991 (4322)		0.996 (55242)	
: Low Temp (4)	1.120 (226)		1.011 (4092)	
: Warmup (4)	0.987 (222)		1.021 (4092)	
Combined (62)	0.997 (4770)	*0.443*	0.999 (63426)	*0.401*
(2) ^{137}Cs: Steady State (41)	0.980 (3477)		0.995 (41943)	
: Low Temp (8)	0.973 (575)		0.998 (8184)	
Combined (49)	0.979 (4052)	*0.173*	0.996 (50127)	*0.238*
(3) ^{137}Cs: Aq. Solution (13)	1.008 (1191)		0.986 (13299)	
: Crys. SS (17)	1.030 (1106)		0.996 (17391)	
: Crys. LT (10)	1.006 (589)		0.975 (10230)	
Combined (40)	1.023 (2866)	*0.731*	0.988 (40920)	*0.036*
Randomly generated (90)	0.985 (7870)	*0.174*	0.997 (92070)	*0.260*

[a] Groups 1a and 1b on nongrounded Al; group 2 on grounded Al; group 3 in plastic (acetate) encapsulation. [b] Probability (P) is expressed as the likelihood that a value of the statistic (of the same or lesser size) would be observed in a random distribution.

TABLE II: Serial Tests and Probabilities

Source[a]	Linear correlation coefficient (r), $s^2/\hat{\sigma}^2$	Degrees of freedom	Linear slope (b)[c] $s^2/\hat{\sigma}^2$	Probability, P^b
(1a) ^{60}Co: SS	2.013	40	1.971	*d*
: LT	1.244	49	1.232	
: WU	0.840	22	0.849	
Combined	1.441	111	1.422	*0.998*
(1b) ^{137}Cs: SS	2.890	54	2.843	*d*
: LT	1.966	4	1.973	
: WU	1.007	4	1.026	
Combined	2.709	62	2.670	>*0.9999*
(2) ^{137}Cs: SS	0.682	41	0.683	*d*
: LT	0.461	8	0.451	
Combined	0.646	49	0.645	*0.025*
(3) ^{137}Cs: AS	0.256	13	0.256	*d*
: SS	0.656	17	0.633	
: LT	0.629	10	0.613	
Combined	0.519	40	0.506	*0.004*
Random generated	1.011	1024	1.008	*0.577*

[a,b] Same as in Table I. [c] Values of $s^2/\hat{\sigma}^2$ (combined) based on randomly rearranged data are 1a: 1.016; 1b: 1.036; 2: 0.866; and 3: 0.984. [d] P's are 1a(SS): 0.9998; 1b(SS): >0.9999; 2(SS): 0.061; 3(SS + AS): 0.006.

unity in each group of data sets, the mean square of the standardized slopes within each such group would be expected to be (and is—Table II) almost identical with that of the 0,1 standardized correlation coefficients.

A large number of data sets of random deviates fit to a Gaussian distribution and truncated to integers—a very close approximation to the Poisson for the mean involved (*ca.* 529)—were generated using the IBM RANDU subroutine and employing continually changing starting values. These sets of random deviates were then analyzed using the same computer programs as were used with the detected emissions sets.

To test further for the existence of serial relationships in the data sets based on radioactive emissions, each set of count totals was shuffled randomly with respect to order (using a RANDU type subroutine) prior to recalculating each regression slope and the variance of 0,1 standardized statistics based on each randomized order.

Results

The results of the analyses are given in Tables I and II. These tables list summaries of all the runs made and known to the authors which meet the aforesaid counting conditions during the period Dec 10, 1971 through Oct 31, 1972. No runs have been omitted from the analyses other than the two previously noted which had at least one zero total in each.

Based on the 262 data sets derived from the radioactive γ emissions as well as on those from the computer generated random deviates, the nonserial χ^2 tests and the index of dispersion comparisons are, with one marginal exception, well within the 0.05 probability limit: the nonserial tests do not differentiate the distributions of randomly generated numbers or those of the radioactive counting sequences from the theoretic (random) expectation. With the exception of ref 1 and 3 as noted, this conclusion is entirely consistent with the recorded literature known to the authors.

On the other hand, using the same 262 data sets derived from the radioactive emissions, the tests of the linear correlation coefficients (r) and of the slopes of the linear regressions (b) show marked deviations from the expectation: all probabilities, when grouped as shown in Table II, lie outside $0.05P$ and three fourths are well outside $0.01P$.[12] However when the order within each set is randomized, the sets cannot be differentiated from the expectation with respect to these same statistical tests (Table II, footnote c). Similar tests of the sets based on the randomly generated numbers also cannot be differentiated from the expectation.

Table II, footnote d, also lists the probabilities associated with the variances of the regression slopes when only "steady state" counting environments are considered. Again, three of the four groups have probabilities which lie outside $0.01P$. While the value of $0.06P$ for group 2 is within the $0.05P$ limit, when these 41 sets of 1024 were subdivided into 82 half-sets ($41 \times 1\text{–}512$ and $41 \times 513\text{–}1024$), the mean square ($s^2/\hat{\sigma}^2$) of standardized half-to-half slopes was 0.553—the probability associated with this value is $0.0003P$.

The serial tests also indicate a rather pronounced effect due to lowering the source temperature and subsequent raising of the source temperature to ambient. In all four groups the effect of lowering the temperature was to lower the variance of the correlation coefficients and the slopes. However only in group 1a were sufficient runs carried out to draw this conclusion with any reasonable degree of an assurance of repeatability. Whether the effect is, in fact, an effect of temperature, *per se,* or of changing the electronic environment (by contact of the metal substrate with the liquid nitrogen in groups 1a and 1b) has not been determined.

Discussion

In experimental testing of the hypothesis of randomness by measuring emissions of radioactive decay events, the conformance of the data distributions as shown by nonserial statistical tests with the theoretic (random) expectation must be considered necessary but insufficient *proof* that the data are, in fact, random. Thus the nonconforming serial statistical tests such as are illustrated in Table II raise substantial questions relative to the general applicability of the thesis of randomness.

The existence of both significantly *high* and *low* serial tests does not in itself suggest that the nonserial tests are faulty—but it does imply that such tests fail to measure the nonrandom characteristics shown by the serial ones.

Even though the present work was originally undertaken to explore the possibility that additional examples of low or restricted values of the index of dispersion might be observed, the marked similarity of the variance tests of the 0,1 standardardized slopes of the regressions with those of the correlation coefficients as well as the agreement with theoretical expectation of the index of dispersion values indicates that variance *per se* does not contribute significantly to the observed anomalies reported herein.

Possible artifactual errors which might contribute to the nonconformance of serial statistics can be classified as faults in the experimental design or as equipment-based shortcomings such as drift.

Adventitious movement of the source with respect to the detector (during the 13.7 min of counting ^{137}Cs or 6.9 min of counting the ^{60}Co series) would cause an artifactual increase both in the nonserial and serial tests used in this paper. Also any time-phased change in detected emissions during cooling (by adventitious condensation of moisture in the path of the emissions, for example) must also result in an increase of the several statistics. Neither can cause a *decrease* in the variance of the slopes of the regressions however. Thus since the high values of the variances of the slopes (and of the correlation coefficients) have been observed during "steady state" counting of nongrounded sources only, the authors have concluded that such artifactual effects have not occurred to any significant degree.

A discussion of the effect of small angle detectors was included in ref 1 (supplemental note 1). To determine statistical properties of radioactive emissions, the use of such detectors (assuming independent decay events and random emissions) can be considered as noncasual for statistical abnormalities insofar as the detected emissions are concerned with the single exception (b) discussed below. Since all the statistics discussed in this paper depend only on detected emissions, such a conclusion also applies here.

If emissions (between decay events and detection) are pushed into or out of the solid angle detection zone deliberately or adventitiously one of two effects result.

(a) If the *number* of detected emissions is *consistently* increased (or decreased), the net effect is to enlarge (or make smaller) the solid angle detection zone. From the standpoint of the detector it is as if the source were larger (or smaller) than it actually is. This situation has, in itself, no adverse effect on any of the statistical analyses employed in this paper.

(b) If the number of detected emissions is *inconsistently* increased or decreased or both within the overall elapsed time of each 1024 data set, the observed counts would vary more than would be expected on the basis of random emissions. Such an effect would overall be manifested by an *increase* both in the index of dispersion (s^2/m) and in the $s^2/\hat{\sigma}^2$ of the regression slopes. Since the $s^2/\hat{\sigma}^2$ statistic is *less* than is expected in groups 2 and 3, the likelihood that such an artifact would be causal of the overall phenomenon is very low.

Within the subject of potential equipment-based artifacts, only the inherent drift of the detector–PMT appears to be of such a magnitude as to influence the slope variances (or those of the correlation coefficients) to any extent at all. The rest of the equipment (preamplifier to printout) was independently shown to be artifact-free—at least during the course of the specific equipment checks cited; further, the method of temperature stabilization of the detector–PMT and the use of the highly stabilized high-voltage supply appears to preclude any possible artifact from such causes.

For purposes of discussion this inherent drift capability can be separated into two components: long-term drift and short-term drift in which the term short term drift refers to shift of the PMT output within the time of accumulation of an individual data set. In each case the effect of drift is to change the count rate and the number of counts within the data set artifactually.

According to the manufacturer (*cf.* note 8), the short-term drift is rarely if ever greater in magnitude than the long-term drift, and thus the showing below that the long-term drift (as shown by mean shift) is of a very low order

TABLE III: Compressed data for 23 Data Sets[a]

Group	Run	Mean + (deviations from mean)[b]	Intercept and slope[c] + (deviations from linear trend)[b]
1a	Z20	519.5 + (−3.1, −2.5, −2.5, −0.8, +2.0, +2.7, +1.4, +2.8)	515.2 + 0.94n + (+0.2, −0.2, −1.1, −0.3, +1.5, +1.3, −1.0, −0.5)
	Z97	486.5 + (+2.2, −0.3, +1.3, −0.5, +1.2, −0.9, −0.7, −2.3)	488.6 − 0.45n + (+0.6, −1.4, +0.6, −0.7, +1.4, −0.2, +0.4, −0.7)
	X01	714.3 + (−1.8, −1.8, +0.7, −0.4, +0.3, +0.7, +2.6, −0.2)	712.5 + 0.41n + (−0.4, −0.8, +1.4, −0.2, +0.1, +0.1, +1.6, −1.6)
	V04	328.5 + (−0.4, −2.4, −1.1, −0.3, +1.3, +0.9, +1.0, +1.0)	326.7 + 0.40n + (+1.0, −1.4, −0.5, −0.1, +1.1, +0.3, +0.0, −0.5)
1b	Q01	305.5 + (+2.5, +0.9, +1.9, −1.1, −0.9, −1.1, −0.0, −2.1)	308.0 − 0.55 n + (+0.6, −0.5, +1.0, −1.4, −0.6, −0.2, +1.3, −0.2)
	Q11	371.8 + (−1.9, −1.8, −1.7, +1.3, +0.7, +1.7, +0.8, +0.7)	369.6 + 0.49n + (−0.2, −0.6, −1.0, +1.6, +0.5, +1.0, −0.4, −1.0)
	Q18	408.0 + (−3.9, −2.2, −0.8, +1.5, +0.9, +0.6, +1.1, +2.8)	404.4 + 0.80n + (−1.1, −0.2, +0.4, +1.9, +0.5, −0.6, −0.9, +0.0)
	Q19	360.0 + (+3.0, +0.8, +0.0, −0.8, −0.6, −0.7, −1.5, −0.3)	362.0 − 0.44n + (+1.5, −0.3, −0.6, −1.0, −0.4, −0.0, −0.4, +1.2)
	VCB	384.7 + (+1.4, +2.5, +1.9, +1.7, −0.4, −2.0, −2.3, −2.8)	388.3 − 0.80n + (−1.4, +0.5, +0.7, +1.3, +0.0, −0.8, −0.3, −0.0)
	QQ3	542.2 + (+3.3, −0.3, +1.0, +0.0, −1.6, −0.9, −1.1, −0.4)	544.2 − 0.44n + (+1.7, −1.4, +0.4, −0.2, −1.4, −0.2, +0.0, +1.1)
3AS	Q61	550.4 + (+0.1, −0.9, +0.1, −0.8, +1.1, −0.2, +0.0, +0.5)	550.0 + 0.10n + (+0.5, −0.6, +0.2, −0.8, +1.1, −0.4, −0.2, +0.2)
	Q62	551.2 + (−0.0, −1.3, +0.8, +1.8, +0.6, −0.2, +0.6, −2.2)	551.8 − 0.12n + (−0.5, −1.6, +0.6, +1.7, +0.7, −0.0, +0.9, −1.8)
	Q63	551.0 + (−0.7, +0.2, +1.9, −0.2, −1.0, −0.3, +0.4, −0.3)	551.2 − 0.04n + (−0.8, +0.1, +1.8, −0.2, −1.0, −0.2, +0.5, −0.2)
	Q64	551.2 + (+0.6, −1.5, −0.4, +0.4, +1.0, −1.5, +1.1, +0.3)	550.7 + 0.10n + (+0.9, −1.3, −0.2, +0.4, +1.0, −1.6, +0.9, −0.1)
	Q65	550.6 + (−0.2, +0.2, −1.3, +2.1, −0.9, −0.4, +0.1, +0.4)	550.4 + 0.04n + (−0.1, +0.3, −1.2, +2.2, −0.9, −0.5, −0.0, +0.2)
	Q66	549.2 + (−1.2, +0.7, +0.1, +0.3, +0.3, −0.1, −0.3, +0.3)	548.9 + 0.06n + (−0.9, +0.8, +0.1, +0.3, +0.3, −0.2, −0.5, +0.1)
	Q67	550.3 + (+0.9, −0.4, +0.3, −0.7, −0.6, +0.3, +0.4, −0.1)	550.5 − 0.04n + (+0.8, −0.5, +0.2, −0.7, −0.6, +0.3, +0.5, −0.0)
	Q68	549.9 + (+0.5, −0.4, −1.5, −0.6, +0.1, +0.5, +1.3, +0.0)	549.3 + 0.14n + (+1.0, −0.0, −1.3, −0.6, +0.1, +0.3, +1.0, −0.4)
	Q69	550.1 + (−0.0, +1.2, −0.8, −1.0, +0.5, +0.2, −0.4, +0.4)	550.1 − 0.01n + (−0.0, +1.2, −0.8, −1.0, +0.5, +0.2, −0.4, +0.4)
	Q70	549.8 + (+0.6, +0.2, −1.0, +0.8, −0.0, −0.3, −0.3, −0.0)	550.1 − 0.07n + (+0.3, +0.1, −1.1, +0.8, +0.0, −0.2, −0.2, +0.2)
	Q71	549.5 + (−0.7, +0.2, +1.1, −1.6, −0.5, +0.6, +1.5, −0.5)	549.1 + 0.08n + (−0.4, +0.4, +1.2, −1.6, −0.5, +0.4, +1.3, −0.8)
	Q72	549.6 + (+1.3, −1.5, −0.4, −0.4, +0.3, +0.7, +0.8, −0.8)	549.5 + 0.01n + (+1.3, −1.5, −0.4, −0.4, +0.3, +0.6, +0.8, −0.8)
	Q73	549.9 + (+0.5, −0.1, +0.6, −0.6, −0.6, +0.5, −1.0, +0.6)	550.1 − 0.06n + (+0.3, −0.2, +0.6, −0.6, −0.6, +0.6, −0.8, +0.8)

[a] Compressed data calculated as $2\sqrt{\text{sum of 128 count totals}}$. [b] The entries in the line for Q01, for example, correspond to observed $2\sqrt{\text{count}}$ values of 305.5 + 2.5 = 308.0, 305.5 + 0.9 = 306.4, 305.5 + 1.9 = 307.4 (and so on) on the left and to 308.0 − 1(0.55) + 0.6 = 308.0, 308.0 − 2(0.55) − 0.5 = 306.4, 308.0 − 3(0.55) + 1.0 = 307.4 (and so on) on the right. (Occasional discrepancies of ±0.1 or ±0.2 in other lines are due to rounding. [c] Since $\Sigma(n - \bar{n})^2 = 42$, the rms coefficient of n for Poisson randomness should be about 0.15.

is presumptive evidence that the short-term drift is also of a low order.

Long term drift is often stated in terms of drift per day or drift per 8 hr. Although the authors have drawn, in this paper, no detailed statistical conclusions based on mean shifts over long periods (*e.g.*, 8 hr), such shifts have indeed been examined. Thus when six series encompassing 70 data sets (35 sets in daily sequences of 10, 12, and 13 series based on grounded aluminum substrates—group 2—and a similar 35 sets based on nongrounded substrates —group 1b) were examined, the largest apparent 8-hr shift amounted to only *ca.* 0.6% or, if linear over this 8-hr period, only *ca.* 0.02% per 13.7 min counting period. In these same six series, essentially no overall mean shift was observed in three of them. The largest set-to-set shift within each of these six series was under 0.5%—just in excess of a not too unexpected 3σ, assuming the Poisson applies. For a mean of 400 counts, the *maximal* long term mean shift, if linear, thus amounts to only ±0.1 count over each 1024 count set, a change which is, overall, insignificant with respect to increased slope variance.

In these considerations it is necessary to differentiate mean shift from drift of the PMT since (as mentioned in the Experimental Section) drift is not linearly related to mean shift (the drift affects photopeak *position* within the particular SCA window settings employed). The observed long-term mean shift (and even the short-term ones) may also be due to factors other than PMT drift *per se*, such as small, nonrecorded adjustments in the source–detector geometry, excessive (but not expected) temperature changes in the ambient, and/or even a randomness type factor in decay itself. (Note: The authors recognize that this argument relative to mean shift based on radioactive counting is open to some question since the results of this paper raise substantial questions relative to randomness of detected emissions. However, no better evidence of PMT behavior is available since the γ's required to pulse the detector-PMT cannot readily be generated in any other way.)

The potential effect of short-term drift on the output of the detector-PMT (and the resultant effect on the detected counts) was also discussed in the Experimental Section. The *maximal* expected drift was shown to be of only small moment ($< 0.5\sigma$) under the conditions cited, a finding which is entirely consistent with the observations of mean shift cited.

Certainly for high variance values of the regression slopes, the small (but not negligible) potential drift of the detector-PMT might be responsible for a minor portion of the observed variances but in no manner can it be causative in the two groups exhibiting *low* slope variances. Actually, if such drift is indeed a factor in any appreciable degree in the high variance examples (groups 1a and 1b), the low variances of groups 2 and 3 are likely to be *understated* (*i.e.*, the listed results are likely to be higher than they would have been in the absence of drift).

In the two cases of nongrounded substrates (groups 1a and 1b), there existed ten data sets (out of the 173 listed ones) in which the values of the standardized slopes of the linear regressions were above 2.56 (*i.e.*, the 1% level of significance) whereas only two are expected. These ten contribute greatly to the excessively high levels of the variances since the variance is proportional to the sum of squares of such standardized values. Of these ten, none was individually unusual in the nonserial tests; the overall index of dispersion for these ten was 0.994 which is indistinguishable from the overall value of all 173 runs.

One of the referees suggested that it might be instructive to list the values of these ten series in compressed form as $2\sqrt{\text{sum of 128 counts}}$ since, for Poisson distributions, $2\sqrt{\text{sum}}$ − mean has very nearly unit variance. Thus the sizes of changes from the expectation on the approximately minute-to-minute time scale can be appreciated. Table III lists the deviations of such double-square-roots-from-run-means for each of these ten series and the deviations from the linear trend in each case as well. (In the table note that run Q11 may be a step change.) Also included in Table III for comparison purposes are similar analyses of the 13 data sets that comprise the aqueous solution runs of group 3; these latter represent a series in which the variances of the slopes are extremely low.

It is of interest that excessive mean shifts have previously been reported during counting of β-emissions from thin layers of carbon-14 containing material when the source holder was nongrounded; the authors apparently did not question the statistics of counting, however.[13] A significant statistical effect associated with nongrounded substrates was, however, observed during counting of the carbon-14 submonolayers previously cited.[1]

While the meaning of high (or low) variance of the regression slopes can be interpreted as higher than expected (or lower than expected) shift of the mean, a similar analysis of the high and more particularly of the low variances of the linear correlation coefficient does not appear to have such a straightforward physical explanation. At this time all the authors wish to conclude with respect to these serial tests is that they show that the time-related order of the data is not consistent with random expectation. Other serial tests might also have been used, permitting increased (or decreased) definition of the nonrandom phenomenon herein described.

Conclusion

γ Emissions detected following radioactive decay of cobalt-60 and cesium-137 nuclei deviate significantly from the theoretic (random) expectation as shown by tests of serial statistics even though nonserial tests fail to differentiate the same data distributions from the (random) expectation. Insofar as emissions and decay events are appropriately interrelated, the evidence is inconsistent with the thesis of decay independence.

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Supplementary Material Available. Supplemental notes and material will appear following these pages in the microfilm edition of this volume of the journal. Photocopies of the supplementary material from this paper only or microfiche (105 × 148 mm, 20× reduction, negatives) containing all of the supplementary material for the papers in this issue may be obtained from the Journals Department, American Chemical Society, 1155 16th St., N.W., Washington, D. C. 20036. Remit check or money order for $3.00 for photocopy or $2.00 for microfiche, referring to code number JPC-73-3114.

References and Notes

(1) J. L. Anderson, *J. Phys. Chem.*, **76**, 3603 (1972).
(2) *Cf.* E. Rutherford and H. Geiger, *Phil. Mag.*, **20**, 698 (1910); L. F. Curtiss, *J. Res. Nat. Bur. Stand. A*, **8**, 339 (1932). See also, A. P. Kovarik, *Phys. Rev.*, **13**, 272 (1919), which describes β emissions.
(3) The "exception": W. Kutzner, *Z. Phys.*, **21**, 281 (1924); **44**, 655 (1927); L. F. Curtiss, *J. Res. Nat. Bur. Stand. A*, **4**, 595 (1930).
(4) J. Berkson, "Examination of Randomness of Alpha Particle Emissions," in "Research Papers in Statistics," F. N. David, Ed., Wiley, New York, N. Y., 1966, pp 37–54.
(5) See paragraph at end of paper regarding supplementary material.
(6) Canberra Industries, Inc., Model 3015 High-Voltage Power Supply. This unit is designed to have less than 0.05% voltage variation over an 8-hr period with a 3° temperature variation and less than 0.01% output voltage change for a 10% change in nominal line voltage. Experimental checks of the power supply unit did not indicate any adverse deviation from the design specifications.

(7) Model numbers were: multichannel analyzer, ND Model 2200; detector–PMT, CI Model 802-1; preamplifier, CI Model 805; amplifier, CI Model 1417B; and timing SCA, CI Model 1437.

(8) As shown by actual data on which the window used for counting the plastic encapsulated CsCl in aqueous solution was established, these settings were determined by observing the counts per channel (averaged over at least 10 channels) at a series of baseline (0 to 10.0) settings with the window (also 0 to 10.0) set at 0.10 and with the amplifier coarse gain held at 8 and fine gain at 0. The values shown are baseline settings followed by the counts in parentheses: 3.6(16); 3.7(11); 3.8(10); 3.9(8); 4.0(12); 4.1(22); 4.2(71); 4.3(190); 4.4(352); 4.5(448); 4.6(367); 4.7(221); 4.8(84); 4.9(22); 5.0(5); 5.1(1); and 5.2(1). In this case a baseline of 3.9 was selected with the window at 1.30. Note: The summation of counts for the baseline of 3.9 and a window of 1.3 (assuming linearity of window) is 1803. The actual 13 × 1024 series (hereinafter detailed) had a mean of slightly less than 600 since the source-to-detector subtended solid angle was smaller during the actual counting series cited. Any adventitious variation in detector–PMT output (which causes a shift primarily in the *position* of the photopeak) is thus seen to be unlikely to cause a significant shift in counts per channel. While the manufacturer (CI) has stated in a personal communication that as much as a 1% shift in the PMT output over a 1-hr period is relatively rare and unexpected, even a 1.0% net overall shift in 13.7 min (the duration of ^{137}Cs counting sequences) would have caused a maximal shift of *ca.* 1 count (for the means of 600). As is discussed later in this paper, such an unusual and artifactual shift is relatively insignificant ($<0.5\sigma$) for the serial tests used.

(9) The equations used to calculate these statistics are listed below where the symbols are defined as: a_i denotes the counts per unit time, m denotes the mean of the data set ($= (1/n)\Sigma a_i$), t_i denotes the time expressed as a digit (1 to 1024) at which the a_i count total was observed, $\bar{t}$ denotes the mean of the times ($= 512.5$), and n denotes the value 1024. For the nonserial χ^2 tests, o_i are the observed frequencies and f the theoretic frequencies with a minimum of at least 5 components in each theoretic cell, p being the number of such cells.

χ^2

$$\chi^2 = \sum_{i=1}^{p}(o_i - f)^2/f$$

Index of dispersion

$$s^2/m = \{1/(n-1)\}\sum_{i=1}^{n}(a_i - m)^2/m$$

Linear correlation coefficient (r)

$$r = \left\{\sum_{i=1}^{n}(t_i - \bar{t})(a_i - m)\right\} / \left\{\sum_{i=1}^{n}(t - \bar{t})^2 \cdot \sum_{i=1}^{n}(a_i - m)^2\right\}^{1/2}$$

Slope of the regression (b)

$$b = \sum_{i=1}^{n}(a_i - m)(t_i - \bar{t}) / \sum_{i=1}^{n}(t_i - \bar{t})^2$$

The relationship of r to b may be expressed

$$r = b\left\{\sum(t_i - \bar{t})^2/(n-1)s^2\right\}^{1/2} = \text{constant}\cdot(b/m^{1/2})/(s^2/m)^{1/2}$$

(10) J. S. Bendat and A. G. Pierson in "Random Data; Analysis and Measurement Procedures," Wiley, New York, N. Y., 1971, pp 126–128.

(11) N. R. Draper and H. Smith in "Applied Regression Analysis," Wiley, New York, N. Y., 1966, p 18. Since for Poisson distributions (and that is, essentially, the hypothesis being tested) the sample mean (m) is the best estimate of the population variance, the expression for the standard deviation of the theoretic slope reduces to the form shown.

(12) It may be worthy of note that when one of the data sets of the 111 group 1a series (*i.e.*, Z20—see Table III) is removed, the $s^2/\hat{\sigma}^2$ falls to *ca.* 1.10 and the whole remaining group cannot be differentiated from the (random) expectation. However there is no reason known to the authors to suspect that any artifactual factor caused the excessively high value in Z20 particularly since the run made 30 min earlier appeared completely normal with respect to the tests employed. The significant deviation from the expectation of the autocorrelation coefficients of this group of data sets (supplemental notes) is *not* appreciably affected by the removal of Z20. Further, no such similar removal of a single run has any particular effect in group 1b.

(13) H. J. Perkins and M. D. MacDonald, *Science*, **138**, 1259 (1962).